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Dynamic Subcriticality Measurements Using the $^{252}$Cf-Source-Driven Noise Analysis Method


Oak Ridge National Laboratory, Instrumentation and Controls Division
Oak Ridge, Tennessee 37831-6305

and

Y. Hachiya

Power Reactor and Nuclear Fuel Development Corporation, Tokyo, Japan

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Abstract—Dynamic measurements of the subcritical neutron multiplication factor $k_{\text{eff}}$ using the $^{252}$Cf-source-driven neutron noise analysis method were performed for an unreflected 25.1-cm-i.d. cylindrical tank containing aqueous uranyl nitrate as the solution height was changed at rates of 1 to 23 cm/min, with corresponding changes in $k_{\text{eff}}$ from $4 \times 10^{-4}$ to 0.01/s.

These experiments, which were the first test of the method to measure $k_{\text{eff}}$ while it is changing, showed the following:

1. This method has the capability to measure subcriticality for a multiplying system to a $k_{\text{eff}}$ as low as 0.30.

2. Experimental $k_{\text{eff}}$ values can be obtained from the ratio of spectral densities with as little as 6 s of data accumulation and a small fraction of a second analysis time while the solution tank is drained from a height of 29.3 to 0.5 cm in ~60 s, with corresponding changes in $k_{\text{eff}}$ from 0.95 to 0.30.

3. The measured $k_{\text{eff}}$ values obtained do not depend on the speed at which the solution height is changed or whether it is filling or draining.

4. The results of the dynamic measurements agreed with the static measurements.

5. Where static measurements were practical (limited to $k_{\text{eff}}$ down to ~0.5 by detection efficiency) with $^4$He proportional counters sensitive to leakage neutrons only, the results agreed with those from measurements with scintillation detectors sensitive to gamma rays and neutrons escaping from the system.

6. As in previous experiments, the ratios of spectral densities at low frequency were used successfully to obtain $k_{\text{eff}}$ values using a modified point kinetics interpretation of the data.

7. The neutron multiplication factors from independent measurements using the break frequency noise analysis method agree with the values of $k_{\text{eff}}$ from the measured ratios of spectral densities down to $k_{\text{eff}}$ values of 0.65.

8. The effectiveness of this method for systems where conditions are changing probably exceeds the dynamic requirements of most nuclear fuel plant processing applications.

9. Calculated $k_{\text{eff}}$ values using the KENO Monte Carlo code and Hansen-Roach cross sections compare well with the experimental values.
compressed through four 0.63-cm-diam threaded steel tie rods, equally spaced around the circumference of the vessel, joining the base and a stainless steel upper flange. A 0.63-cm-thick, 32- x 32-cm square acrylic plate on top of the upper flange provided access for the source and minimized evaporation of the solution.

The height of solution above the base plate was determined by an ultrasonic device with a transducer mounted on the acrylic cover of the vessel over a circular opening. Every 0.125 s, this device sampled and stored in the computer the solution height to within ±0.2 mm. Figure 1 is a photograph of the experimental vessel showing the mounted transducer and the 252Cf source ionization chamber at the top of the solution.

The vessel was mounted on a 122- x 122-cm square aluminum table 76 cm above a steel grating covered with a stainless steel sheet. This grating was 3.6 m above the concrete floor of the experiment cell. The cell in which the equipment was assembled was ~9.1 x 12.2 x 9.1 m high with thick concrete walls and roof. The experiment vessel was located 4.4 m from the 9.1-m south side of the cell, 3.0 m from the 12.2-m east side, and ~1.85 m from a 2.9-m-diam empty steel tank (2.5 cm thick) also present in the cell. The steel tank was located in the cell such that its axis was at the 9.1-m south side of the cell and 6.2 m 12.2-m east side.

The aqueous uranyl nitrate contained a solution density of 1.643 g/cm³, with content <0.1 wt% HNO₃. Uranium isotopes 235U = 1.02, 235U = 93.2, 236U = 0.41, 5.37 wt% and was free of significant impurities.

