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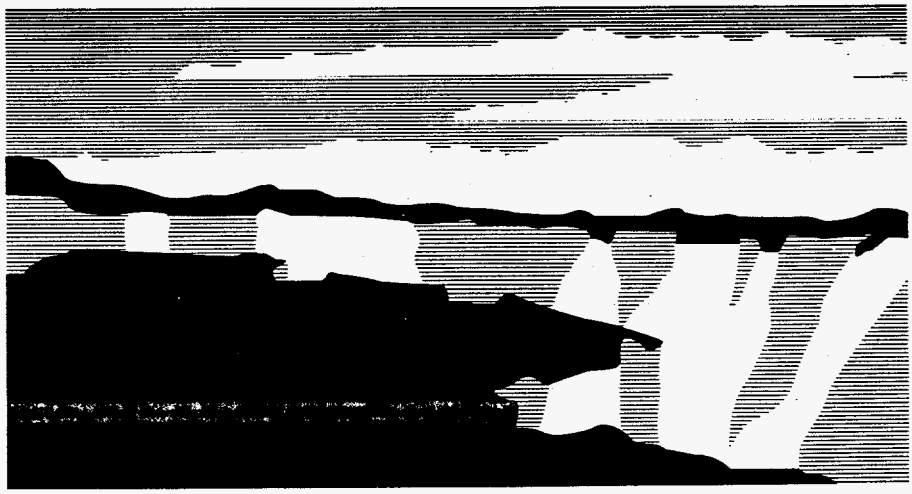

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ANALYSIS OF ACTIVE NEUTRON MULTIPLICITY DATA FOR Y-12 SKULL OXIDE SAMPLES*

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ABSTRACT

Previous work on active neutron multiplicity measurements and analyses is summarized. New active multiplicity measurements are described for samples of Y-12 skull oxide using an Active Well Coincidence Counter and MSR4 multiplicity electronics. Neutron multiplication values for the samples were determined from triples/doubles ratios. Neutron multiplication values were also obtained from Monte Carlo calculations using the MCNP code and the results compared with the experimental values. A calibration curve of AmLi source-sample coupling vs neutron multiplication was determined and used for active multiplicity assay of the skull oxides. The results are compared with those obtained from assay with the conventional calibration-curve technique, where the doubles rate is calibrated vs the ^{235}U mass. The coupling-multiplication relationship determined for the skull oxides is compared with that determined earlier for pure high-enrichment uranium metal and pure uranium oxide. Conclusions are drawn about the application of active multiplicity techniques to uranium assay. Additional active multiplicity measurements and calculations are recommended.

INTRODUCTION

Passive neutron multiplicity counting has become a standard nondestructive analysis technique for the assay of impure plutonium samples or plutonium samples whose characteristics are either not well known or whose characteristics cannot be assumed. The measured singles, doubles, and triples count rates from a sample are used to solve for neutron multiplication, (alpha, n) neutron yield, and effective ^{240}Pu mass. The technique does not require a calibration curve; only detector parameters and nuclear fission parameters are needed. An introduction to the technique can be found in Ref. 1, which contains numerous references to the literature.

Active neutron multiplicity counting[2-5] has a similar motivation for uranium assay as passive multiplicity counting has for plutonium assay. Because calibration curves for uranium assay with the Active Well Coincidence Counter (AWCC)[6] and similar instruments are sensitive to the enrichment, density, and material composition of the samples, assay samples must match calibration standards closely to produce good assay results. This presents a problem for the assay of uranium samples for which a suitable calibration curve is not available or for which the sample characteristics are either not well known or cannot be assumed.

An active multiplicity technique would be very useful if the measured singles, doubles, and triples count rates could be solved for neutron multiplication (M), source-sample coupling (C), and ^{235}U mass (m); coupling is defined as the number of ^{235}U fissions induced by AmLi source neutrons per AmLi neutron per gram of ^{235}U. There are two complications, however. First, the coupling and ^{235}U mass always appear in the multiplicity equations as the product Cm,[5] so the multiplicity equations cannot determine the ^{235}U mass until the coupling is known. Second, the singles rate is generally not useful because it depends on the scattering of the AmLi source neutrons off of the sample; the amount of

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scattering depends on the sample and is usually not negligible.

The ratio of the triples-to-doubles count rates produces an equation for the neutron multiplication because the product *Cm* cancels in the ratio; thus, the multiplication can be determined without knowing the coupling.

