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Title: ACTIVE NEUTRON MULTIPLICITY ANALYSIS AND MONTE CARLO CALCULATIONS

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ACTIVE NEUTRON MULTIPLICITY ANALYSIS AND MONTE CARLO CALCULATIONS*

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ABSTRACT

Active neutron multiplicity measurements of high-enrichment uranium metal and oxide samples have been made at Los Alamos and Y-12. The data from the measurements of standards at Los Alamos were analyzed to obtain values for neutron multiplication and source-sample coupling. These results are compared to equivalent results obtained from Monte Carlo calculations. An approximate relationship between coupling and multiplication is derived and used to correct doubles rates for multiplication and coupling. The utility of singles counting for uranium samples is also examined.

INTRODUCTION

Calibration curves for the Active Well Coincidence Counter (AWCC)[1] are normally obtained by counting uranium standards and plotting coincidence rate vs ^{235}U mass. Because of neutron absorption and multiplication in the uranium samples, the calibration curves are nonlinear and are sensitive to the geometry and ^{235}U density of the samples. For accurate assays, therefore, the assayed samples must be very similar to the standards used to obtain the calibration curve. However, an appropriate calibration curve is not always available—either because suitable standards are not available or because the characteristics of the assay samples are not well known. The purpose of this study is to explore the possibilities of using multiplicity counting and Monte Carlo calculations to improve the assays of samples for which suitable calibration curves are not available.

MULTIPLICITY COUNTING

Passive neutron multiplicity counting[2] was developed for the assay of impure plutonium samples because the impurities in the samples produce excess coincidence counts through fissions induced by neutrons created by (alpha, n) reactions with the impurities. If the amount and type of impurity is not known, then there is no appropriate calibration curve to use for accurate assays with conventional coincidence counting. The multiplicity counting technique has worked well for some material categories because the multiplicity measurement provides a third quantity that can be used to solve for the (alpha, n) rate. The situation for active multiplicity counting, however, is much more difficult because it involves the coupling of the AmLi source neutrons to the assay sample. The coupling is defined as the average number of fissions induced by an AmLi neutron per gram of ^{235}U in the sample; the coupling depends on the sample's geometry, ^{235}U density, chemical and isotopic composition, and location in the detector. Previous work on active multiplicity counting is described in Ref. 3.

In active and passive multiplicity counting, three equations relate the singles, doubles, and triples count rates to three unknown quantities; the singles and doubles rates are the same as the totals and real coincidence rates in conventional coincidence counting. The equations for active multiplicity counting are[3]

$$S = S_0 + B + S_s + YCmM\varepsilon\nu_{s1} \tag{1}$$

$$D = \frac{YCmf_d\varepsilon^2\nu_{s2}}{2} \bullet c_d \tag{2}$$

$$T = \frac{YCmf_t\varepsilon^3\nu_{s3}}{6} \bullet c_t \tag{3}$$

where

$$c_d = M^2\left[1 + c_1(M-1)\right] \tag{4}$$

$$c_t = M^3\left[1 + c_2(M-1) + c_3(M-1)^2\right]$$

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S, D, T	=	singles, doubles, and triples rates
S_0	=	singles rate from the AmLi sources without a sample
B	=	background singles rate
S_S	=	change to S_0 due to scattering and absorption of AmLi neutrons by the sample
Y	=	AmLi source yield (neutrons/s)
C	=	coupling
m	=	^{235}U mass
M	=	neutron multiplication
$\nu_{s1}, \nu_{s2}, \nu_{s3}$	=	1st, 2nd, and 3rd moments of the first-fission neutron distributions
c_1, c_2, c_3	=	nuclear data constants
fd, ft	=	doubles and triples gate fractions
ε	=	neutron detection efficiency
c_d, c_t	=	doubles and triples multiplication correction factors.

Equations (2) and (3) determine the neutron multiplication M and the product of coupling and ^{235}U mass (Cm) from the measured doubles and triples rates and from the known detector parameters; Eq. (1) can then be used to determine the singles perturbation term S_S. If S_S can be neglected relative to S_0, for samples with high ^{235}U mass and high multiplication, M and Cm can also be obtained from Eqs. (1) and (2), that is, from conventional coincidence counting data.

