

*Title:* **Non Destructive Analysis of Shielded Highly Enriched Uranium**

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## Non Destructive Analysis of Shielded Highly Enriched Uranium

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### Abstract

A new method for the nondestructive analysis of shielded highly enriched uranium (HEU) is presented. This method uses a source of 14-MeV neutrons to penetrate the shielding material and induce fission in the HEU sample. The 14-MeV neutron source is pulsed at a rate of 50 Hz. Between pulses neutrons are detected with a medium efficiency  $^3\text{He}$  based neutron detector. These neutrons are delayed neutrons from fission products resulting from HEU fission events induced during the pulse, as well as from fission events induced by the delayed neutrons from the fission products. The time history of neutron detection is recorded and subsequently analyzed according to the Feynman reduced variance method to yield rates of detection of one (N1), two (N2) and three (N3) neutrons from fission events. Ratios of N2/N1 and N3/N1 are compared to model calculations to extract the neutron multiplication. The rate N1/sec is used to deduce the HEU system mass. Measurements were performed on a bare 4.79 kg HEU system, and this HEU system shielded within lead, iron, carbon, and beryllium hemispheres. The major effect of the shielding materials was to increase the system neutron multiplication, resulting from reflection of neutrons back onto the HEU.

### 1. INTRODUCTION

In previous measurements [1] we have developed a method for determining the neutron multiplication of sub-critical bare metal systems of highly enriched uranium (HEU). This method uses delayed neutrons from fission products as the driving neutron source for bare metal HEU systems. In this work we present results for HEU system enclosed within various shielding materials of low density, (carbon and beryllium), medium density (iron) and high density (lead ).

The method presented here uses delayed neutrons from fission products distributed throughout the HEU sample as a source of interrogating neutrons. The fission products are from fission events produced by an external radiation probe of pulsed 14-MeV neutrons. Neutrons are detected between pulses of the interrogating probe by a medium efficiency neutron detector system. The neutron detection times are recorded, and subsequently analyzed with the Feynman reduced variance method [2,3,4]. This analysis provides a measure of the number of “single”, “double”, and “triple” neutron events detected from fission events. Ratios of doubles/singles and triples/singles are compared to model calculations to extract values for the multiplication of the HEU sample. Neutron detection probabilities are determined for each shielding configuration by multiplicity measurements on a  $^{252}\text{Cf}$  neutron source.

## 2. Experimental Apparatus

### 2.1. External Interrogation Probe

The external radiation probe consisted of 14-MeV neutrons from a pulsed neutron generator. The neutron generator produced neutrons by means of the deuteron + triton nuclear reaction which yields  $\sim 14$ -MeV neutrons and  $\sim 3.4$ -MeV alpha particles. The neutron generator produced pulses of neutrons of approximately  $20\ \mu\text{s}$  width at a frequency 50 Hz. The neutron intensity was  $\sim 1 \times 10^6$  neutrons /pulse providing a total neutron output of approximately  $5 \times 10^7$  neutrons/sec into  $4\pi$  steradian.

### 2.2. Neutron Detector System

The neutron detector system contained forty-eight  $^3\text{He}$  gas ionization tubes, arranged into sixteen modules, each containing three tubes. The three tubes were within a polyethylene cavity, which thermalized the neutrons. The neutron detection occurs by the neutron +  $^3\text{He} \Rightarrow$  proton +triton reaction. The  $^3\text{He}$  gas pressure was two atmospheres. The half-life for neutron die away in the detector was determined to be 54.1 microseconds from a Rossi- $\alpha$  type time distribution measurement of a  $^{252}\text{Cf}$  neutron source. The detector modules formed an enclosure 40 cm x 40 cm x 116 cm. The ends of the enclosure were open. The experimental geometry is shown schematically in Fig. 1. The total efficiency for detection of fission neutrons was