The 252Cf was electroplated on one parallel-plate ionization chamber, and the spontaneous fission rate was 60 000/s (~0.1 µg 252Cf). The ionization chamber was mounted at the center of a 2.54-cm-o.d., 1.27-cm-i.d. Lexan tube with the 252Cf source was sealed from the solution with steel and epoxy. The Lexan tubing and the signal protruded out the top of the solution through a hole in the lid of the experimental vessel. The source could be located anywhere along the axis of the cylindrical experimental vessel. In most of the experiments, the source was located at the bottom of the experimental vessel. In practical applications, the bottom of the tank was a convenient location for the source. For measurements at various solution heights, the source was located on the axis at the vertical midsection of the solution, but in some measurements, its location on the axis was varied.

Two types of detectors were used in the experiments: commercially available 3He proportional counters primarily for static measurements and an assembled composite 6Li-glass organic scintillator for both static and dynamic measurements. Proportional counters (5.1-cm-diam, 38-cm Reuter-Stokes model RS-P4-1641-101) with their axes parallel to the axis of the vessel adjacent to the outer surface of the east and west sides, 180 deg apart azimuthally, detected leakage neutrons. One end of each detector was at the bottom of the tank. Thus, as the solution height changed, the active length of the source was essentially the height of the solution.

Two such detectors on the east and west sides of the tank were present for the dynamic measurements.

The scintillation detectors were 6Li-glass organic scintillators sensitive to fast neutrons and gamma rays. Each scintillator consisted of a 15.2- x 15.2- x 0.5-cm-thick organic scintillator optically coupled to a 15.2- x 15.2- x 3-cm-thick 6Li-glass scintillator sensitive to fast neutrons and gamma rays. The 6Li-glass scintillator was installed in the experimental vessel with the organic scintillator optically coupled to the 6Li-glass so that the organic was almost neutrons. Each detection channel consisted of two adjacent 15.2- x 15.2- x 10.7-cm-thick 6Li-glass organic scintillators, each mounted in an aluminum box (10 cm wall) that formed a detection channel.
with time is compared with the results of measurements where the solution height was fixed. For these comparisons, the neutron multiplication factor obtained for the average height in the dynamic measurements was compared with static measurements fixed at the average height. The results are also compared with those from break frequency noise analysis measurements.

STATIC MEASUREMENTS

Static measurements were performed to obtain reference measurements to compare with the results of dynamic measurements. These measurements were performed as a function of solution height with the $^{252}$Cf source located on the axis at the center of the solution. In the static measurements, the detectors were $^3$He proportional counters 180 deg apart, adjacent to the outer surface of the vessel with their axes parallel to the axis of the experimental vessel and $^6$Li-glass scintillators located as shown in Fig. 2. The length the $^3$He detectors was more than the height of the solution.

Typical ratios of spectral densities as a function of frequency are shown in Fig. 3. These ratios were examined visually to determine the range of frequencies over which the ratio of spectral densities was constant. This range of frequencies is generally greater for more subcritical systems because the frequency response of the experimental system $H_f$ has significant amplitude at higher frequency. A range of frequencies over which the ratio was arithmetically averaged was then selected to eliminate high-frequency points for which the statistical uncertainty was large and would lead to high or low values of the ratio that would distort the average value of the ratio. Thus, both of these criteria, constant ratio and s
**TABLE I**

Ratios of Spectral Densities $G_{13}G_{11}/G_{13}G_{13}$ at Low Frequency for Various Solution Heights from Static Measurements with Central Source and $^3$He Proportional Counters Adjacent to the Outer Surface of the Experimental Vessel