Because C and m always appear as the product Cm, C and m cannot be obtained separately from coincidence or multiplicity counting—additional experimental data or calculations are required. There is no known way to measure the coupling, so we studied the problem with calculational procedures.

MONTE CARLO CALCULATIONS

We modeled the AWCC and various samples with the MCNP code[4] to obtain the neutron multiplication and coupling for a wide range of ^{235}U masses and multiplications. There were two goals. The first was to see whether calibration curves could be generated computationally when the samples are well characterized but standards are not available. The second was to see whether computations can be used to assist in the assay of samples with variable geometry.

MEASUREMENTS

For comparison with the Monte Carlo calculations, a series of HEU metal standards and a series of oxide standards were measured in an AWCC at Los Alamos. The metal standards consisted of stacks of disks; the ^{235}U masses ranged from 246 g to 3686 g and the diameters were either 6 cm or 7 cm. The oxide standards were cans of powder with diameters of 10 cm; the ^{235}U masses ranged from 117 g to 903 g with varying enrichments such that the uranium mass was always about 1 kg. Multiplicity circuitry was used so that the triples rate would be available to determine an experimental value for the neutron multiplication from the triples/doubles ratio (Eqs. 2 and 3). The AWCC was configured in its most compact fast mode (8-in. cavity height, cadmium lined) with the nickel reflector and polyethylene shield rings in place. The samples were centered on a 2-in.-high lab jack. The doubles rate vs ^{235}U mass for the oxide and metal standards is shown in Fig. 1; note the sensitivity of the shape and size of the calibration curves to the material type.

To study the assay of samples with variable geometry, we used data from Y-12 for an assortment of samples for which no calibration curve is available. These samples were mostly metals in assorted geometries with good book values for their ^{235}U masses. Details about the samples are shown in Table I. The AWCC was configured in fast mode with a 17-in. cavity height to accommodate tall sample cans; the nickel reflector and the polyethylene shield rings were not used. The doubles rate vs ^{235}U mass for this group is shown in Fig. 2.

EXPERIMENTAL MULTIPLICATION AND COUPLING

From the Los Alamos measurements of the metal standards we calculated the multiplication using Eqs. (2) and (3); then we calculated the multiplication correction factor from Eq. (4) and, using the known ^{235}U mass, we

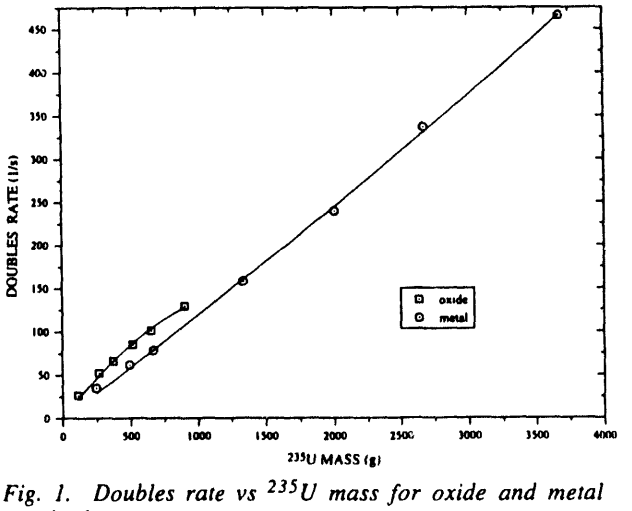

Fig. 1. Doubles rate vs ^{235}U mass for oxide and metal standards.

