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*Title:* **Energy-Dependent Bias in Plutonium Verification Measurements Using Thermal Neutron Multiplicity Counters**

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# ENERGY-DEPENDENT BIAS IN PLUTONIUM VERIFICATION MEASUREMENTS USING THERMAL NEUTRON MULTIPLICITY COUNTERS

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# ENERGY-DEPENDENT BIAS IN PLUTONIUM VERIFICATION MEASUREMENTS USING THERMAL NEUTRON MULTIPLICITY COUNTERS*

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## ABSTRACT

Multiplicity analysis algorithms are extended to include the effect of ($\alpha$,n) neutron energies on the detector efficiencies, induced fission probabilities, and induced fission factorial moments. The analysis is restricted to plutonium oxide. Bias is calculated as a function of ($\alpha$,n) neutron energy for six thermal neutron coincidence counters: HLNC, AWCC, 3RMC, PSMC, PYRO, and 5RMC. Ring ratio data for the 3RMC are used to reduce energy-dependent bias in the verification of impure plutonium oxide. The utility of the AWCC as a multiplicity counter is considered.

## 1. INTRODUCTION

Although thermal neutron multiplicity counting has become a routine technique for the verification of impure plutonium samples, important characteristics of multiplicity counters and assumptions made in the multiplicity analysis algorithms have not been adequately studied. The moments formalism for multiplicity analysis [1,2] provides very convenient algorithms for the calculation of neutron multiplication, ($\alpha$,n) neutron yield, and effective $^{240}$Pu mass, from which the plutonium mass is calculated from the isotopic composition.[3] The moments analysis presently used for IAEA plutonium verification measurements [4] is based on a number of simplifying assumptions, including the following.

1. There is no neutron moderation in the sample.
2. There is no neutron absorption in the sample except for induced fission reactions.
3. Neutron multiplication is constant throughout the sample.
4. Neutron multiplication is independent of neutron energy.
5. Neutron detection efficiency is independent of sample geometry.
6. Neutron detection efficiency is independent of neutron energy.

These assumptions are never quite true, and sometimes they are not even good approximations. The validity of the assumptions depends strongly on the characteristics of the material being verified. Presently, the IAEA verifies inventories of impure plutonium oxide. The inventories are cans of impure oxide with plutonium content in the kilogram range. The samples do not have a large hydrogenous content, so neutron moderation and absorption in the samples are not large effects. Kilogram quantities of impure plutonium oxide have small dimensions relative to the sample cavity of a typical multiplicity counter, so the dependence of the neutron detection efficiency on sample geometry is small. However, neutrons from ($\alpha$,n) reactions on impurities can have energies from a fraction of an MeV to several MeV. Because the neutron detection efficiency depends on neutron energy in a thermal neutron counter, the amount and type of impurities can have a significant effect on the accuracy of multiplicity assay if the efficiency is assumed to be constant. Similarly, because neutron multiplication of ($\alpha$,n) neutrons depends on the ($\alpha$,n) neutron energy, an assay bias can result from the assumption that the neutron multiplication is constant. Finally, neutrons emitted from the center of the sample have a greater neutron multiplication than those emitted from the surface of the sample, so a significant bias can also result from this effect if the multiplication is assumed to be constant; this is very important for highly multiplying plutonium metal samples [5] and is significant for kilogram quantities of plutonium oxide. To minimize the effects of variable ($\alpha$,n) energies from impure plutonium samples, thermal neutron counters designed to be used as multiplicity counters have

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been configured so that the neutron detection efficiency is insensitive to neutron energy; the design is ultimately constrained by practical considerations—especially cost.

This study is concerned with the assay bias that results from variable (α,n) neutron energies in plutonium oxide samples. It is assumed that the effects of moderation and absorption in the samples are small, that the neutron detection efficiency is constant over the sample volume, and that the neutron multiplication is constant for an individual sample. The last assumption was made for two reasons. First, it is important to study the energy-dependent effects independently of other effects; second, the bias that results from nonconstant neutron multiplication will be similar from one oxide sample to another if they have similar masses, densities, and shapes. The moments equations are extended to allow for variable (α,n) neutron energies. Assay bias is then calculated as a function of neutron energy for several thermal neutron counters assuming that the energy-independent moments equations are used for the analysis. An assay procedure is developed based on the ring ratio as an approximate measure of the (α,n) neutron energy; the procedure is applied to a measurement data set acquired from impure plutonium oxides.