<table>
<thead>
<tr>
<th>Solution Height (cm)</th>
<th>Bandwidth of Measurement (kHz)</th>
<th>Number of Data Samples (10^3)</th>
<th>Ratios of Spectral Densities* (10^{-2})</th>
<th>Upper Limit of Frequency Range for Ratio (kHz)</th>
<th>Neutron Multiplication Factor</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Measurement</td>
<td>Calculation</td>
</tr>
<tr>
<td>30.5</td>
<td>20</td>
<td>80</td>
<td>$100 \pm 1$</td>
<td>3</td>
<td>0.963</td>
</tr>
<tr>
<td>30.5</td>
<td>40</td>
<td>80</td>
<td>$96 \pm 1$</td>
<td>3</td>
<td>0.964</td>
</tr>
<tr>
<td>27.9</td>
<td>40</td>
<td>80</td>
<td>$139 \pm 1$</td>
<td>3</td>
<td>0.945</td>
</tr>
<tr>
<td>25.4</td>
<td>50</td>
<td>240</td>
<td>$190 \pm 1$</td>
<td>3</td>
<td>0.919</td>
</tr>
<tr>
<td>22.9</td>
<td>20</td>
<td>40</td>
<td>$248 \pm 3$</td>
<td>5</td>
<td>0.884</td>
</tr>
<tr>
<td>20.3</td>
<td>100</td>
<td>160</td>
<td>$304 \pm 3$</td>
<td>8</td>
<td>0.843</td>
</tr>
<tr>
<td>20.3</td>
<td>100</td>
<td>40</td>
<td>$303 \pm 5$</td>
<td>7</td>
<td>0.843</td>
</tr>
<tr>
<td>15.2</td>
<td>50</td>
<td>840</td>
<td>$465 \pm 3$</td>
<td>5</td>
<td>0.704</td>
</tr>
<tr>
<td>10.2</td>
<td>100</td>
<td>9360</td>
<td>$525^a$</td>
<td>0.4^a</td>
<td>0.570</td>
</tr>
</tbody>
</table>

*Uncertainties are one standard deviation of the mean.

**TABLE II**

Ratios of Spectral Densities at Low Frequency and Neutron Multiplication Factors from Static Measurements with the Source at the Bottom of the Solution

<table>
<thead>
<tr>
<th>Solution Height (cm)</th>
<th>Ratio of Spectral Densities*</th>
<th>Neutron Multiplication Factors, $k_{eff}$</th>
<th>Number of Data Blocks (10^3)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^3$He Detectors (10^{-4})</td>
<td>$^3$He Detectors Scintillators</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Scintillators (10^{-4})</td>
<td>$^3$He Detectors Scintillators</td>
<td></td>
</tr>
<tr>
<td>29.2</td>
<td>$453 \pm 12$ (5)</td>
<td>$436 \pm 5$ (2)</td>
<td>$0.952 \pm 0.002$</td>
</tr>
<tr>
<td>25.8</td>
<td>$767 \pm 40$ (5)</td>
<td>$774 \pm 20$ (2)</td>
<td>$0.924 \pm 0.005$</td>
</tr>
<tr>
<td>20.3</td>
<td>$1590 \pm 20$ (5)</td>
<td>$1616 \pm 10$ (6)</td>
<td>$0.853 \pm 0.005$</td>
</tr>
<tr>
<td>15.3</td>
<td>$2896 \pm 90$ (5)</td>
<td>$2843 \pm 10$ (10)</td>
<td>$0.732 \pm 0.015$</td>
</tr>
<tr>
<td>10.1</td>
<td>$4508 \pm 90$ (5)</td>
<td>$4477 \pm 60$ (10)</td>
<td>$0.535 \pm 0.030$</td>
</tr>
<tr>
<td>9.8</td>
<td>---</td>
<td>$4723 \pm 60$ (10)</td>
<td>---</td>
</tr>
<tr>
<td>9.0</td>
<td>---</td>
<td>$4960 \pm 60$ (10)</td>
<td>---</td>
</tr>
<tr>
<td>7.5</td>
<td>---</td>
<td>$5293 \pm 210$ (10)</td>
<td>---</td>
</tr>
<tr>
<td>6.5</td>
<td>---</td>
<td>$5650 \pm 60$ (10)</td>
<td>---</td>
</tr>
<tr>
<td>5.7</td>
<td>---</td>
<td>$5943 \pm 360$ (10)</td>
<td>---</td>
</tr>
</tbody>
</table>

*aValues in parentheses are the upper limit of the frequency range in kilohertz over which the ratio was averaged. The precision given is one standard deviation of the mean.