Table I. Y-12 Miscellaneous Samples			
Sample	^{235}U Mass (g)	Form	Description
1	6 241	metal	4 pieces
2	8 007	oxide	powder
3	10 934	metal	broken button
4	14 087	metal	5 pieces
5	16 252	metal	2 cylinders
6	16 294	metal	chunks
7	16 298	metal	plates
8	16 298	metal	1 piece

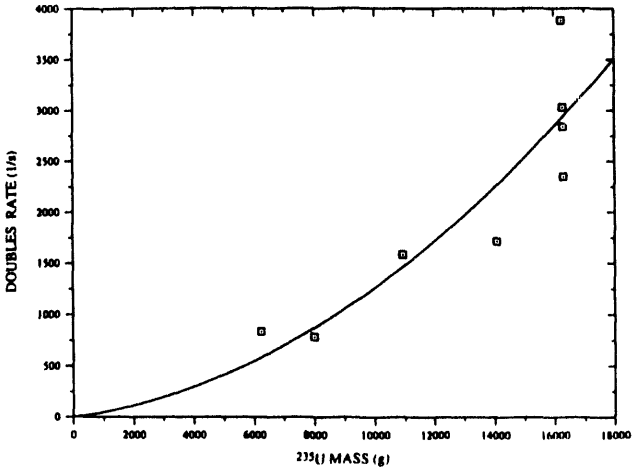

Fig. 2. Doubles rate vs ^{235}U mass for Y-12 miscellaneous samples.

calculated the coupling from Eq. (2). Figure 3 shows the doubles rate, coupling, and doubles multiplication correction factor (c_d) plotted vs the ^{235}U mass for the metal standards. This figure illustrates the compensation that occurs between multiplication and coupling. Over the mass range of the standards, the multiplication correction factor increases by about a factor of 3, whereas the coupling decreases by about a factor of 3 as a result of sample self-shielding. The product of the coupling and the multiplication correction factor is insensitive to the ^{235}U mass, so the doubles rate is almost linear with ^{235}U mass.

MONTE CARLO AND EXPERIMENTAL COMPARISON

For the Los Alamos metal and oxide standards, the experimental and Monte Carlo results for the multiplication are shown in Fig. 4. The experimental values come directly from Eqs. (2) and (3). The multiplication values

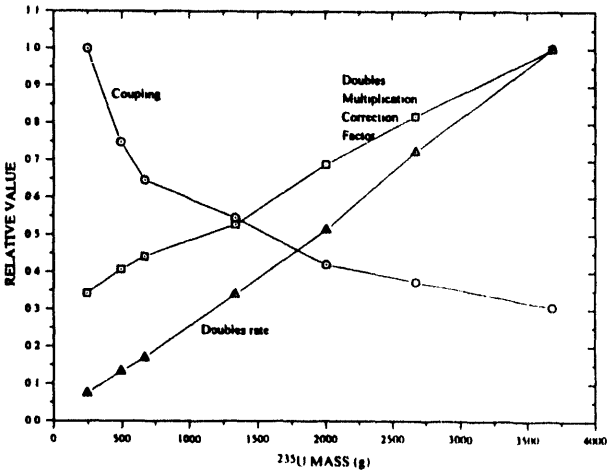

Fig. 3. Doubles rate, coupling, and doubles multiplication correction factor vs ^{235}U mass for metal standards.

agree within statistical errors. A typical random error (1 standard deviation) for the measured multiplication is 1% and for the Monte Carlo multiplication is 2%. The experimental and Monte Carlo results for the coupling are shown in Fig. 5. The experimental values come from the measured multiplications, the known masses, and Eq. (2). The Monte Carlo results for coupling were consistently biased relative to the experimental values, so they were normalized to force agreement with the experimental results averaged over all samples. With the exception of one of the metal standards, the relative coupling values agree within statistical errors. A typical random error (1 standard deviation) is 2% for the Monte Carlo value and 3% for the experimental value. Note in Fig. 5 that the measured coupling is higher for oxides and lower for metals than the Monte Carlo values. Thus, if a metal calibration curve is used as a reference, then an oxide calibration curve derived from Monte Carlo calculations would be biased by about 6%. All of these calculations are preliminary, so better results might be obtained with improved modeling.