## 2. ENERGY DEPENDENCE

### 2.1 Thermal Neutron Detector Efficiencies

Calculations were done for six thermal neutron detectors. The neutron detection efficiency vs neutron energy is shown in Fig. 1 for each of the detectors. The dependence of the efficiency on energy was determined from Monte Carlo calculations using the Los Alamos MCNP code.[6] Most of the calculations on the energy dependence of the efficiency were performed at the time the counters were designed. Typically, the efficiency was calculated at 0.1, 0.2, 0.5, 1, 2, 3, 4, and 5 MeV. The efficiencies plotted in Fig. 1 are straight-line connections between available calculated efficiencies. The detector efficiencies given below for each counter are efficiencies for $^{252}$Cf neutrons emitted at the center of the sample cavity.

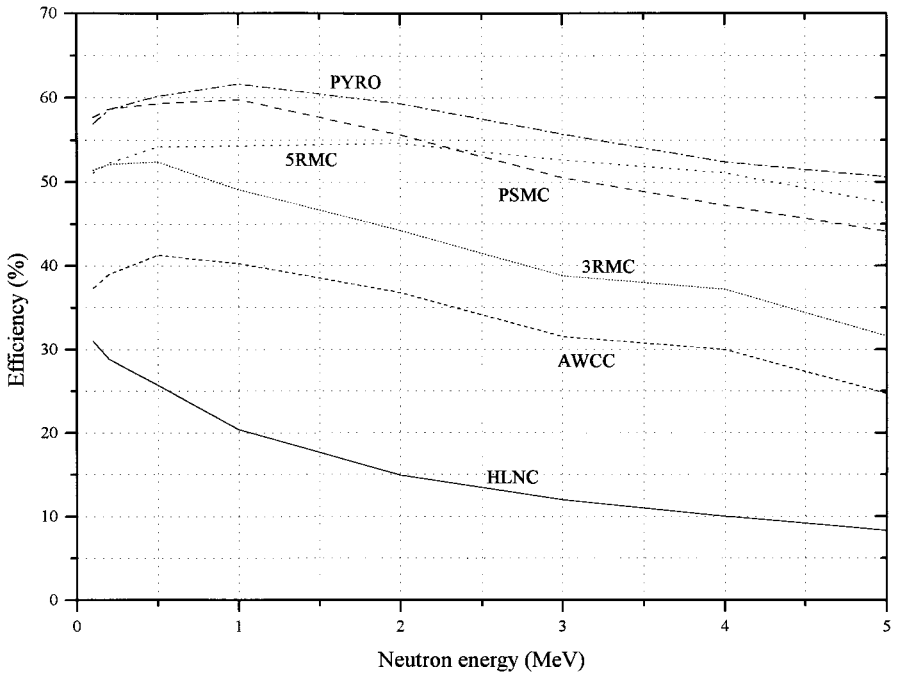


**Fig. 1. Neutron detection efficiency vs neutron energy for six thermal neutron coincidence counters.**

(1) HLNC (High-Level Neutron Coincidence Counter) [7]
This counter has 18 $^{3}$He tubes in one ring. The HLNC has a thin polyethylene body at the vertical center to flatten the efficiency profile and maintain easy portability. Thus, the neutron detection efficiency is low (18%) and the dependence of the efficiency on energy is large.

(2) AWCC (Active Well Coincidence Counter) [8]
The AWCC has 42 $^{3}$He tubes in two rings. Although designed for active neutron assay, it makes a good passive neutron counter if the americium-lithium (AmLi) sources are removed and the polyethylene end

plugs are replaced with graphite plugs. With graphite plugs the efficiency is calculated to be 36% and the efficiency has medium energy dependence.