*bUncertainties are statistical precision of the ratio of spectral densities and uncertainties in the parameters of Appendix B. The largest contribution to the uncertainty was from $I_s/I$ (e.g., 80% of the uncertainty for a solution height 6.5 cm). All tables in this paper other than Tables II and III have uncertainties from the precision of the ratio of spectral densities only.
Fig. 5. Ratios of spectral densities as a function of frequency as the experimental vessel was filled at a rate of 1 cm/s (continued on following pages).
in the measured neutron multiplication factor varies from 0.01 at the lowest $k_{\text{eff}} (-0.31)$ to $<0.001$ at higher $k_{\text{eff}}$ values (0.93).

The first three entries of Table III are for the same solution height. Intercomparison of these measurements (102 s of data accumulation time) shows the ratios and the resulting neutron multipliers are reproducible even at $k_{\text{eff}}$ values as
The experimental vessel was drained at three different rates with the source on the bottom of the tank. The fastest rate was such that the height varied from 29.5 to -6 cm in 60 s. This limit on the draining rate resulted from the 1.27 cm diam opening in the bottom of the tank. Other draining rates were such that the height of solution changed 3 and 5 cm/min.

For a draining rate corresponding to a change of solution height of -3 cm/min, the data accumulation time before the Fourier processor uploaded the data to the VAX computer for interpretation was 13 s while that for more rapid draining of the tank (5 and 23 cm/min) was 6.4 s. Thus, these data have a larger statistical error than data for filling the experimental vessel because the data accumulation time is a factor of 8 to 16 shorter. The ratios of spectral densities at low frequency for their draining rates are plotted as a function of solution height in Fig. 6. The \( k_{s} \) values obtained from these measurements are in agreement with those from filling the vessel (Fig. 7).

A draining rate that corresponds to a change in solution height of 23 cm/min, the data accumulation time before the Fourier processor uploaded the data to the VAX computer for interpretation was 13 s while that for more rapid draining of the tank (5 and 23 cm/min) was 6.4 s. Thus, these data have a larger statistical error than data for filling the experimental vessel because the data accumulation time is a factor of 8 to 16 shorter. The ratios of spectral densities at low frequency for their draining rates are plotted as a function of solution height in Fig. 6. The \( k_{s} \) values obtained from these measurements are in agreement with those from filling the vessel (Fig. 7).

For a draining rate that corresponds to a change in solution height of 23 cm/min (\( \Delta k_{s} / s = 0.01 \)), the solution was drained continuously from 28.9 to 5.7 cm.

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The draining rate was limited by the size of the bottom of the tank. The results of the data interpretation are given in Table IV. The precision of the measured ratio of spectral densities is such that the precision in the \( k_{s} \) value is down to \( 0.01 \) (solution height of 12 cm) for the collection time of 6.4 s and the draining rate cm/min. For the last four entries of Table IV, the solution height was fixed at 5.72 cm and the average value obtained is 0.30. The ratios of spectral densities and neutron multiplication factors are plotted as a function of solution height in Figs. 5 and 6, which are compared with the other dynamic measurements.

The rate of change of the neutron multiplication factor in this experiment was 0.01 in \( k_{s} \) per sec. The neutron multiplication factor from all dynamic measurements (filling at a rate of 1 cm/min and draining at rates of 3, 5, and 23 cm/min) and the static measurements are in agreement. Thus, over the range of filling and draining rates investigated, the \( k_{s} \) measured do not depend on the rate of change or whether the solution height is increased, decreased, or stationary. The dynamic capability of the method may be more than that required for most nuclear processing or reprocessing plant applications.

### Solution Perturbed by Bubbles

After the experimental vessel was filled to 29 cm, exploratory experiments were performed with air bubbles introduced into the bottom of the tank by letting the pump running with no fuel solution in the tank. As a result, air was continuously added to the bottom of the tank through a 1.3-cm-diam hole at a radius of 7.6 cm. Air bubbles moved vertically through the solution, displacing fuel solution from the interior to the upper surface of the solution. This displacement of fuel solution from the interior to the upper surface decreased the activity. For this measurement, the source was on axis at the bottom of the tank.