COUPLING vs MULTIPLICATION

As the ^{235}U mass or the ^{235}U density of a sample increases, the neutron multiplication increases and the coupling decreases, so it is interesting to study coupling vs multiplication. If coupling could be expressed in terms of multiplication and ^{235}U mass [$C = C(m, M)$], then Eqs. (2) and (3) could be used to determine the ^{235}U mass. Monte Carlo calculations were performed for the Los Alamos AWCC and ^{235}U samples as follows. All samples were in 4-in.-diam by 4-in.-high cylinders. The samples were centered radially in the AWCC 2 in. above the bottom AmLi source. The enrichment was always 93%. For each ^{235}U mass the density was either maximum (a 4-in.-diam metal cylinder at the bottom of the

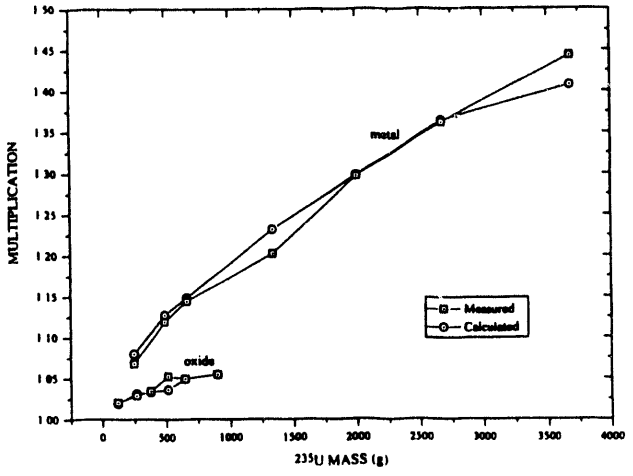

Fig. 4. Multiplication vs ^{235}U mass for Los Alamos oxide and metal standards.

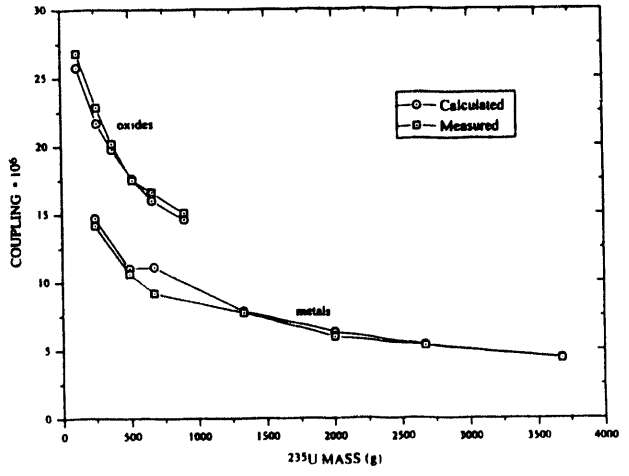

Fig. 5. Measured and calculated coupling vs ^{235}U mass for metal and oxide standards.

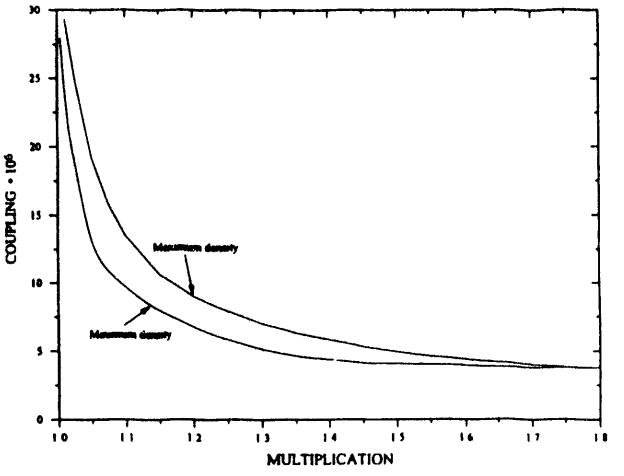

Fig. 6. Multiplication vs coupling for high-density and low-density samples.

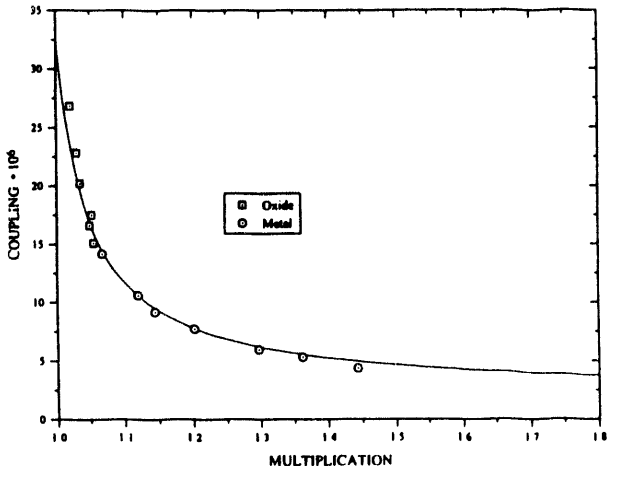

Fig. 7. Coupling vs multiplication for oxide and metal standards.