(3) 3RMC (3-Ring Multiplicity Counter) [9]
The 3RMC is a prototype AWCC with the addition of an extra ring of $^{3}$He tubes and with a change from polyethylene to graphite end plugs. It has 60 $^{3}$He tubes and an efficiency of 45%. Although now called the 3RMC, it was not designed to be a multiplicity counter. The efficiency has medium energy dependence.

(4) PSMC (Plutonium Scrap Multiplicity Counter) [10]
This counter was designed to be used as a multiplicity counter. An essential design goal was to obtain a good compromise between performance and cost. The PSMC is sometimes called a 3 1/2 ring counter; it has four rings, but the outer two rings have only half the usual number of $^{3}$He tubes. This design was chosen to reduce the energy dependence of the efficiency compared to a 3-ring design while maintaining high detection efficiency and moderate cost. The PSMC has 80 $^{3}$He tubes and an efficiency of 55%. The energy dependence of the efficiency is medium to low.

(5) PYRO (Pyrochemical Counter) [11]
The PYRO counter was also designed to be a multiplicity counter. It has 126 $^{3}$He tubes in four complete rings. The efficiency is 58% and the energy dependence of the efficiency is low.

(6) 5RMC (5-Ring Multiplicity Counter) [11]
Originally called the Dual-mode Multiplicity Counter, the 5RMC was designed to be used as a research multiplicity counter. In its present, fixed configuration it has a polyethylene body and graphite end plugs. It has 130 $^{3}$He tubes in five complete rings. The efficiency is 53% and the energy dependence of the efficiency is low.

The energy dependence of the efficiencies is emphasized in Fig. 2, where the efficiencies for each detector have been normalized to the detector efficiency at 2 MeV.

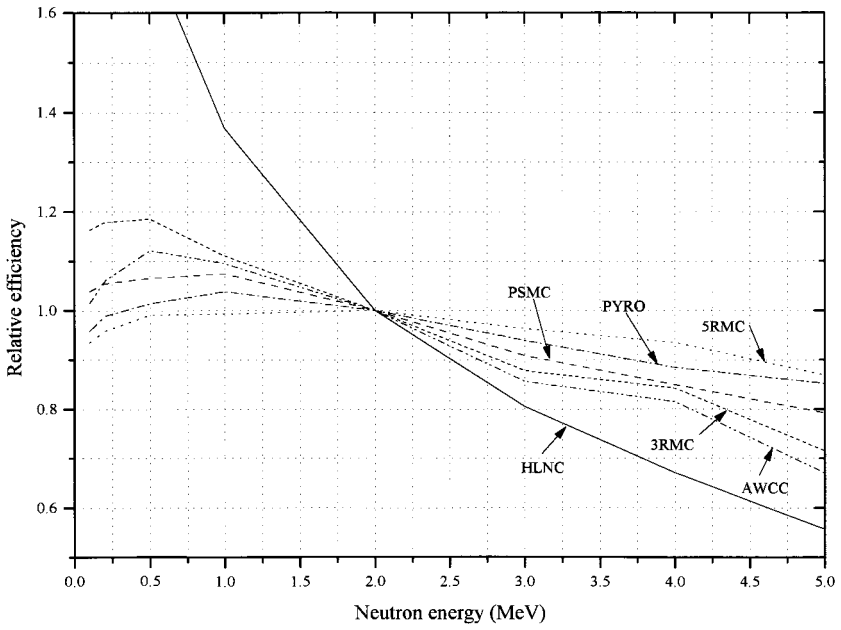


**Fig. 2. Neutron detection efficiency vs neutron energy for six thermal neutron coincidence counters normalized to the 2-MeV efficiency for each counter.**

## 2.2 Induced Fission Multiplicities

The average number of fission neutrons emitted in the neutron-induced fission of $^{239}$Pu (the nubar value) depends on the energy of the neutron inducing the fission. The values for the first, second, and third factorial moments of the fission neutron distribution as a function of neutron energy were taken from Ref. 12. Curves fit to these values are plotted in Fig. 3. For simplicity, only induced fission in $^{239}$Pu is treated in the multiplicity calculations.