These exploratory experiments were the first experiments in which a solution was perturbed during type of measurement (a perturbed solution tank be typical of some tanks in in-plant applications). These exploratory measurements, no quantitative characterization of the bubbling was performed on the estimate that the volume of air continuously introduced into the solution was \( 500 \) cm\(^3\)/min. Data accumulation rates were such that 102 s of data were accumulated by the Fourier analyzer before the data file was uploaded to the VAX computer.

The ratios of spectral densities at low frequency with and without bubbles are given in Table V all with the \( k_{s} \) values obtained using the same parameters with and without bubbles. The assumption of the same parameters can be used is probably valid since the air introduction was not a large perturbation.
associated with the bubbles is a small decrease (~1.6 x 10^-3).

BREAK FREQUENCY NOISE ANALYSIS METHOD

The break frequency noise analysis (BFNA) method provides another technique for measuring the subcritical neutron multiplication factor and thus can be compared to the results of the californium-source-driven noise analysis method. It is not completely independent of the californium-source-driven noise method since the BFNA method uses a reference reactivity of $k_{eff} = 0.95$ measured by the californium method.

The various spectral densities as a function of frequency can be least-squares fitted to obtain the fundamental mode break frequency $f_b$. The reactivity at a given subcritical state is related to the fundamental mode break frequency at that subcritical state $f_b$ (or to the prompt neutron decay constant $\alpha = 2\pi f_b$) and to the fundamental mode break frequency at delayed criticality $f_{bdC}$ (or prompt neutron decay constant $\alpha_{DC} = \beta/L$) as follows:

$$\alpha = \frac{f_b}{f_{bdC}} = \frac{1 - k_{eff}}{k_{eff} \beta} + 1.$$

Equation (1) must be corrected for the changes the neutron lifetime and in the effective delayed neutron fraction $f_{Dc}$ from the delayed critical state to the subcritical state of interest as follows:

$$\frac{1 - k_{eff}}{k_{eff} \beta} + 1 = \frac{f_b}{f_{bdC}} \frac{l_{DC}}{l_{DC} \beta}.$$

For the reactivity changes in these experiments corrections for neutron lifetime and effective delayed neutron fraction changes were made. The ratios of prompt neutron lifetimes and the ratios of effective delayed neutron fractions were obtained from calculations of these quantities as a function of solution height, which used fixed source forward fluxes and adjoint fluxes from 5x transport the calculations.

The break frequency for each solution height was obtained by fitting the various CPSDs and APSDs as functions of frequency $\omega$ to functional forms $H(\omega)$

<table>
<thead>
<tr>
<th>Solution Height (cm)</th>
<th>Fundamental Mode Break Frequency ($s^{-1}$)</th>
<th>Percentage Change in Reactivity from BFNA, $k_{eff}$ (5)</th>
<th>Ratio of Spectral Densities$^a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>29.5</td>
<td>1369 ± 7$^b$</td>
<td>0</td>
<td>6.13$^c$</td>
</tr>
<tr>
<td>29.5</td>
<td>1385 ± 7</td>
<td>0</td>
<td>6.13$^c$</td>
</tr>
<tr>
<td>27.6</td>
<td>1375 ± 7</td>
<td>0</td>
<td>6.13$^c$</td>
</tr>
<tr>
<td>27.6</td>
<td>1781 ± 13</td>
<td>1.1</td>
<td>8.6</td>
</tr>
<tr>
<td>25.7</td>
<td>2320 ± 9</td>
<td>2.4</td>
<td>11.3</td>
</tr>
<tr>
<td>23.8</td>
<td>2889 ± 12</td>
<td>3.7</td>
<td>14.7</td>
</tr>
<tr>
<td>22.0</td>
<td>3563 ± 14</td>
<td>5.3</td>
<td>18.6</td>
</tr>
<tr>
<td>20.2</td>
<td>4327 ± 8</td>
<td>6.5</td>
<td>23.4</td>
</tr>
<tr>
<td>18.4</td>
<td>5175 ± 20</td>
<td>9.7</td>
<td>28.8</td>
</tr>
<tr>
<td>16.5</td>
<td>6218 ± 13</td>
<td>13.5</td>
<td>36.9</td>
</tr>
<tr>
<td>14.7</td>
<td>7468 ± 18</td>
<td>18.5</td>
<td>47.9</td>
</tr>
<tr>
<td>13.1</td>
<td>8955 ± 8</td>
<td>24.6</td>
<td>62.8</td>
</tr>
<tr>
<td>11.4</td>
<td>10537 ± 31</td>
<td>32.3</td>
<td>77.8</td>
</tr>
<tr>
<td>9.9</td>
<td>12325 ± 98</td>
<td>44.3</td>
<td>97.7</td>
</tr>
<tr>
<td>8.3</td>
<td>14388 ± 143</td>
<td>56.1</td>
<td>122.2</td>
</tr>
<tr>
<td>7.3</td>
<td>16414 ± 197</td>
<td>71.3</td>
<td>152.2</td>
</tr>
</tbody>
</table>