One possibility for determining the coupling is to create a calibration curve of coupling vs multiplication. The triples/doubles ratio determines the multiplication and then the multiplication determines the coupling from the calibration curve. Once the coupling is known, the multiplicity equations determine the ^{235}U mass. A calibration curve of coupling vs multiplication is insensitive to ^{235}U mass and density.[5]

Active multiplicity measurements of pure, high-enrichment uranium (HEU) metal and pure, enriched uranium oxide are discussed in Ref. 5. Active multiplicity measurements of HEU metal pieces at Savannah River and Y-12 are discussed in Ref. 4. The present work concerns the measurement of skull oxides (impure oxides) at Y-12 with the AWCC and the application of active multiplicity analysis to the data.

SAMPLES

Seven skull oxide (impure U_3O_8) samples were measured. These contained 55% to 84% uranium by weight and contained erbium—a strong thermal-neutron absorber. The uranium enrichment was 93.15% and the can diameter was 6 inches for all samples. Detailed information on each sample is shown in Table I. The fill heights were determined with a segmented gamma scanner and the uranium masses were determined by modified Davies-Gray titration.

EXPERIMENTAL SETUP

The measurements were performed in a standard AWCC operated in the fast mode with a cavity height of 14 inches. The polyethylene rings for the end plugs and the nickel reflector were not used. Because the impure oxides contained erbium, an erbium liner was used in the sample cavity to reduce the sensitivity of the measurements to the erbium impurity.

For all measurements the samples were centered in the AWCC and placed on a spacer such that the bottoms of the cans were 2 inches above the bottom end plug.

An MSR4 multiplicity shift register[7] was used to collect the multiplicity data using an IBM-type personal computer running the MULTI multiplicity code, which was a forerunner of the current Windows NCC code.[8] The predelay was set to 3 μs and the gate was set to 64 μs.

MEASUREMENT RESULTS

The room background and AmLi doubles and triples count rates were negligible. Each sample was measured for 1000 s (10 runs of 100 s each). The doubles count rates and the triples/doubles ratios are shown in Table II. The counting-

Table I

Skull Oxide Sample Data

Sample number	Net weight (g)	Weight fraction uranium	Uranium (g)	^{235}U (g)	Fill height (cm)	^{235}U density (g/cm^3)
1	5296	0.6750	3575	3330	9.47	1.928
2	10756	0.5548	5967	5558	22.95	1.328
3	9568	0.6759	6467	6024	16.75	1.972
4	9609	0.7210	6928	6453	19.24	1.839
5	10770	0.8310	8950	8337	18.70	2.444
6	12116	0.7861	9524	8872	21.17	2.297
7	12706	0.8377	10644	9915	24.41	2.227

Table II			
Skull Oxide Measurement Data			
Sample number	^{235}U mass (g)	Doubles rate (1/s)	Triples/Doubles ratio
1	3330	358	0.189
2	5558	516	0.201
3	6024	709	0.267
4	6453	568	0.221
5	8337	679	0.266
6	8872	812	0.272
7	9915	915	0.282

statistics standard deviations for the doubles rates are about 1% and for the triples/doubles ratios are about 2%.

These errors correspond to ^{235}U assay mass errors (standard deviations) of about 2% from counting statistics.

MONTE CARLO CALCULATIONS

Because the samples had known masses, fill heights, enrichment, and diameter, and because the samples were known to be uniform, it was possible to calculate the neutron multiplication for each sample and to compare the result with that obtained from the triples/doubles ratio. The multiplication of each sample was calculated using the MCNP Monte Carlo code.[9] The comparison is shown in Table III and Fig. 1. The standard deviations for both the measured and calculated multiplication values are typically 0.006. Good agreement was found for all samples except number 3. For this sample the disagreement was so large that the result was rejected as an outlier pending additional measurements of the sample. For all other calculations and comparisons, the remaining six samples were used.

The Monte Carlo calculations also give the coupling for each sample. The coupling is plotted vs the multiplication in Fig. 2 with a least-squares fit to the calculated points. The coupling increases rapidly as the multiplication decreases; this is a result of the increasing sample penetration by the source neutrons. Note that the calculated points do not lie on a smooth curve; this is because the samples differ in both mass and density.