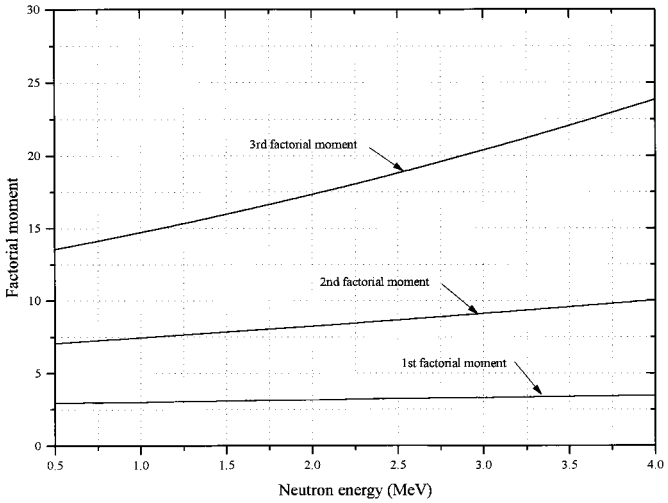


**Fig. 3. Factorial moments vs neutron energy. The factorial moments are the neutron multiplicity distributions from the neutron-induced fission of $^{239}$Pu.**

## 2.3 Induced fission Probabilities

The probability that a neutron will induce a fission in the sample depends on the neutron energy and the characteristics of the sample. The same probability relative to that of fission spectrum neutrons also depends on the neutron energy and the characteristics of the sample. Thus the inclusion of ($\alpha$,n) energies into the model for multiplicity analysis makes the problem sample dependent. To study the sensitivity of the analysis to sample characteristics, Monte Carlo calculations were performed with the MCNP code for 1-kg and 2-kg cans of plutonium oxide in the 3RMC. A 10-cm can diameter and a density of 2 g/cm$^3$ was assumed. The plutonium was 94% $^{239}$Pu and 6% $^{240}$Pu. The probability for a neutron to induce a fission (first fission only) was calculated for energies from 0.5 MeV to 4 MeV for the 1-kg and 2-kg samples. These probabilities relative to the same probabilities for neutrons with a $^{240}$Pu spontaneous fission spectrum are plotted in Fig. 4. The relative values for the 1-kg and 2-kg samples are within 2% to 3% of each other over this energy range, so the sample dependence for plutonium oxide is not large. The 1-kg values were used for the remainder of the multiplicity calculations.

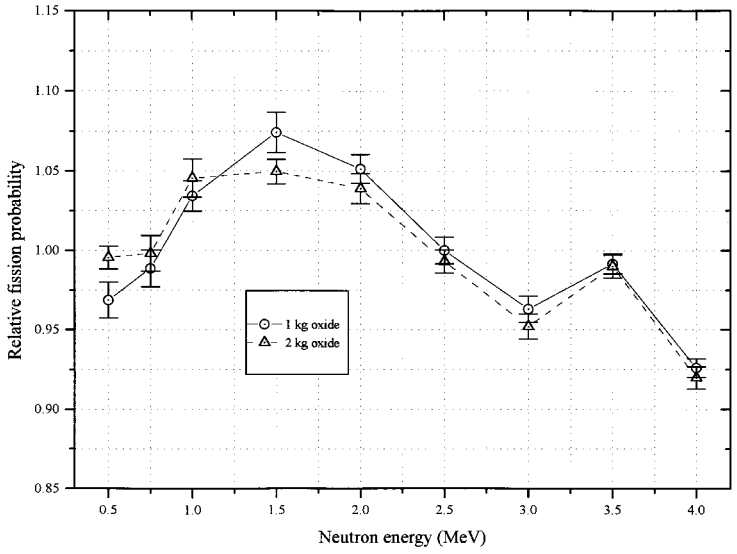


**Fig. 4. Relative fission probability vs neutron energy for 1-kg and 2-kg plutonium oxide samples. The probability is relative to the fission probability for fission spectrum neutrons.**

## 2.4 Energy Dependent Multiplicity Equations

Monoenergetic ($\alpha$,n) neutrons were used for the bias calculations. The multiplicity equations from Ref. 1 were modified so that the following quantities are functions of the ($\alpha$,n) neutron energy:

$\varepsilon_\alpha$ = efficiency for detecting an ($\alpha$,n) neutron,

| | | |
|---|---|---|
| r | = | probability for an (α,n) neutron to induce a fission relative to that for a fission spectrum neutron (first fission only), and |
| $\nu_{i1}^{\alpha}, \nu_{i2}^{\alpha}, \nu_{i3}^{\alpha}$ | = | 1st, 2nd, and 3rd factorial moments of the neutron multiplicity distribution from fission induced in $^{239}$Pu by (α,n) neutrons. |

The revised equations for the singles, doubles, and triples rates are

$$S = mF_0\nu_{s1}\left[\varepsilon M + (1 - rp)\alpha\varepsilon_{\alpha} + rp\alpha\nu_{i1}^{\alpha} M \varepsilon\right] \quad , \tag{1}$$

$$D = \frac{mF_0\varepsilon^2 f_d M^2}{2}\left[x(\nu_{s1}, \nu_{s2}) + rp\alpha\nu_{s1} x(\nu_{i1}^{\alpha}, \nu_{i2}^{\alpha})\right] \quad , \text{ and} \tag{2}$$

$$T = \frac{mF_0\varepsilon^3 f_t M^3}{6}\left[y(\nu_{s1}, \nu_{s2}, \nu_{s3}) + rp\alpha\nu_{s1} y(\nu_{i1}^{\alpha}, \nu_{i2}^{\alpha}, \nu_{i3}^{\alpha})\right] \quad , \tag{3}$$

where

$$x(\nu_1, \nu_2) = \nu_2 + z\nu_1\nu_2,$$

$$y(\nu_1, \nu_2, \nu_3) = \nu_3 + z(3\nu_2\nu_2 + \nu_1\nu_3) + 3z^2\nu_1\nu_2^2,$$

$$z = \frac{M - 1}{\nu_{i1} - 1},$$

$$p = \frac{M - 1}{M\nu_{i1} - 1},$$

| | | |
|---|---|---|
| S, D, T | = | singles, doubles, and triples rates, |
| m | = | effective $^{240}$Pu mass, |
| $F_0$ | = | $^{240}$Pu spontaneous fission rate per unit mass of $^{240}$Pu, |
| α | = | (α,n) to spontaneous fission neutron ratio, |
| M | = | neutron multiplication for fission spectrum neutrons, |
| ε | = | detection efficiency for fission spectrum neutrons, |
| $f_d$, $f_t$ | = | doubles and triples gate fractions, |
| $\nu_{s1}$, $\nu_{s2,}$ $\nu_{s3}$ | = | 1st, 2nd, and 3rd factorial moments of the neutron multiplicity distribution for $^{240}$Pu spontaneous fission, and |
| $\nu_{i1}$, $\nu_{i2,}$ $\nu_{i3}$ | = | 1st, 2nd, and 3rd factorial moments of the neutron multiplicity distribution for fission spectrum induced fission of $^{239}$Pu. |

## 3. BIAS CALCULATIONS

The purpose of the bias calculations is to determine the bias in a multiplicity verification measurement as a function of (α,n) neutron energy if the data are analyzed with the energy-independent multiplicity equations. To calculate the bias for a particular detector and one set of parameters, the singles, doubles, and triples rates are calculated from the energy-dependent equations (Eqs. 1–3) for a specified effective $^{240}$Pu mass ($m_0$), neutron multiplication, alpha, and (α,n) neutron energy; then the count rates are used to solve for effective $^{240}$Pu mass (m) using the energy independent multiplicity equations. The bias, expressed in percent, is $(m - m_0)/m_0 \cdot 100$.

Figure 5 shows the bias vs neutron energy for the six neutron detectors for a multiplication of 1.1 and an alpha of 3 — typical parameters for 1-kg plutonium oxide samples with moderate impurities. Also shown in Fig. 5

is the bias plot for an idealized detector called FLAT, which has constant neutron detection efficiency vs neutron energy. The three detectors designed to be multiplicity counters (5RMC, PYRO, and PSMC) have the lowest bias over most of the energy range from 0.5 MeV to 4 MeV, as expected. The 5RMC is a good approximation to the FLAT counter. The 3RMC, which was not designed to be a multiplicity counter, has a very high relative efficiency at low energies (see Fig. 2) and has a larger bias over most of the energy range than the AWCC, even though the 3RMC has three rings of tubes and the AWCC only has two. The HLNC — with its single ring of tubes and thin polyethylene body — has the largest bias over most of the energy range. The bias of the PSMC, for example, varies from -7% at an ($\alpha$,n) energy of 0.5 MeV to +6% at an ($\alpha$,n) energy of 4 MeV; thus, even for counters designed to be multiplicity counters, the bias can be significant.