Note: The change in neutron lifetime for a change in solution height from 29.5 cm to a solution height of 7.3 cm was a factor of 2.1.

$^a$Uncertainties are taken from Tables III and VI.
$^b$Uncertainties are one standard deviation of the mean from the least-squares fitting of data.
$^c$Average of values from Table V unperturbed by air bubbles.
$^d$Assumed to be equal to the average value from the ratio of spectral densities.
fractions, are higher than those from the ratios of spectral densities. In previous experiments, the validity of the BFNA method was usually limited to $k_{eff}$ values down to $\sim 0.8$.

CONCLUSIONS

The results and conclusions of these experiments are as follows:

1. The capability to measure the subcriticality for a multiplying system to $k_{eff}$ as low as 0.3 was demonstrated.

2. Experimental $k_{eff}$ values were obtained from the ratio of spectral densities in times as short as 6 s of data accumulation, and a small fraction of a second analysis time as a solution tank was drained from a height of 29.5 to 6.5 cm in $\sim 60$ s with corresponding changes in $k_{eff}$ from 0.95 to 0.30.

3. The measured $k_{eff}$ values obtained did not depend on the speed at which the solution height was changed or whether the tank was filling or draining.

4. The results of the dynamic measurements agreed with the static measurements.

5. Where static measurements were practical (limited to $k_{eff}$ down to $\sim 0.5$ by detection efficiency) with $^{3}\text{He}$ proportional counters sensitive to neutrons, the results agreed with those from measurements of spectral densities at low frequency were used successfully to obtain $k_{eff}$ values using a modified point interpretation of the data.

6. As in previous experiments, the ratios of spectral densities at low frequency were used successfully to obtain $k_{eff}$ values using a modified point interpretation of the data.

7. The neutron multiplication factors from independent measurements using the BFNA method with the values of $k_{eff}$ from the measured spectral densities down to $k_{eff}$ values of 0.65.

8. The effectiveness of this method for where conditions are changing as demonstratedly exceeds the dynamic requirements of most fuel plant processing applications.

9. Calculated $k_{eff}$ values using the KENO Carlo code and Hansen-Roach cross sections well with the experimental values.

The demonstrated dynamic capability of the method down to neutron multiplication factors allows continuous monitoring of the criticality of process tanks or dissolvers and thus allows for the elimination of excess conservatism from plant design, lead...
Fig. 9d. CPSD $G_{23}$ and APSDs $G_{22}$ and $G_{33}$ as a function of frequency for a solution height of 29.5 cm of source on the bottom of the tank and the scintillators located as shown in Fig. 2. (Lines are fitted functions.)

For detectors 2 and 3 located outside the fissile material system, as well as the APSD of a detector [Eq. (A.2)], were derived previously using the Langevin equation approach with the assumption that detector noise and reactor noise are not correlated:

\[
G_{11}(\omega) = 2|h_1(\omega)|^2 \left( \frac{31}{2} F_c q_c^2 + F_c q_c^2 \right), \tag{A.1}
\]

\[
G_{22}(\omega) = |h_2(\omega)|^2 \left[ 2W_2 F q_2^2 + W_2^2 (\bar{q}_2)^2 + \frac{W_2^2 (\bar{q}_2)^2}{\bar{p}^2} |H_s(\omega)|^2 G_s \right], \tag{A.2}
\]

\[
G_{12}(\omega) = 2h_1^*(\omega)h_2(\omega)\bar{q}_c - \frac{W_2 \bar{q}_2}{\bar{p}} H_s(\omega) \frac{\bar{p}_c F_c I_c}{I}, \tag{A.3}
\]

and

\[
G_{23}(\omega) = h_2^*(\omega)h_3(\omega) \frac{W_2 \bar{q}_2}{\bar{p}} \frac{W_3 \bar{q}_3}{\bar{p}} |H_s(\omega)|^2 G_s, \tag{A.4}
\]

where

\[
h_1(\omega), h_2(\omega), h_3(\omega)
\]

