Title: Subcritical Neutron Multiplication Measurements of HEU Using Delayed Neutrons as the Driving Source

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A new method for the determination of the multiplication of highly enriched uranium systems is presented. The method uses delayed neutrons to drive the HEU system. These delayed neutrons are from fission events induced by a pulsed 14-MeV neutron source. Between pulses, neutrons are detected within a medium efficiency neutron detector using $^3$He ionization tubes within polyethylene enclosures. The neutron detection times are recorded relative to the initiation of the 14-MeV neutron pulse, and subsequently analyzed with the Feynman reduced variance method to extract singles, doubles and triples neutron counting rates. Measurements have been made on a set of nested hollow spheres of 93% enriched uranium, with mass values from 3.86 kg to 21.48 kg. The singles, doubles and triples counting rates for each uranium system are compared to calculations from point kinetics models of neutron multiplicity to assign multiplication values. These multiplication values are compared to those from MCNP K-Code calculations.
Introduction

The problem of determining the neutron multiplication of subcritical systems of highly enriched uranium (HEU) has always been more difficult than the equivalent problem in plutonium systems. In the latter, the driving neutron source is usually neutrons from the spontaneous fission of $^{239}$Pu. The $^{239}$Pu is evenly distributed throughout the volume of the plutonium. Point kinetic models using multiplicity analysis of the observed neutron time distributions have been successfully applied to determine the multiplication of subcritical plutonium systems.

Since $^{235}$U has a long half life for spontaneous fission ($\sim 2.5 \times 10^7$ years), no passive neutron technique exists for extracting attributes of an HEU system, rather, an active method using an external neutron source is required. This driving neutron source is seldom evenly distributed throughout the system volume. The best situation occurs when it is possible to place the neutron source centered within the volume of the HEU system. For this case the ratio of the neutron counting rates in a detector external to the HEU volume, with and without the HEU in place, determines the "leakage" multiplication. If the center of the assembly is not accessible, the source is placed at a fixed external position opposite the neutron detector. The leakage multiplication determined in this manner has been shown to be dependent upon the exact geometrical relationships between the HEU system, the neutron source, and the detector, as well as upon the neutron source energy distribution.

A new method for the investigation of HEU systems is presented that uses an external radiation probe of 14-MeV neutrons to induce fission of the uranium in the sample. The 14-MeV neutron source is operated in a pulsed mode with a pulse width of approximately 20 microseconds at a frequency of 50 Hz. The total neutron output is approximately $5 \times 10^7$ neutrons per second. Between pulses of the interrogating probe and after the "prompt" neutron signal has died away, neutrons are detected by a high-efficiency neutron detector system consisting of $^3$He gas tubes located within polyethylene moderating material. The HEU sample is located within a cadmium-lined box within the neutron detector assembly to shield the sample from low-energy neutrons. The neutron detection times are recorded and subsequently analyzed with the Feynman reduced variance-to-mean method [1,2,3]. This analysis provides a measure of the number of "single," "double," and "triple" neutron events that originate from both the delayed neutrons and delayed neutron induced fission chain events. The driving neutron source is the delayed neutrons born from fission products that have been distributed throughout the system by the interactions of uranium nuclei with the 14-MeV neutrons during the interrogating probe pulse.

Experimental Results

A series of measurements was made on a set of bare, enriched uranium metal hemispheres with a constant inner radius of 2.34 cm. The outer radius varied from 4.00 cm to 6.67 cm in steps of approximately 1/3 cm. The mass of these nine HEU systems vary from 3.86 kg to 21.48 kg.

The N1, N2, and N3 counting rates as a function of time after the neutron beam burst are shown in Fig. 1 for the 14.36 kg sample. The N1/sec data, which decays sharply from the earliest times
Fig. 1. The singles (circles), doubles (diamonds), and triples (triangles) measured for the 14.36-kg HEU sample.

measured at 2000 microseconds after the beam burst, contains contributions from the initial neutron burst as well as scattered neutron background. The counting rates for doubles and singles are essentially constant. Similar results are obtained for the other eight HEU configurations.

The neutron background was measured with the neutron detector empty, and subtracted from the results with the HEU samples. The background-corrected results between 8000 and 16000 microseconds for N1/sec, N2/sec, and N3/sec were averaged and statistical errors were assigned.