Table III			
Measured and Calculated Neutron Multiplications			
Sample number	Measured multiplication	Calculated multiplication	Ratio (meas./calc.)
1	1.094	1.095	0.999
2	1.106	1.102	1.004
3	1.168	1.130	1.034
4	1.125	1.136	0.990
5	1.167	1.171	0.997
6	1.173	1.168	1.004
7	1.182	1.180	1.002

The Multiplication Correction Factor

The doubles rate (D) is given by[5]

$$D = kCmc_d, \qquad (1)$$

where k is a constant, C is the coupling, m is the ^{235}U mass, and c_d is the doubles multiplication correction factor. The multiplication correction factor accounts for the increase in the doubles count rate as a result of multiplication and is given by[2]

$$c_d = M^2\left[1+\frac{\nu_{s1}\nu_{f2}}{\nu_{s2}(\nu_{f1}-1)}(M-1)\right], \qquad (2)$$

where ν_{s1} and ν_{s2} are the first and second factorial moments of the multiplicity distribution of neutrons from the fission of ^{235}U induced by low-energy neutrons and where ν_{f1} and ν_{f2} are the same moments for fissions induced by fission-spectrum neutrons.

A plot of the multiplication correction factor vs the neutron multiplication is shown in Fig. 3.

Combined Multiplication and Coupling Effects

The product of the doubles multiplication correction factor and the coupling is plotted vs the multiplication in Fig. 4. If this product were constant, the calibration curve of doubles rate vs ^{235}U mass would be linear. The multiplication correction factor and the coupling tend to compensate each other as the multiplication changes, as Fig. 4 shows. For multiplication values between about 1.11 and 1.19, a linear calibration curve will work fairly well.

Calibration-Curve Analysis

The doubles rates are plotted vs the ^{235}U masses in Fig. 5. A linear calibration curve was determined by least-squares fitting of the six data points to a straight line passing through the origin. The deviations of the points about the calibration curve are shown in Fig. 6, expressed as percent mass deviations; these points are labeled "conventional analysis." The root-mean-square (rms) deviation about the calibration curve is 9.1%. The counting-statistics standard deviations of the doubles rates are approximately 1%.

Multiplicity Analysis

A multiplicity analysis was performed as follows. The measured triples/doubles ratio was used to determine the neutron multiplication. The multiplication correction factor was calculated from the multiplication using Eq. (2). The coupling was calculated from the calibration curve of coupling vs multiplication (Fig. 2). The assay ^{235}U mass was then determined from Eq. (1), using a value for k such that the assays agree on average with the known masses. The deviations of the assays from the known values are plotted in Fig. 6 and are expressed as percent mass deviations. The rms deviation about the average is 4.4%. The assay-mass standard deviation from counting statistics is approximately 2%, so the dominant error is from the estimate of the coupling from the coupling calibration curve.

Coupling vs Multiplication

A calibration curve of coupling vs multiplication was determined for pure HEU metal and pure, enriched uranium oxide in an earlier experiment.[5] The result is shown in Fig. 7. The calibration curve was obtained by least-squares fitting to the pure metal and oxide data points. Also plotted on the graph are the six data points for the skull oxide, normalized as a group to the calibration curve. For the measurements of the pure oxide and metal samples, the AWCC was configured in its standard fast mode—i.e., it had a cavity height of 8 inches and had the polyethylene rings and nickel reflector in place. The absolute values for the couplings are thus much different than for the present experiment with the skull oxides. The relative values of coupling vs multiplication, however, are very similar.

Monte Carlo calculations were used to obtain the coupling vs multiplication for cans of oxide with various masses and densities. The mass range was 2.5 kg to 10 kg and the ^{235}U density range was 1.25 g/cm^3 to 2.5 g/cm^3; this spans the variety of skull oxide samples. The results are shown in Fig. 7, where the coupling is plotted for the highest and lowest densities for ^{235}U masses of 2.5, 5, 7.5, and 10 kg; the coupling values were normalized as a

group to the calibration curve. Such Monte Carlo calculations can be used to extend the calibration curve of coupling vs multiplication [$C = C(M)$] to a calibration curve of coupling vs multiplication and ^{235}U mass [$C = C(M,m)$]. This should improve the estimate of the coupling for multiplicity assays using Eq. (1).

CONCLUSIONS

From the present and previous work on active multiplicity measurements, the following conclusions can be drawn:

The triples/doubles ratio provides a good measure of the neutron multiplication. The neutron multiplication is required to perform active multiplicity assays and is valuable by itself to authenticate uranium samples.