**$\sim 27\%$  Neutron Detector**

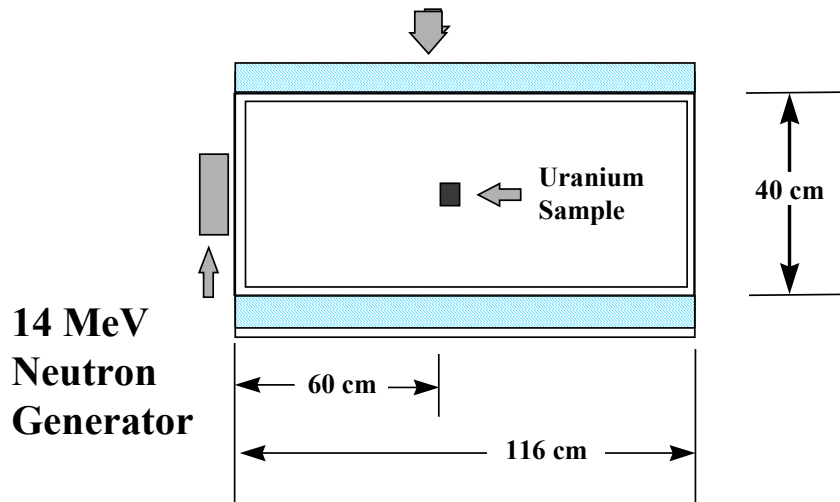


FIG. 1. Plan view of the experimental apparatus.

approximately 27% determined from a multiplicity analysis for a  $^{252}\text{Cf}$  neutron source located at the center of the enclosure. The HEU sample within the various shielding configurations was placed within a cadmium-lined box to prevent low energy neutrons from reaching the sample. The neutron generator was located at one of the open ends of the detector 60 cm from the center of the HEU sample

Signals from the  $^3\text{He}$  neutron detectors were amplified and discriminated to form logic pulses in the Los Alamos designed preamplifier, amplifier, discriminator, ECL logic pulse module, (PADEM) [5] then configured as input to the 15 channels of the custom Los Alamos designed pulse arrival time recording module (PATRM) [6]. The PATRM uses a logic pulse, designated VETO, to control data acquisition in active mode. At the onset of the VETO pulse, the PATRM disables all data inputs, writes a code marker to its internal memory at the next two memory locations, stops the internal clock counter and resets this counter to zero. Upon lifting of the VETO, the clock counter and all data inputs are enabled. The result of the measurement is a history of the signal arrival times relative to the end of the VETO pulse. The clock frequency of the PATRM was 5 megahertz, allowing the signal arrival time to be measured with an accuracy of  $0.2\ \mu\text{s}$ . Up to a million events can be recorded before transferring the data to a small computer for analysis and archiving. The VETO input to the PATRM was the logic pulse of length 1000  $\mu\text{s}$  derived from the trigger pulse from the neutron generator. The PATRM was disabled during the beam pulse and for most of the time in which the  $^3\text{He}$  detectors were able to detect neutrons from the interrogating probe burst. The majority of pulses recorded by the PATRM were from delayed neutrons from fission products and neutrons from fission events induced by the delayed neutrons.

### 3. Highly Enriched Uranium Sample And Shielding Materials

The HEU sample used was configured as a solid sphere of radius 4.0 cm and mass 4.79 kg, from a set of nesting hollow hemispheres of average density  $18.675\text{g/cm}^3$ , enriched to 93.12%  $^{235}\text{U}$  [7]. The shielding materials, listed in Table I, were hollow hemispheres that enclosed the HEU. The thickness of the shielding materials is also listed in the table.

### 4. Measurement Protocol.

Each configuration of the HEU shells was investigated with delayed neutrons from fission events initiated by 54,000 14-MeV neutron pulses. A small plastic scintillator viewed by a fast photo-multiplier tube was placed at a distance of one meter from the neutron source and was used as a relative 14-MeV neutron flux monitor.

### 5. Experimental Observables, Feynman Variance Analysis

We have applied the reduced variance method of analysis developed initially by Feynman, deHoffmann, and Serber [2], and used by other investigators [3,4] of spontaneous fissioning systems. The time history of the neutron events recorded in the PATRM module, is examined to construct the Feynman histograms,  $\text{FN}_{\text{ORDER}}$ , formed by randomly opening an inspection time interval of fixed width and then counting the number of neutron signals that are present within this time interval. A histogram array is then incremented according to the number of neutrons found, and the process is repeated for the duration of the measurement. The resulting distribution then contains the number of times that zero, one, two, three, etc., neutrons were found within the inspection interval. The  $\text{FN}_{\text{ORDER}}$  distributions are converted to probability distributions  $\text{P}_{\text{ORDER}}$  by dividing the  $\text{FN}_{\text{ORDER}}$  by the number of times the inspection interval ( $\text{NUM\_INT}$ ) was opened.

