NUCLEAR DATA AND MEASUREMENTS SERIES

ANL/NDM-48

^{235}U Fission Mass and Counting Comparison and Standardization

by

W.P. Poenitz, J.W. Meadows, and R.J. Armani

May 1979

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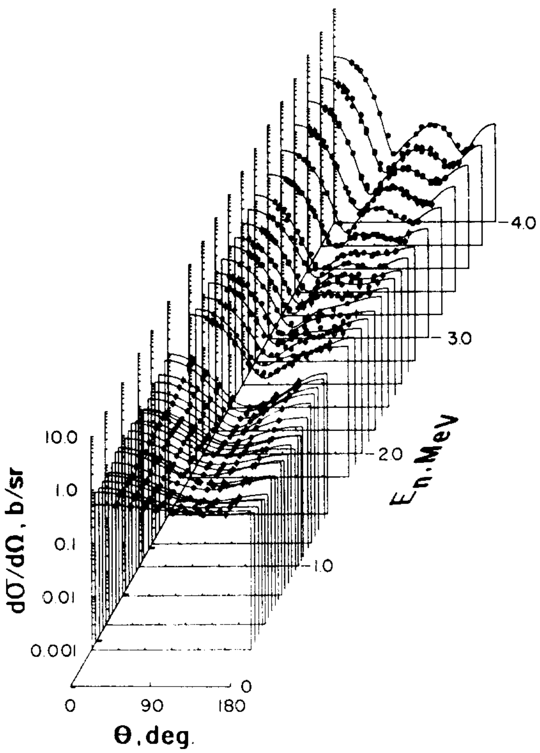

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NUCLEAR DATA AND MEASUREMENTS SERIES

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^{235}U FISSION MASS AND COUNTING COMPARISON AND STANDARDIZATION*

by

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ABSTRACT

^{235}U sample mass intercomparisons carried out at different laboratories were compiled. The compilation reveals a trend with the NBS mass scale being systematically higher by 0.7% than other mass scales. Present measurements by fast neutron fission counting confirm this difference. The present measurements result in a unified mass scale with about 0.6% uncertainty. Mass scales from LASL, ANL (#1) and the University of Michigan are in excellent agreement ($\sim \pm 0.1\%$) and within $\sim 0.3\%$ of the unified mass scale. The uncertainty of the unified mass scale established with the present measurements reduced the uncertainty for ^{235}U mass and fission counting by about a factor of 2 compared with the NBS ^{235}U mass scale against which all previous comparisons were made.

*This work supported by the U.S. Department of Energy.

I. INTRODUCTION

The fast neutron fission cross section of ^{235}U is an important standard which is used as a reference in most other fission cross section measurements. Experimental data for this cross section which were measured in the last 15 years above an energy of ∿100 keV can generally be found in a ± 3-4% wide band. Though very few discrepancies exist between the recent experimental data, the level of uncertainty is still unsatisfactory relative to the ∿1% requested by reactor calculations and designers. Little improvement can be expected in the knowledge of the $^{235}U(n,f)$ cross section, except from a new generation of experiments carried out at a ∿1% uncertainty level. Therefore, a specialists discussion meeting was organized in June of 1978 at the National Bureau of Standards in order to discuss the planning of such future measurements.[1]

It was recognized at this meeting that an increasingly important question for such future measurements is the standardization of the mass of the ^{235}U samples used in these experiments. The establishment of an U.S. fissile mass standard sample set was suggested at this meeting.[2] Such a standard set does not yet exist. However, several sample sets are in use at different laboratories and intercomparisons have been carried out among some of these laboratories. Most of these intercomparisons were made with a set of samples established at the National Bureau of Standards (NBS). The mass scale of the NBS sample set is stated with a 1.2% uncertainty. This appears inadequate for future ^{235}U cross section measurements by about a factor of 2-3. However, it may be possible to establish an improved mass scale by utilizing the various intercomparisons carried out in the past. A compilation of past intercomparisons made at various laboratories at different times may be less consistent than an intercomparison of the different mass scales in a single experiment. The major part of the present report describes the intercomparison of several samples representing four different mass scales.