Active multiplicity is effective for the assay of irregular, high-mass pieces of HEU metal. Uniform metal pieces, such as the 18-kg Y-12 cylindrical ingots, are best assayed with conventional coincidence counting and a calibration curve of doubles rate vs ^{235}U mass.

Active multiplicity is potentially useful for the assay of impure uranium oxides, such as the Y-12 skull oxides. For the small data set available from the present experiment, the rms deviation of the assay masses from the known masses was reduced from 9.1% for conventional assay to 4.4% for multiplicity assay. A much larger data set is needed to draw a conclusion.

The use of calibration curves of coupling vs multiplication looks promising for the assay of uranium samples whose detailed characteristics either are not known or cannot be assumed. Additional Monte Carlo calculations are needed to study coupling vs multiplication for various densities and geometries; one case of practical importance is the calculation of coupling vs multiplication for cans of HEU oxide mixed with varying amounts of matrix materials.

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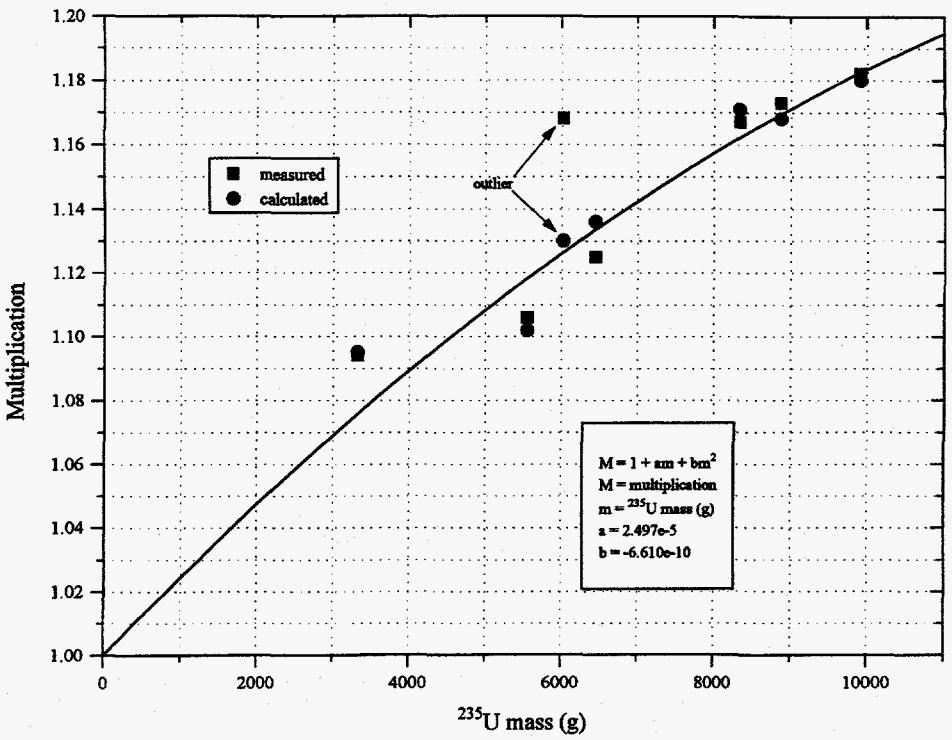

Fig. 1. Measured and calculated neutron multiplication vs ^{235}U mass.