can) or minimum (the ^{235}U spread uniformly throughout the can). Figure 6 shows coupling vs multiplication for maximum and minimum densities. Because these two curves are similar, the multiplication gives a rough estimate of the coupling for any mass and density. The average of these two curves is shown in Fig. 7 with the experimental values of coupling and multiplication for the Los Alamos metal and oxide standards.

The curve in Fig. 7 can be used as an approximate Monte Carlo calibration curve of coupling vs multiplication. With additional Monte Carlo calculations, this curve could be extended from the form $C = C(M)$ to the more accurate and useful form $C = C(m, M)$.

MULTIPLICITY ANALYSIS OF Y-12 SAMPLE DATA

The coupling calibration curve from the last section was applied to the assay of the samples in the Y-12 data set, although the AWCC configuration for these measurements is different from the Monte Carlo model and the samples are not well represented by homogeneous ^{235}U distributions in 4-in.-diam by 4-in.-high containers. The measured triples/doubles ratio was used to obtain the multiplication from Eqs. (2) and (3). The multiplication correction factor was calculated from Eq. (4) and the coupling from the calibration curve in Fig. 7. The measured doubles rate was then divided by the multiplication correction factor and the coupling. The corrected doubles rate should be a linear function of ^{235}U mass according to Eq (2). Figure 8 shows the doubles rates for the Y-12

samples before and after correction for multiplication and coupling. The doubles rates after correction are normalized for plotting on the same scale with the uncorrected rates.

PRECISION OF MULTIPLICITY ANALYSIS

If the assay of ^{235}U mass is based on a calibration curve of doubles rate (corrected for multiplication and coupling) vs ^{235}U mass, then the assay precision for metals and oxides vs ^{235}U mass is shown in Fig. 9. These curves were derived from observed errors in the triples/doubles ratio for the Los Alamos metal and oxide standards.

NET SINGLES COUNTING

In general, singles counting (or totals counting) is risky because of neutron scattering and absorption by the sample and because of background fluctuations. However, if the ^{235}U mass is large, singles counting can be useful. Figure 10 shows calibration curves of net singles rate vs ^{235}U mass for the Los Alamos oxide and metal standards. The singles rate without a sample is 4555 counts/s. Note that a 1% change in the singles rate corresponds to about a 25% change in the ^{235}U mass for the smallest metal standard.

If the sample scattering and absorption term S_S in Eq. (1) can be neglected, then Eqs. (1) and (2) can be used to calculate the multiplication. The multiplication was calculated from Eqs. (1) and (2) with $S_S = 0$ for the metal and oxide standards. The metal results are shown in Fig. 11 together with the results from Monte Carlo calculations and from the triples/doubles ratio. Except for the smallest standards, the multiplication values agree within

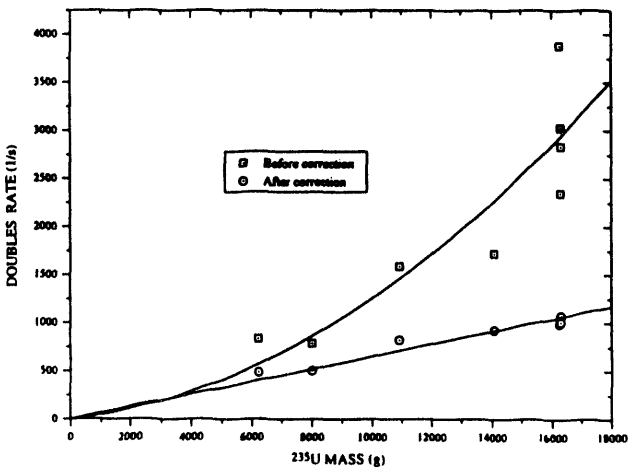

Fig. 8. Doubles Rates vs ^{235}U mass before and after correction for multiplication and coupling.