II. COMPILATION OF SOME OF THE PREVIOUS MASS INTERCOMPARISONS

Several intercomparions of ^{235}U samples were made during the last several years.[3-6] Most of these intercomparisons were relative to the NBS sample set. Samples from all sets were used in the present measurements (described in Section III) except for samples from the University of Michigan. Figure 1 is a schematic diagram which summarizes the relative differences between the stated masses of the samples and those determined relative to other samples by alpha or fission counting. The origin of the arrow indicates the origin of the measurement, e.g. the comparison between the two samples of Meadows (ANL M-5-2 and ANL M-SST-5) and the NBS sample 25S-2-5 was carried out by Meadows. Figure 1 also contains information on sample deposit thicknesses which is of interest in understanding the thermal fission comparisons. (E.g., the difference between the alpha counting intercomparison and the fission counting intercomparison between the NBS reference deposit and the LASL spare #1 sample might be due to the fission fragment absorption correction for the substantially different sample thicknesses.)

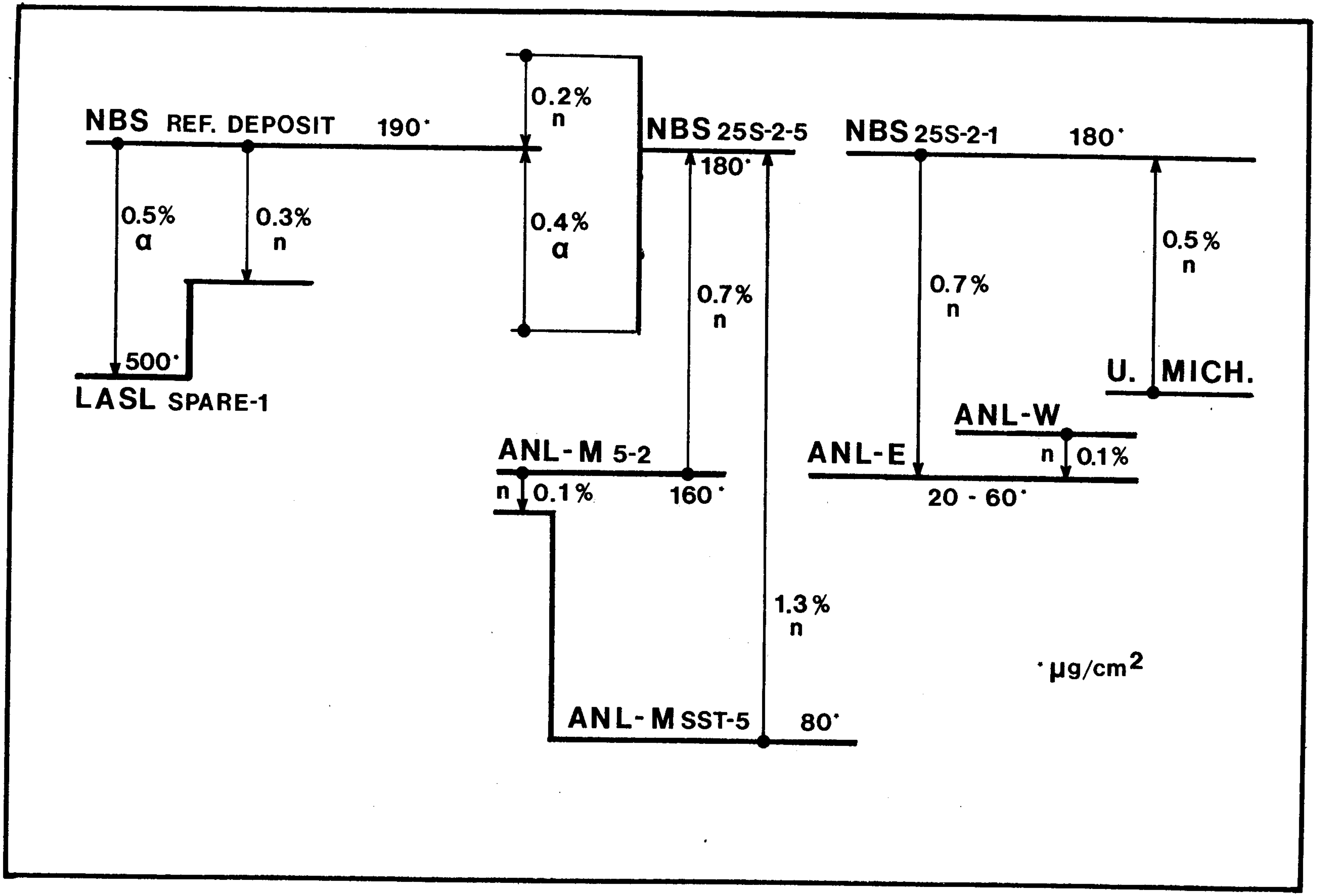

Fig. 1. Comparison of ^{235}U Mass Scales from Previous Intercomparisons. n indicates thermal neutron intercomparison and α indicates α-counting intercomparison.