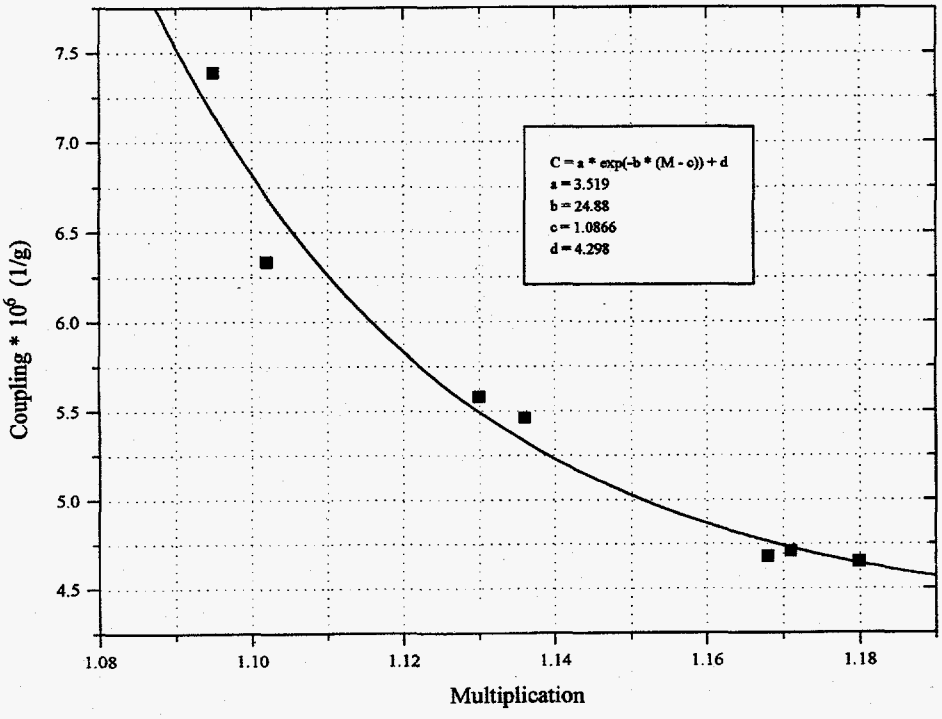

Fig. 2. Coupling vs multiplication for the skull oxides from Monte Carlo calculations.

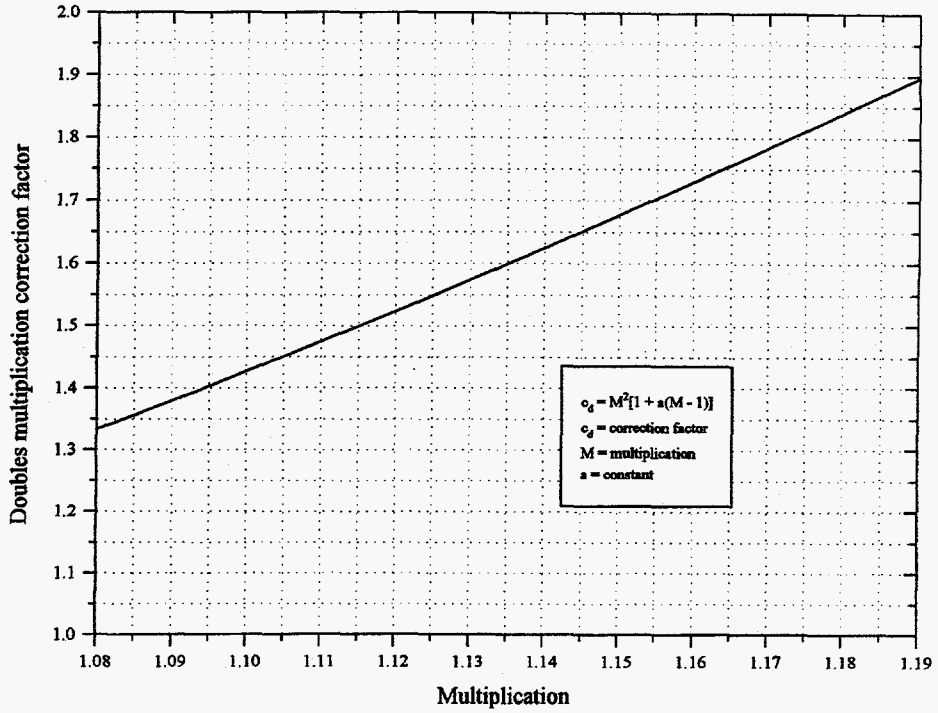

Fig. 3. Multiplication correction factor for the doubles rate vs multiplication calculated from Eq. (2).