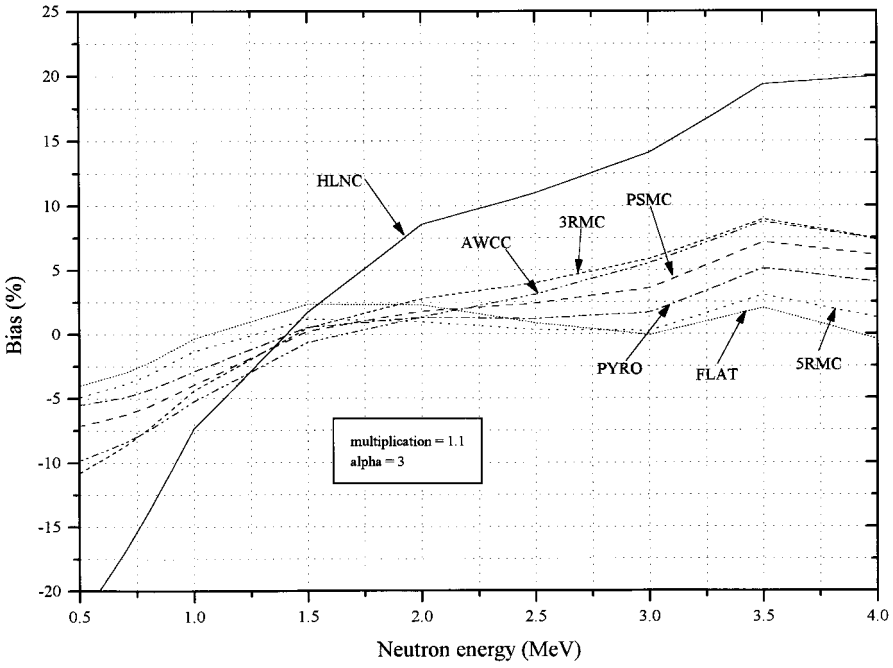


**Fig. 5. Assay bias vs ($\alpha$,n) neutron energy for multiplication = 1.1 and alpha = 3 for six thermal-neutron coincidence counters and for an idealized counter (FLAT) with constant efficiency.**

## 4. RING RATIO ANALYSIS

The ring ratio is the ratio of total neutron count rates in two rings of a detector. The ring ratio is a function of neutron energy, so the measured ring ratio can provide a crude estimate of the average energy of the neutrons from a sample. The energy-dependent multiplicity equations can be solved for multiplication, alpha, ($\alpha$,n) energy, and effective $^{240}$Pu mass from the measured singles, doubles, and triples rates and the measured ring count rates. Figure 6 shows the count rate ratio of the inner to the outer ring in the 3RMC vs neutron energy determined for ($\alpha$,n) neutrons from aluminum, fluorine, silicon, oxygen, magnesium oxide, and boron by measuring a series of small plutonium samples with known impurities; the average ($\alpha$,n) energy for each sample was taken from Ref. 3. The ring ratio for ($\alpha$,n) neutrons from Li was measured using an AmLi source; the average ($\alpha$,n) energy was taken from Ref. 8. A calibration curve of ring ratio vs neutron energy was calculated from the seven data points.

Figure 7 shows the bias vs ($\alpha$,n) neutron energy, estimated from the ring ratios, for a series of measurements of impure plutonium oxide made with the 3RMC and analyzed with energy independent multiplicity analysis.[9] The samples typically had 800 g of plutonium, multiplication values of 1.08 to 1.10, and alpha values of 2 to 10. A weighted, linear least squares fit to the bias has a slope of 5.0 ± 0.4 %/MeV. The average bias is low because most of the ($\alpha$,n) neutron energies are between 1 and 2 MeV (see Fig. 5).