$$\text{P}_{\text{ORDER}} = \text{FN}_{\text{ORDER}} / (\text{NUM\_INT}).$$

The first, second and third reduced factorial moments of the  $P_{\text{ORDER}}$  distributions are then formed.

$$\text{Mom1} = \sum \{m * P_m\} \text{ and}$$

$$\text{Mom2} = \sum 1/2 * \{m * (m-1) * P_m\}.$$

$$\text{Mom3} = \sum 1/6 * \{m * (m-1) * (m-2) * P_m\}.$$

The first moment is the average, or mean, number of neutrons found in the time interval. The first moment divided by the interval value ( $\Omega$ ) is the average neutron counting rate during the measurement,

$$N1/\text{sec} = \text{Mom1} / \Omega.$$

The second moment represents the variance of the distribution. The difference between the variance and the square of the mean is a measure of the amount of correlation present in the data. This difference divided by the inspection interval width is then the number of correlated neutrons per second,  $N2/\text{sec}$ , present in the data;

$$N2/\text{sec} = \{ \sum m(m-1) * P_m - 1/2 * (\sum m * P_m)^2 / (\sum m * P_m) \} / \Omega.$$

The following combination of the first, second and third reduced moments divided by the interval width is the number of triplet neutrons per second,  $N3/\text{sec}$ , present in the data,

$$N3/\text{sec} = (\text{Mom3} - \text{Mom2} * \text{Mom1} + (1/3) * \text{Mom1}^3) / \Omega.$$

For a neutron source that has no correlation's, such as neutrons from an Americium-Lithium source, the above distributions are those of a pure Poisson distribution, and  $N2/\text{sec}$  and  $N3/\text{sec}$  are identically zero. Thus the observation of nonzero values for  $N2/\text{sec}$  and  $N3/\text{sec}$  are positive indicators of correlations in the neutron signal, which in turn is a positive indicator of neutrons from fission events.

For a passive interrogation of a system containing a spontaneously fissioning system such as  $^{240}\text{Pu}$  or  $^{252}\text{Cf}$ , the fission rate is essentially constant with time, although the time between any two fission events is completely random. The fission rate is not necessarily constant for a system such as  $^{235}\text{U}$  undergoing active interrogation. In order to at least partially compensate for any time dependence, we form many separate Feynman distributions. The first distribution is for an interval starting 1000  $\mu\text{s}$  after the neutron generator burst. The inspection window is opened once for each probe burst to form this first Feynman histogram. The inspection time interval was chosen to be 200  $\mu\text{s}$ . The second Feynman histogram begins one inspection time interval later, the third Feynman histogram two time intervals later, etc. For the data described here, the neutron generator produced a beam burst every 20,000  $\mu\text{s}$ . Thus, ninety-five Feynman distributions were generated. From these Feynman distributions ninety-five values for  $N1/\text{sec}$ ,  $N2/\text{sec}$  and  $N3/\text{sec}$  were calculated. Each of these distributions are for inspection intervals that are random in time with respect to any fission event.

## 6. Experimental Results

### 6.1. The time dependence of N1/sec, N2/sec and N3/sec

The N1/sec, N2/sec and N3/sec distributions for the 4.79 kg HEU sphere are shown on a log scale in Fig. 2. The curve for the “singles” measurement N1/sec is shown as diamonds,

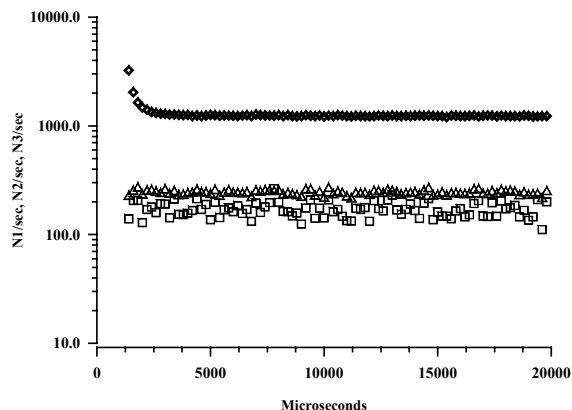


FIG. 2. The time dependence of N1/sec(diamonds), N2/sec (triangles) and N3/sec (squares) after the 14-MeV neutron beam bursts for the bare configuration containing 4.79 kg of HEU.