Figure 1 reveals other differences which cannot be explained by sample thickness variations. The mass of NBS 25S-2-5, determined relative to the NBS reference deposit, differs by 0.6% depending on whether alpha-counting or thermal neutron fission counting was used. The two samples ANL M-5-2 and ANL M-SST-5 are consistent within 0.1% when compared with each other, but differ by 0.6% in the intercomparison with NBS 25S-2-5. Such differences appear to be indicative of the uncertainty of these measurements and are probably within the estimated errors. The differences shown in Fig. 1 are those reported by the experimenters, no attempt was made to revise this information. (E.g. the sample mass of ANL M-5-2 should be restated based on a newer isotopic dilution analysis for this material (as noted in Section III).

Figure 1 shows that the NBS mass scale, against which most other samples were measured, appears to be higher than any other mass scale. The average difference is 0.7% which is well within the stated uncertainty of 1.2% for the NBS mass scale. The perception of Fig. 1 is that six mass scales agree rather well, possibly within ±0.1 - 0.2%. This would suggest that the ^{235}U mass problem could be considered as solved after adjustment of the NBS mass scale and resolution of the above mentioned possible inconsistency between the two Meadows' foils. However, closer scrutiny reveals that many of the mass scales of the previous intercomparisons shown in Fig. 1 were not independent. The NBS samples were made from the same material as the LASL samples, and the isotopic composition of this material contributed to the determination of both mass scales. The ANL M-5-2 and ANL-East samples consist of the same material, and the good agreement shown in Fig. 1 appears accidental after updating ANL M-5-2. ANL West contributed to establishing the ANL-East mass scale, thus the good agreement between both is not too suprising either. For a long time ANL-West has also compared its mass scale with LASL. Though adjustments were not made as a result of the comparisons, indirect dependence may be suggested.[24] Therefore, of the six mass scales perceived to be in good agreement, only three scales are truely independent. Consideration of the previous intercomparisons suggests that their utilization for establishment of a unified mass scale requires a reduction of uncertainties to a <0.5% level and a careful consideration of the independence of contributing mass scales.

III. DESCRIPTION OF SAMPLES UTILIZED IN THE PRESENT INTERCOMPARISON

Eight samples were used in the present measurements. These samples were made from three fissile materials and represented four different mass scales. The physical parameters of these samples, the fissile materials and their isotopic composition and specific activities are summarized in this section. The four different mass scales and their representation in the present measurements are described briefly.

III.1. Fissile Materials

Values for the isotopic compositions and the specific activities are summarized in Table 1.

TABLE 1. Isotopic Composition and Specific Activity

Material	Isotopic Composition/wt%				Specific Activity/αpm/μg				
	U-234	U-235	U-236	U-238	Isotopic Dilution	Isotopic Composition Half Lives	Colorimetric	Others	Average
LASL INS-1	0.0608	99.7476	0.0655	0.1261	13.34 ± 0.02	13.31 ± 0.16	-	13.30 ± 0.08	13.33 ± 0.01
ANL U5-S-U4	1.028	98.403	0.447	0.122	146.2 ± 0.3	147.9 ± 0.9	146.1 ± 0.9	-	146.4 ± 0.3
ANL M-TH	0.852	93.244	0.334	5.570	-	123.1 ± 0.4	123.8 ± 0.7	-	123.3 ± 0.3

LASL INS-1

This material has been in use at LASL for a long period of time. The NBS used the same material. Values for the isotopic composition were derived from measurements at the EURATOM Central Bureau for Nuclear Measurements at Geel, at NBS, and at LASL. The specific activity for this material was determined at CBNM by isotopic dilution, and derived from the isotopic composition and known half-lifes. An additional value exists from LASL, but the technique for the measurement of this value is unidentified.

ANL U5-S-U4

This material was obtained by spiking high-purity ^{235}U with ^{234}U.[13] The isotopic composition was derived from two sets of measurements at ANL-West (Idaho) and from the original spiking procedure. Values for the specific activity were obtained from the isotopic composition and the known half-lives, from an isotopic dilution analysis at ANL-West, from a colorimetric comparison with a standard solution at ANL-East and absolute alpha counting.