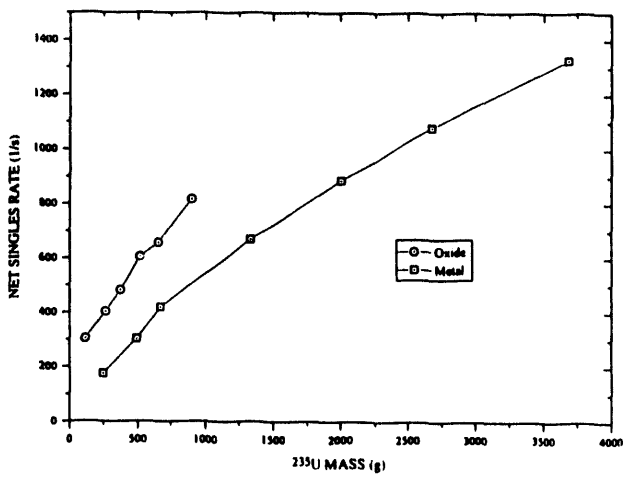

Fig. 10. Net singles rate vs ^{235}U mass for oxide and metal standards.

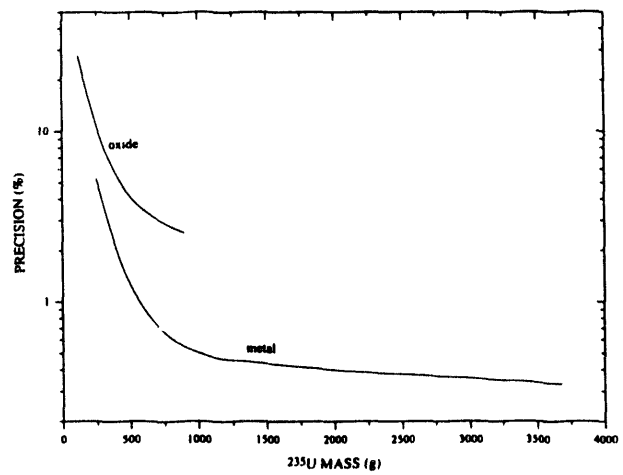

Fig. 9. Precision of assay ^{235}U mass vs ^{235}U mass from counting statistics only (1 hour measurement).

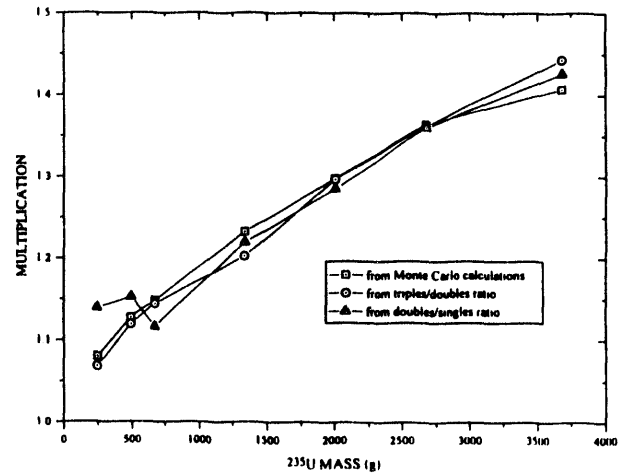

Fig. 11. Multiplication vs ^{235}U mass for metal standards.

a few percent. The oxide results are shown in Fig. 12 together with the results from Monte Carlo calculations and from the triples/doubles ratio. The results using Eqs. (1) and (2) are poor for the smallest standards because the assumption that $S_S = 0$ is inadequate for the small ^{235}U masses.