the “doubles”, N2/sec, distribution as triangles and the “triples”, N3/sec, distribution as squares. No data is recorded during the burst and for the following 1000  $\mu$ s. At the earliest times, there are a substantial number of neutrons from the neutron beam burst and from fission events induced during the burst. From 1000 to 2000  $\mu$ s, a sharp decrease in the N1/sec distribution is observed, reflecting the time constant associated with the neutron detector. The N2/sec and N3/sec distributions are essentially constant throughout the full time of the measurement. Similar distributions were obtained for each of the HEU plus shielding configurations listed in Table I. After background corrections, average values for N1/sec, N2/sec and N3/sec were calculated from the data between 8000 and 16000  $\mu$ s, and a statistical error assigned by calculating the variances about the mean for the forty independent measurements.

### C. N1/sec, N2/sec and N3/sec Observables for Shielded HEU Configurations

The values for the ratios N2/N1 and N3/N1 are shown in Fig. 3 for the HEU bare, and in 1.27 cm, 3.18 cm and 4.45 cm of lead shielding, in Fig. 4 for the HEU bare and in 1.27 cm and 3.18 cm of iron shielding, and in Fig. 5 for the HEU bare and in 1.27 cm and 3.18 cm of  $^{12}\text{C}$ , and 4.13 cm of  $^9\text{Be}$ . For all cases the N2/N1 and N3/N1 ratios are larger for the shielded configurations compared to the bare HEU configuration.

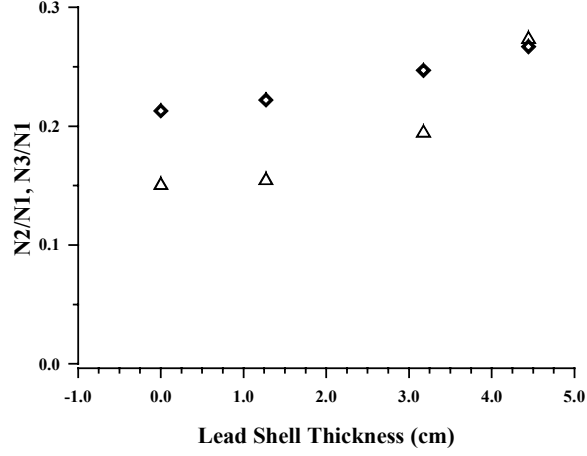


FIG. 3.  $N2/N1$  and  $N3/N1$  for the HEU sample bare and in 1.27 cm, 3.18 cm and 4.45 cm of lead.

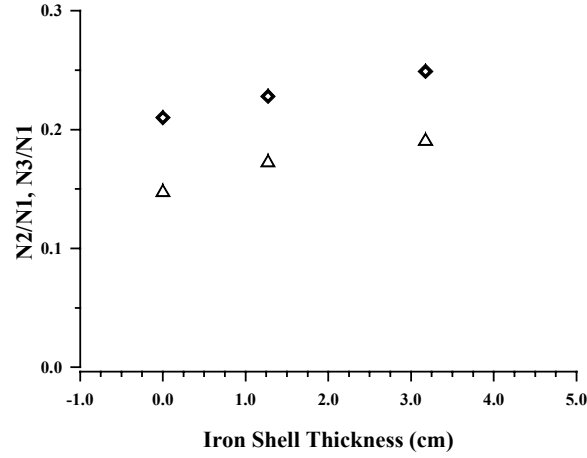


FIG. 4.  $N2/N1$  and  $N3/N1$  for the HEU sample bare and in 1.27 cm and 3.18 cm of iron.

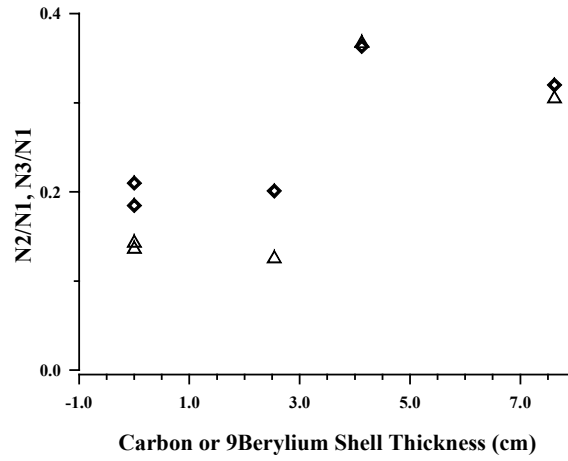


FIG. 5.  $N2/N1$   $N3/N1$  for the HEU sample bare and in 1.27 cm and 3.18 cm of  $^{12}\text{C}$ , and 4.13 cm of  $^9\text{Be}$ .