ANL M-TH

This material was used in the determination of the half-life of ^{234}U.[14] The isotopic composition was determined at ORNL, ANL-West and ANL-East. The specific activity was determined by relative alpha-countings of two sets of foils and subsequent colorimetric analysis of some of the foils. An additional value was obtained from the isotopic composition and known half-lifes. The latter value is not quite independent of the former because the half-life of ^{234}U measured with this material[14] contributes to the value used in the calculation of the specific activity.

III.2. Sample Description

ANL-R-5

This foil is in use for integral fission ratio measurements in fast reactor assemblies. The previously used mass was based upon older values for the specific activity of U5-S-U4 and was updated in Table 2.

ANL N-U5-3

This foil is one of a set of samples prepared for the thermal neutron fission mass intercomparison between NBS, ANL-East and ANL-West (see Section II). Alpha-counting showed that some mass was lost from this foil (1.1%) and the value given in Table 2 was updated for this loss.

ANL M-5-1 and M-5-2

These foils are in use in fission ratio experiments.[12,15] The previously used mass was based upon the isotopic composition and half-lives and was updated in Table 2 by using present values for the

TABLE 2. Sample Set Summary

Sample			Fissile Sample Deposit						Backing		
No.	LAB	Label	Material Label	Material	Deposition Technique	U µg	Diam. cm	Thickness $\mu g/cm^2$	Material	Thickness cm	Diam. cm
1	ANL	R-5	U5-S-U4	U_3O_8	EP[a]	79.8	2.22	20.6	SS	0.013	4.445
2	ANL	N-U5-3	U5-S-U4	U_3O_8	EP	52.09	1.27	41.1	SS	0.025	1.905
3	ANL	M-5-1	U5-S-U4	U_3O_8	EP	1066	2.54	210.4	SS	0.025	6.985
4	ANL	M-5-2	U5-S-U4	U_3O_8	EP	831.8	2.54	164.2	SS	0.025	6.985
5	ANL	M-SST-5	U5-Th	UF_4	EV[b]	411.5	2.54	81.2	SS	0.025	6.985
6	LASL	S1	INS-1	U_3O_8	EV	298.8	2.00	95.1	Pt	0.013	4.763
7	LASL	S3	INS-1	U_3O_8	EV	1689.4	2.00	537.9	Pt	0.013	4.763
8	NBS	25S-2-5	INS-1	U_3O_8	EV	230.6	1.27	182.0	Pt	0.013	1.905

[a]Electroplating.

[b]Evaporation.

specific activity. M-5-2 was also used in the thermal neutron fission mass comparison with NBS (see Section II). Both samples were used in absolute $^{235}U(n,f)$ measurements.[16]

ANL M-SST-5

This foil is one of a set of samples used in the determination of the half-life of ^{234}U.[14] The half-life from this experiment agrees with the presently accepted value within 0.6%. Subsequently this foil was used in fission ratio measurements[10,12] and in the intercomparison with NBS (see Section II). This foil was also used in an absolute $^{235}U(n,f)$ cross section measurement at 800 keV.[17]

LASL S1 and S3

These samples are two of a set of samples used in the measurement of absolute fast neutron cross sections.[18] Another sample of this set was used in an alpha-counting and a thermal neutron fission counting comparison with the reference deposit of the NBS sample set (see Section II). The samples of this set were intercompared by alpha and by thermal fission counting.

NBS 25S-2-5

The NBS sample set consists of a reference deposit and other samples which were measured relative to this reference by alpha and thermal fission counting. This sample was used in a thermal neutron fission counting intercomparison with ANL.

III.3. Mass Scales

Several of the eight samples belong to identical mass scales. In two cases this is very obvious: LASL S1 and LASL S3 belong to the same scale, so do ANL M-5-1 and ANL M-5-2. All samples belong to one of four mass scales which are reasonably independent.

LASL Mass Scale

This mass scale is determined by the high weight of the EURATOM isotopic dilution analysis. This scale is represented by LASL S1 and LASL S2.

NBS Mass Scale

The NBS mass scale is based upon the comparison of the reference deposit with a sample obtained by quantitative deposition, the comparison with ^{239}Pu samples, and the mass determination from the isotopic composition and known half-lives. The latter also contributes to the LASL mass scale, however, the weight of this technique is small for the LASL scale and the NBS mass scale can be considered essentially independent of the LASL mass scale. The NBS mass scale is represented by the sample 25S-5-2.