Comparison to Hage-Cifarelli Formalism

The measured ratios of doubles to singles and triples to singles have been compared to those calculated with the Hage-Cifarelli formalism [3] to extract values of the neutron multiplication. According to this formalism, the R2/R1 and R3/R1 ratios have the following dependence upon the neutron detection probability $\varepsilon$ and the leakage multiplication $M_L$:

\[
R2/R1 = (\varepsilon) \times [M_L \times \{(M_L - 1) \times \nu_{12}/(\nu_{11} - 1)\}], \quad \text{and}
\]

\[
R3/R1 = (\varepsilon^2) \times [M_L^2 \times \{(M_L - 1) \times \nu_{13}/[\nu_{11} \times \nu_{12} + 2 \times (M_L - 1) \times \nu_{12}^2/(\nu_{11} - 1)]\}].
\]

Here $\nu_{11}, \nu_{12},$ and $\nu_{13}$ are the first, second, and third reduced moments of the neutron probability distribution for neutron induced fission of $^{235}$U.

The results for the ratios N2/N1 are shown in Fig. 2, compared to the values calculated with the
Fig. 2. The variation of the ratio of the observables N2 /N1 obtained with delayed neutron interrogation compared to the multiplication obtained from the calculations from the Hage-Cifarelli model.

Hage-Cifarelli formalism. The results for the ratios N3/N1 are shown in Fig. 3. For the calculations, a neutron detection probability of 0.138 obtained from a multiplicity measurement with a $^{252}$Cf neutron source, was used.

Fig. 3. The variation of the ratio of the observables N3 /N1, obtained with delayed neutron interrogation compared to the multiplication obtained from the calculations from the Hage-Cifarelli model.
Total multiplications for these systems have been obtained from MCNP K-Code calculations [4]. These results have been summarized in Table I. The K-Code values are approximately 50% larger than the leakage multiplication values. The relationship for leakage multiplication $M_L$ and total multiplication $M_T$ as given by Serber [5] is

$$M_T = (M_L * v - 1 - \alpha)/(v - 1 - \alpha)$$

where $v$ is the average number of neutrons emitted/fission and $\alpha$ is the ratio of neutron capture cross section to fission cross section. For the case of $^{235}$U, we assume $\alpha = 0$, and $v = 2.41$.

The total multiplication values obtained from the leakage multiplication values are also listed in Table I. These values are plotted against the MCNP K-code calculation values in Fig. 5. A linear
Fig. 5. The total multiplication values from delayed neutron interrogation vs. the MCNP-K-Code values. 

fit to the data yields the linear relationship: 

\[ M_{T\text{-Leakage}} = 1.063 \times M_{K\text{-Code}} \]

The total multiplication values obtained from the data are \( \sim 6\% \) larger than the K-Code values. This difference may be due to the energy distribution of the delayed neutrons. Another possible multiplication enhancement may be due to neutrons reflected back into the HEU system from the polyethylene enclosures of the neutron detector.

**Mass Determination**

According to the Hage-Cifarelli model, the singles counting rate \( N_1/\text{sec} \) should vary as

\[ N_1/\text{sec} = \varepsilon \times M_L \times S_d, \]

where \( S_d \) is the number of delayed neutrons emitted per second, \( M_L \) is the leakage multiplication and \( \varepsilon \) is the neutron detection probability. \( S_d \) should be proportional to the 14 MeV neutron flux, the 14-MeV \( n\text{-fission} \) cross section, the number of delayed neutrons emitted per fission, the mass of the \(^{235}\text{U}\) and the multiplication. Thus the following relationship is obtained:

\[ N_1/\text{sec} = M_L \times (\text{Mass} \times M_L \times A), \text{ or} \]

\[ = A \times M_L^2 \times \text{Mass}, \]
where $A$ (a constant) includes $\varepsilon$, the neutron flux, the $n$-fission cross section and the number of delayed neutrons per fission. The variation of $N_1$/sec divided by the square of the multiplication, as a function of mass, is shown in Fig. 4, along with a linear least squares fit to the data. Solving for the mass yields the following:

$$\text{Mass} = 43.9 \times \frac{(N_1/\text{sec})}{(M_L^2)}.$$  

This linear relationship between the mass and $(N_1/\text{sec})/(M_L^2)$ is indicative that the fission products, and hence the source of delayed neutrons, are evenly distributed throughout the volume of the HEU samples.

**Summary**

A new method of using delayed neutrons to determine neutron multiplication values has been developed and applied to nine bare HEU systems of hollow spheres. These multiplication values are found to be linearly related to those obtained from MCNP K-Code calculations, and are approximately 6% larger. This method of measurement and analysis places the determination of multiplication of HEU systems on an equal basis with plutonium systems driven by spontaneously fissioning $^{239}$Pu.

**References**

5. R. Serber, Los Alamos National Laboratory report LA-335, (July 1945).