## 7. Multiplication Determination.

We compare the ratios of  $N2/N1$  and  $N3/N1$  to values calculated within the Hage-Cifarelli[4] model to extract values of multiplication. According to this model  $N2/N1$  and  $N3/N1$  have the following dependence on the neutron leakage multiplication  $M_L$ :

$$N2/N1 = (\varepsilon) * [M_L * \{(M_L - 1) * v_{12} / (v_{11} - 1)\}] \text{ and,}$$

$$N3/N1 = (\varepsilon^2) * M_L^2 * \{(M_L - 1) / (v_{11} - 1) * [v_{13} + 2 * (M_L - 1) * v_{12}^2 / (v_{11} - 1)]\}.$$

Here  $\varepsilon$  is the neutron detection probability, and  $v_{11}$ ,  $v_{12}$ , and  $v_{13}$  are the first, second, and third moments of the neutron emission probabilities for induced fission. The  $M_L$  values are converted to total multiplication values  $M_T$  using the relationship from Serber[8]

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

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  $^{235}\text{U}$ , we use  $\alpha = 0.03$ , and  $v = 2.62$ .

## 8. Mass Determination

According to the Hage-Cifarelli[4] model, the singles counting rate  $N1/\text{sec}$  should vary as

$$N1/\text{sec} = \varepsilon * M_L * S_d,$$

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-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:

$$N1/\text{sec} = M_L * (\text{Mass} * M_L * A).$$

The constant  $A$  contains the dependencies on the 14-MeV neutron flux, the 14-MeV n-fission cross-section, the number of delayed neutrons emitted per fission and the neutron detection probability. Here  $A$  is determined from the measurement on the bare HEU system. Table I contains the mass values obtained using this approach.



Table 1. Values for the experimental and calculated neutron multiplication and HEU mass.

Material	Thickness cm	Multiplication Total Experiment	Multiplication Total Onedant	Ratio Exp/Calc	Mass Experimental
None	0.00	1.89	1.85	1.02	4.79
Beryllium	1.59	2.17	2.44	0.89	4.33
Carbon	2.54	1.84	1.89	0.97	5.08
Iron	3.18	1.97	1.92	1.03	4.67
Iron	1.27	1.92	1.89	1.02	4.71
Lead	4.45	2.01	1.92	1.04	4.60
Lead	1.27	1.92	1.89	1.02	4.71

## 9. Summary

A new method to determine neutron multiplication values using delayed neutrons has been developed and applied to shielded HEU systems. The neutron multiplication values are found to be within four percent of those calculated using the Onedant computer code, except for the beryllium case where the experimental value disagrees with the calculation by eleven percent. The primary effect of the shielding material is to increase the system neutron multiplication as a result of the reflection of neutrons back onto the HEU. The mass values for the shielded cases agree with the true mass to better than 10 percent.

## References

- [1] HOLLAS, C. L. et al., "Sub-critical Neutron Multiplication Measurements Of HEU Using Delayed Neutrons As The Driving Source", Sixth International Conference on Nuclear Criticality Safety, Versailles (1999),
- [2] FEYNMANN, R. P. et. al., "Dispersion of the Neutron Emission in U-235 Fission", Journal of Nuclear Energy, **3**, (1956) 64-69.
- [3] ROBBA, A. A. et al, "Neutron Multiplication Measurements Using Moments of the Neutron Counting Distribution", Nucl. Instr. and Meth. **215**, (1983) 473-479.
- [4] CIFARELLI, D. M. and HAGE, W. "Three Parameter Analysis of Neutron Signal Correlation Measurements", Nucl. Instr. and Meth. **A251**, (1986) 550-563.
- [5] ARNONE, G. J., private communication.
- [6] ARNONE, G. J, et al , 1992 IEEE Nuclear Science Symposium, Orlando, Florida.
- [7] TUCK, G., "Enriched Uranium-Metal Measurements", NO.1, RFP-907 UC-46 CRITICALITY STUDIES TID-4500, Dow Chemical Company, Rock Flats Division, Golden, Co. (July 1967).
- [8] SERBER, R., "The Definition of Neutron Multiplication", LA-335, (July, 1945).