= response of the electronic component detection systems 1, 2, and 3, respectively, at frequency $\omega$

\[
\bar{q}_c, \bar{q}_a = \text{average charge produced in detector per } ^{252}\text{Cf spontaneous fission and per alpha respectively}
\]

\[
\bar{q}_2, \bar{q}_3 = \text{average charge produced per interaction in detectors 2 and 3, respectively}
\]

\[
W_2, W_3 = \text{detection efficiency of detectors 2 and 3, expressed as counts per \text{fission}}
\]

\[
W_1 = \text{efficiency for detecting } ^{252}\text{Cf fission assumed to be } \approx 1
\]

\[G_s = \text{APSD of the reactor noise-equivalent source}
\]

and an asterisk designates complex conjugates when frequencies much larger than delayed neutron constants,
where

\[ S_R^{-1} = \frac{\text{multiplication of all neutrons}}{\text{multiplication of californium neutrons}} \frac{1}{A} \]

or

\[ S_R^{-1} = \frac{F_c F_e \bar{v}_c + F_i F_i \bar{v}_i}{F_c F_e \bar{v}_c A} \quad (A.8) \]

The factor \( Y \) accounts for the effect of fissions induced by neutrons from the inherent source contributing to \( G_{23} \) but not to \( G_{12} \) or \( G_{13} \). If not all of the \( ^{252}\text{Cf} \) fissions are counted in the detection system due to undetected fissions of californium, producing neutrons that induce fissions in the system that contribute to \( G_{23} \) but not to \( G_{12} \) and \( G_{13} \).

When a multiplying system is far subcritical, the power spectral density of the noise-equivalent source \( G_2 \) must be modified to include \( X' \), a modified form of the neutron dispersion number given by Eqs. (A.5) and (A.6). Then, \( \frac{1 - k_{\text{eff}}}{k_{\text{eff}}} \) is of the form

\[ \frac{1 - k_{\text{eff}}}{k_{\text{eff}}} = \frac{C_1 G_{12} G_{13} / G_{11} G_{23}}{1 - C_2 G_{12} G_{13} / G_{11} G_{23}} \quad (A.10) \]

instead of

\[ \frac{1 - k_{\text{eff}}}{k_{\text{eff}}} = C \frac{G_{12} G_{13}}{G_{11} G_{23}} \quad (A.11) \]

where \( C, C_1, \) and \( C_2 \) are constants involving parameters defined by Eqs. (A.1) through (A.11), some of which depend on solution height.

If it is assumed that reactor and detector noises are completely correlated, \( \bar{v}_c \) is unity because the contribution to the signal from all \( ^{252}\text{Cf} \) fissions is the same. Since there is no significant inherent neutron source in the uranyl nitrate solution comparable in size to the \( ^{252}\text{Cf} \) source, the third term in Eq. (A.5) is zero, and since >99.5% of the \( ^{252}\text{Cf} \) spontaneous fission produces signals in channel 1 and these signals are counted, \( Y = A = S_R = 1 \).

The average number of neutrons per uranium fission was obtained by using the fluxes from transport theory calculations and the ENDF/B-IV data to calculate the total number of neutrons per fission. This averaging accounts properly for the dependence of the number of neutrons per fission on the energy of the neutron inducing the fission. Changing solution height resulted in a ~0.2% change in the number of neutrons per fission, and resulting average number of neutrons per uranium fission was \( \bar{v} = 2.426 \). The value of the average number of prompt neutrons per fission \( \bar{v} = 2.408 \) and was used in the interpretation at all solution heights.