These data were also analyzed with the energy-dependent multiplicity equations (Eqs. 1–3). The (α,n) energy was estimated from the ring ratio curve. The ring ratio for (α,n) neutrons was obtained from the measured inner and outer ring count rates after subtracting the fission neutron contributions to the inner and outer rings. Figure 8 shows the bias vs (α,n) neutron energy for the energy-dependent analysis. A weighted, linear least squares fit to the bias has a slope of 0.3 ± 0.4 %/MeV. Thus, with energy-dependent analysis, the bias no longer shows an energy dependence. This result is better than expected and should not be taken too seriously, because broad (α,n) neutron spectra are represented simply by ring ratios in the energy-dependent analysis. Additional data sets need to be studied.

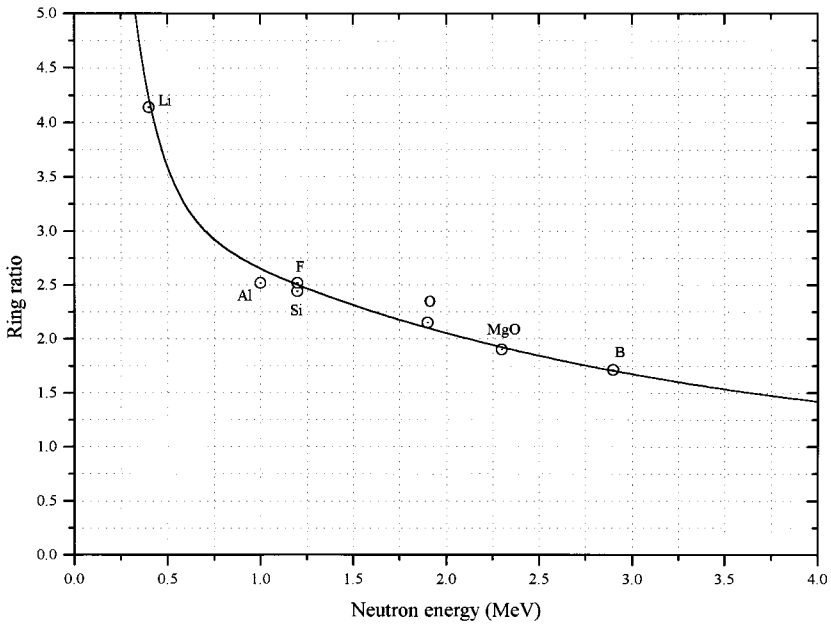


**Fig. 6. Ring ratio vs neutron energy for the 3RMC. Measured values for Li, Al, F, Si, O, MgO, and B are shown.**

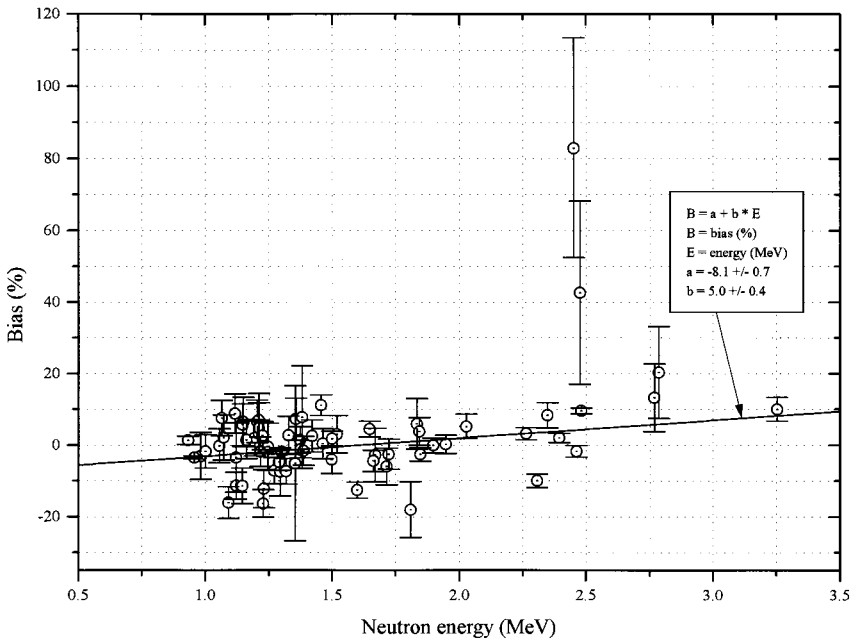


**Fig. 7. Assay bias vs (α,n) neutron energy for multiplicity analysis without using the ring ratio technique. The bias increases with energy with a slope of 5 %/MeV.**