For large metal samples the net singles rate ($S - S_0 - B$) is large compared to S_S, so from Eq. (1) it is nearly proportional to CMm. The product CM is plotted vs M in Fig. 13, where $C(M)$ is taken as the approximate coupling calibration curve discussed above. Note that the curve of CM vs M has a small slope for $M > 1.2$, so a plot of net singles rate vs ^{235}U mass should be nearly linear for highly multiplying samples. Figure 14 shows the net singles rate vs the ^{235}U mass for the set of Y-12 samples, most of which have high multiplication. The straight line shown in Fig. 14 is a least-squares fit to the data. Because CM is nearly constant, the net singles rate is insensitive to multiplication; the scatter of the net singles rates about the fitted line is comparable to the scatter of the corrected doubles rates shown in Fig. 8.

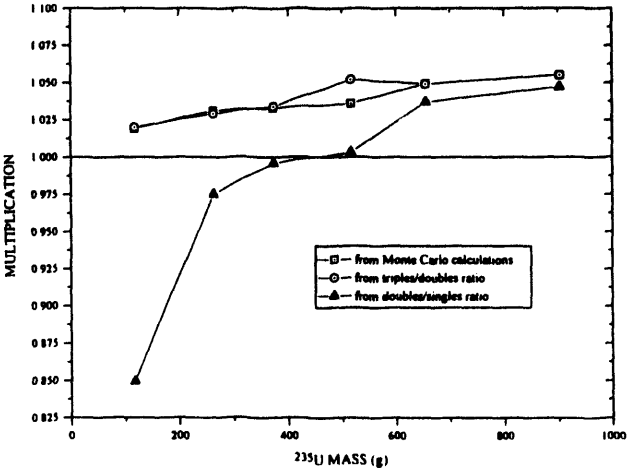

Fig. 12. Multiplication vs ^{235}U mass for oxide standards.

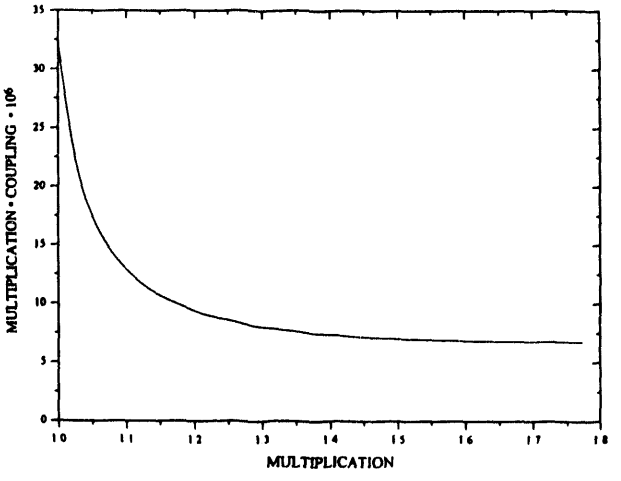

Fig. 13. Multiplication • coupling vs multiplication for average Monte Carlo coupling curve.

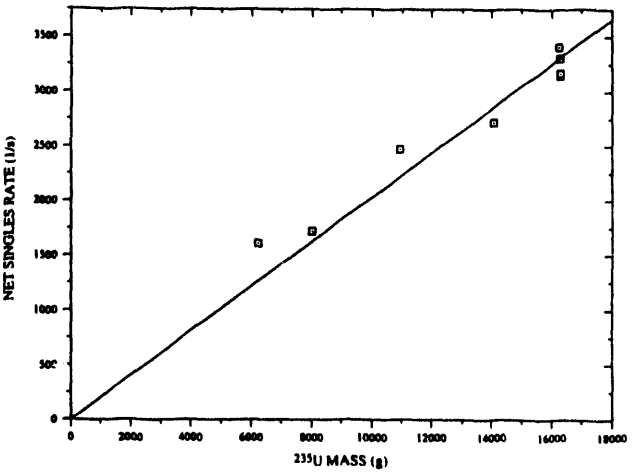

Fig. 14. Net singles rate vs ^{235}U mass for Y-12 miscellaneous samples.

CONCLUSIONS

When calibration standards are not available, Monte Carlo calculations can be used to produce approximate AWCC calibration curves for the assay of uranium samples with known characteristics. For samples with variable geometry and density, multiplicity counting can be used to improve assay accuracy by correcting for variable multiplication and coupling. For high-mass uranium metal samples, net singles counting is a useful assay technique if varying background is not a problem.

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