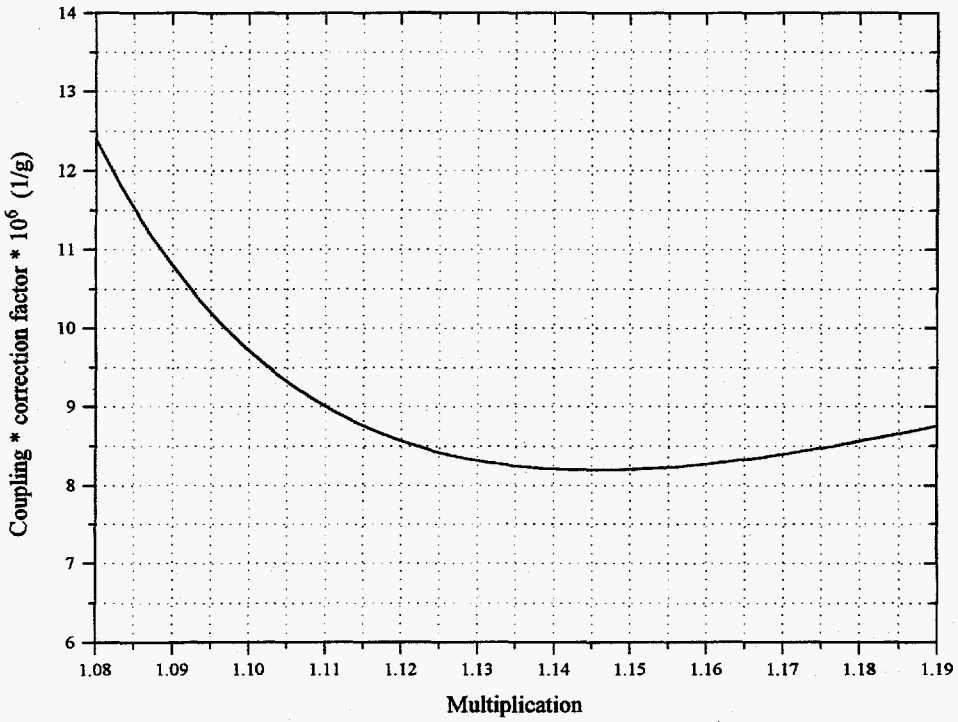

Fig. 4. Product of the coupling and the multiplication correction factor vs multiplication.

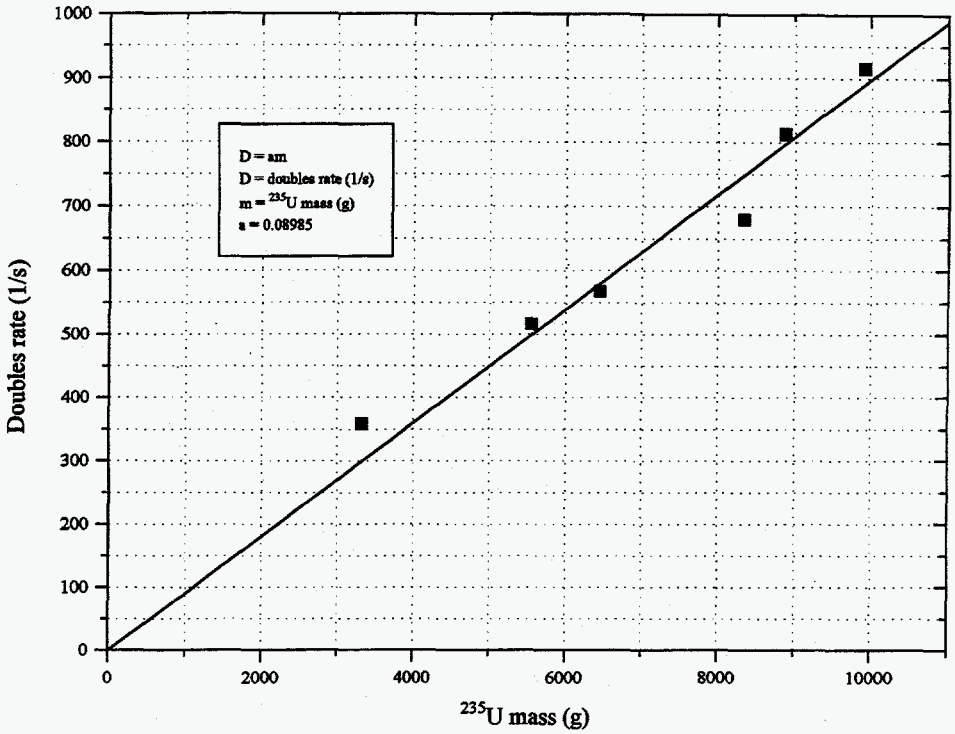

Fig. 5. Doubles rate for the skull oxide samples vs ^{235}U mass.