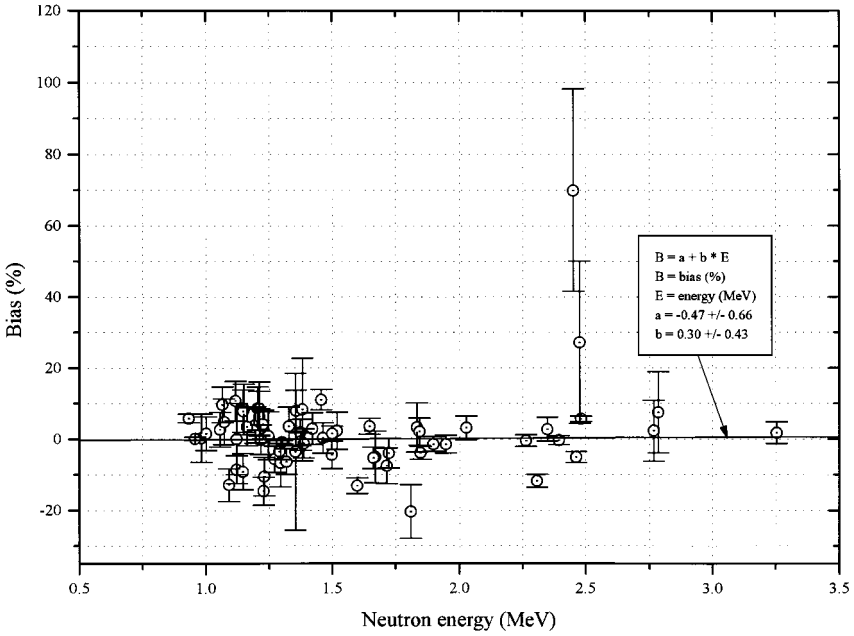


**Fig. 8. Assay bias vs (α,n) neutron energy for multiplicity analysis using the ring ratio technique. The bias shows no energy dependence.**

## 5. COMMENTS AND CONCLUSIONS

The energy of (α,n) neutrons potentially can have a significant effect on the accuracy of verification measurements made with thermal neutron multiplicity counters. Energy-dependent bias increases with increasing multiplication and alpha values. If impure plutonium oxides typically produce (α,n) neutrons with energies in the range of 1 to 2 MeV, then energy-dependent bias should not be a significant problem; in this energy range the bias is not only small, but has positive and negative values that tend to cancel with a broad spectrum (α,n) source (see Fig. 5). Ring ratio measurements and energy-dependent multiplicity analysis should be useful in reducing energy-dependent bias. Measurements of neutron spectra, rather than just ring ratios, should improve the accuracy of energy-dependent multiplicity analysis. The preparation of plutonium oxide impurity standards is planned so that energy-dependent effects can be studied under controlled conditions. The AWCC is expected to perform as accurately as the 3RMC for multiplicity verification measurements; however, longer measurement times are required by the lower AWCC efficiency. Multiplicity measurements with the AWCC are planned for the near future to evaluate its performance.

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FIGURE CAPTIONS

1. Neutron detection efficiency vs neutron energy for six thermal neutron coincidence counters.
2. Neutron detection efficiency vs neutron energy for six thermal neutron coincidence counters normalized to the 2-MeV efficiency for each counter.
3. Factorial moments vs neutron energy. The factorial moments are for the neutron multiplicity distributions from the neutron-induced fission of $^{239}$Pu.
4. Relative fission probability vs neutron energy for 1-kg and 2-kg plutonium oxide samples. The probability is relative to the fission probability for fission spectrum neutrons.
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