The number of delayed neutrons per uranium fission was calculated from the delayed neutron yield, the delayed neutron effectiveness factors obtained from the forward and adjoint fluxes from transport theory, and the delayed neutron spectra of Batch and McKhyder. The effective delayed neutron fission calculated in this way is 0.0075.

The value of the Diven factor \( X = \bar{v} (\bar{v} - 1) \) was obtained from the measurements of prompt neutron multiplicities for uranium. Its value does not depend significantly on the energy of the neutrons inducing fission and varies only from 0.796 to 0.80 when the neutron energy varies from thermal to 20 MeV (Ref. 25). The value of \( X \) used was 0.80. The values of the number of prompt neutrons per \( ^{252}\text{Cf} \) fission its mean square \( \bar{v}_c \) and \( \bar{v}_c^2 \) are from the measurements of prompt neutron multiplicities of Spel et al., \( \bar{v}_c = 3.773 \) and \( \bar{v}_c^2 = 15.818 \).
yields $k_{\text{eff}}$ values that differ from those for the uncorrelated assumption below 0.85, and the difference increases as $k_{\text{eff}}$ decreases to as much as a factor of 4 at $k_{\text{eff}} = 0.35$. The results of the interpretation, assuming that reactor and detector noise are correlated, also differ from other measurements and calculations.

APPENDIX C

CALCULATED NEUTRON MULTIPLICATION FACTORS

The neutron multiplication factors were calculated using the Monte Carlo method with the KENO code and Hansen-Roach and ENDF/B-IV cross sections. The calculations included the source and its Lexan tube and the reflection effect of the scintillation detector as located in the measurements. The results of the calculations are given in Table C.1. The small reaction effect of the detectors that cannot be reliably obtained from the calculations by subtracting the calculated neutron multiplication factors from calculations with and without the detector was included because of the statistical uncertainty of the calculations (±0.005). $k_{\text{eff}}$ values using the ENDF/B-IV data were consistently higher than those from Hansen-Roach cross sections by an average of 2% in $k_{\text{eff}}$. The calculated

<table>
<thead>
<tr>
<th>Solution Height (cm)</th>
<th>Calculated Neutron Multiplication Factors</th>
<th>Hansen-Roach Cross Sections</th>
<th>ENDF/B-IV 27 Group (No Source or Scintillators)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Source and Scintillators$^b$</td>
<td>Source Only$^c$</td>
<td>No Source or Scintillators$^d$</td>
</tr>
<tr>
<td>30.5</td>
<td>0.949</td>
<td>0.950</td>
<td>0.963</td>
</tr>
<tr>
<td>27.9</td>
<td>0.939</td>
<td>0.916</td>
<td>0.902</td>
</tr>
<tr>
<td>25.4</td>
<td>0.894</td>
<td>0.896</td>
<td>0.902</td>
</tr>
<tr>
<td>24.0</td>
<td>0.897</td>
<td>---</td>
<td>0.902</td>
</tr>
<tr>
<td>22.9</td>
<td>0.867</td>
<td>0.870</td>
<td>---</td>
</tr>
<tr>
<td>22.0</td>
<td>0.862</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>20.4</td>
<td>0.844</td>
<td>0.831</td>
<td>0.841</td>
</tr>
<tr>
<td>15.2</td>
<td>0.721</td>
<td>0.721</td>
<td>0.741</td>
</tr>
<tr>
<td>10.2</td>
<td>0.553</td>
<td>0.547</td>
<td>0.544</td>
</tr>
<tr>
<td>5.08</td>
<td>0.241</td>
<td>0.241</td>
<td>0.249</td>
</tr>
</tbody>
</table>

$^a$Statistical error is ±0.005. All calculations included the bottom, sides, and top of the tank. Where the source was included in the calculation, the 2.54-cm-o.d. Lexan tube was included in the calculation. As a result, with the source located at any height of solution, the Lexan tube displaced solution on the axis of the cylinder above the source position.

$^b$Source located at the bottom of the tank and the scintillators as shown in Fig. 2 were included in the calculation.

$^c$Source at the center of solution height on the axis of the tank was included in the calculation.

$^d$Source and scintillators were not included in the calculations.
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