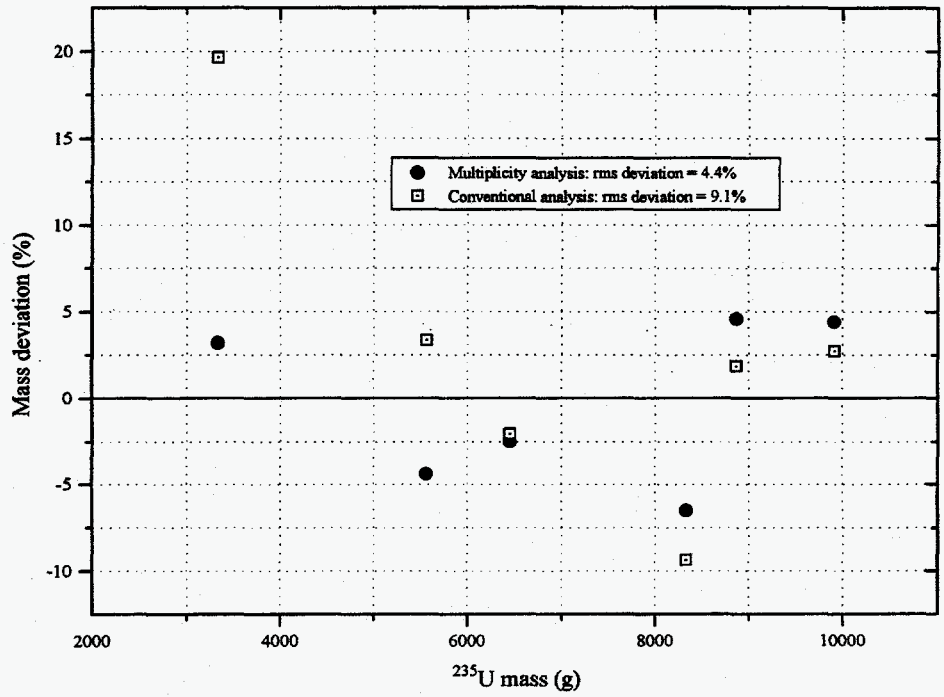

Fig. 6. Mass deviation of ^{235}U vs ^{235}U mass. The points labeled "conventional analysis" are from the calibration curve of doubles rate vs ^{235}U mass.

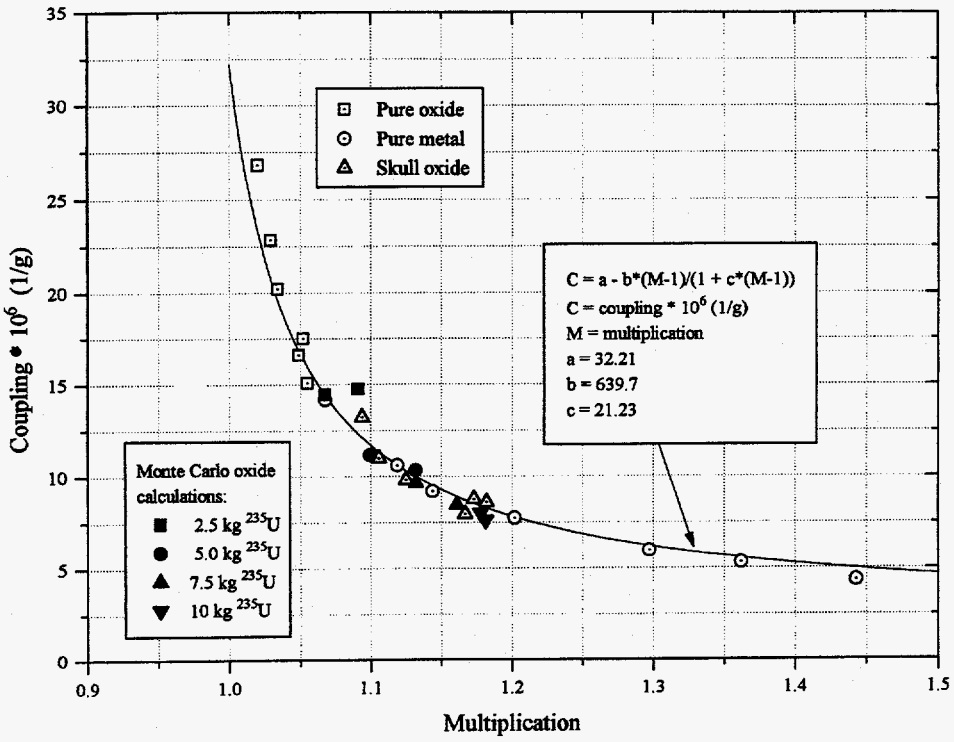

Fig. 7. Coupling vs multiplication for three categories of uranium samples.

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