ANL Mass Scale #1

This mass scale is based upon the determination of the specific activity by three different techniques as described in Section III.1. The major weight is with the isotopic dilution technique. This mass scale is represented by the samples R-1, N-U5-3, M-5-1 and M-5-2.

ANL Mass Scale #2

This mass scale is based upon comparison with a standard solution and the mass determination from the isotopic composition and known half lives. The larger weight comes from the latter technique. This mass scale is represented with the sample M-SST-5.

IV. MASS DETERMINATION AND INTERCOMPARISON BY ALPHA COUNTING

Utilizing the specific activities for the varying materials given in Table 1 permits the determination of the fissile mass by alpha-counting.

IV.1. Low-Geometry Alpha-Counting

Two low-geometry alpha-counters of similar design were available. The alpha-detector was a surface-barrier detector which was placed behind a 1.27 cm diameter aperture. For one of the detectors the aperture was lapped in order to reduce the "lip" to <0.0025 cm. The lip of the second detector's aperture was measured to be 0.0203 cm. Samples could be positioned on a tray at a distance of $\sim$4.57 cm from the aperature, or at larger distances with 5.08 cm increments. Geometry factors were calculated with a Monte Carlo procedure[7] or with an approximate correction to the point source geometry factor.[8] All samples were initially counted at the shortest distance from the aperture with a typical statistical uncertainty of $\sim$0.3%. Samples with a sufficiently high alpha-activity were also counted at the second tray position, thus changing the geometry factor by about a factor of four. Comparison of the decay rates obtained at the different distances increased the confidence in the calculated geometry factors. The ratio of values obtained at the second shelf position to the value obtained at the closest shelf position was

$$1.0002 \pm 0.0009.$$

A comparison was made between the two low-geometry counters using an ^{241}Am sample. A ratio of

$$1.0008 \pm 0.0007$$

was found. The uncertainties for both ratios above is the statistical uncertainty. Based on these values, the estimated systematic uncertainty due to the geometry of the counters does not exceed 0.11%.

A more serious matter is the interpretation of the measured alpha-spectra. All the spectra show a low-energy tail as illustrated in Fig. 2. The possibility of inscattering from the walls and the aperture of the counter can be excluded due to the geometry involved. Backscattering from the sample backings was suggested as a source of this low-energy tail.[9] However, in the present measurements the relative amount of the tail seems independent of the sample thickness and the backing material. The origin of the low-energy tail in the alpha-spectrum could not be established and must therefore be considered an uncertainty. The tail was, on the average, $\sim$0.3% of the total count rate. It may be assumed that the tail extends under the alpha-peak as indicated in Fig. 2. Therefore, we assume that the total amount of these alphas is $\sim$0.4%. One-half, that is 0.2%, were included in the count rate and 0.2% was assumed to be the systematic error.

The results from the low-geometry alpha-counting are given in Table 3. All samples were counted in one of the low-geometry counters before the fission ratio experiment. Several samples were counted again in low-geometry after the fission ratio experiment using either of the two counters.

IV.2. 2π Alpha-Counting

The lower alpha-activity samples were counted with a proportional counter in a 2π geometry. Correction factors for absorption and backscatter were based upon measurements with the low-geometry counter, thus these values are not independent from the low-geometry alpha-counting.

IV.3. Results from the Mass Determination of Alpha Counting

The present results for the alpha-decay rate of the eight samples given in Table 3 were combined with the present compilation of the specific activity of Table 1 in order to derive the mass of uranium for the different samples. The values are given in Table 4 which also contains the values presently quoted for these samples as given in Table 2, and the values which were previously used.

For those samples which consist of the same fissile material the relative alpha-decay rates may be used in order to derive relative mass values. The results of this approach are given in Table 5. The uncertainties of the derived mass values of Table 5 are determined by the uncertainty of the reference mass and the statistical uncertainties of the relative alpha counting. Systematic uncertainties of the alpha-counting should cancel in first order in this procedure.

Table 5 shows that the ANL samples agree rather well within the statistical uncertainties involved. The differences between the LASL and the NBS samples are somewhat larger, though still within a reasonable range of statistical uncertainties. However, it may be noted that the average 0.5% by which the NBS value seems to be higher than that derived with the LASL samples is by chance identical with the difference measured by NBS between the NBS reference deposit and LASL "spare #1" (see Section II, Fig. 1).

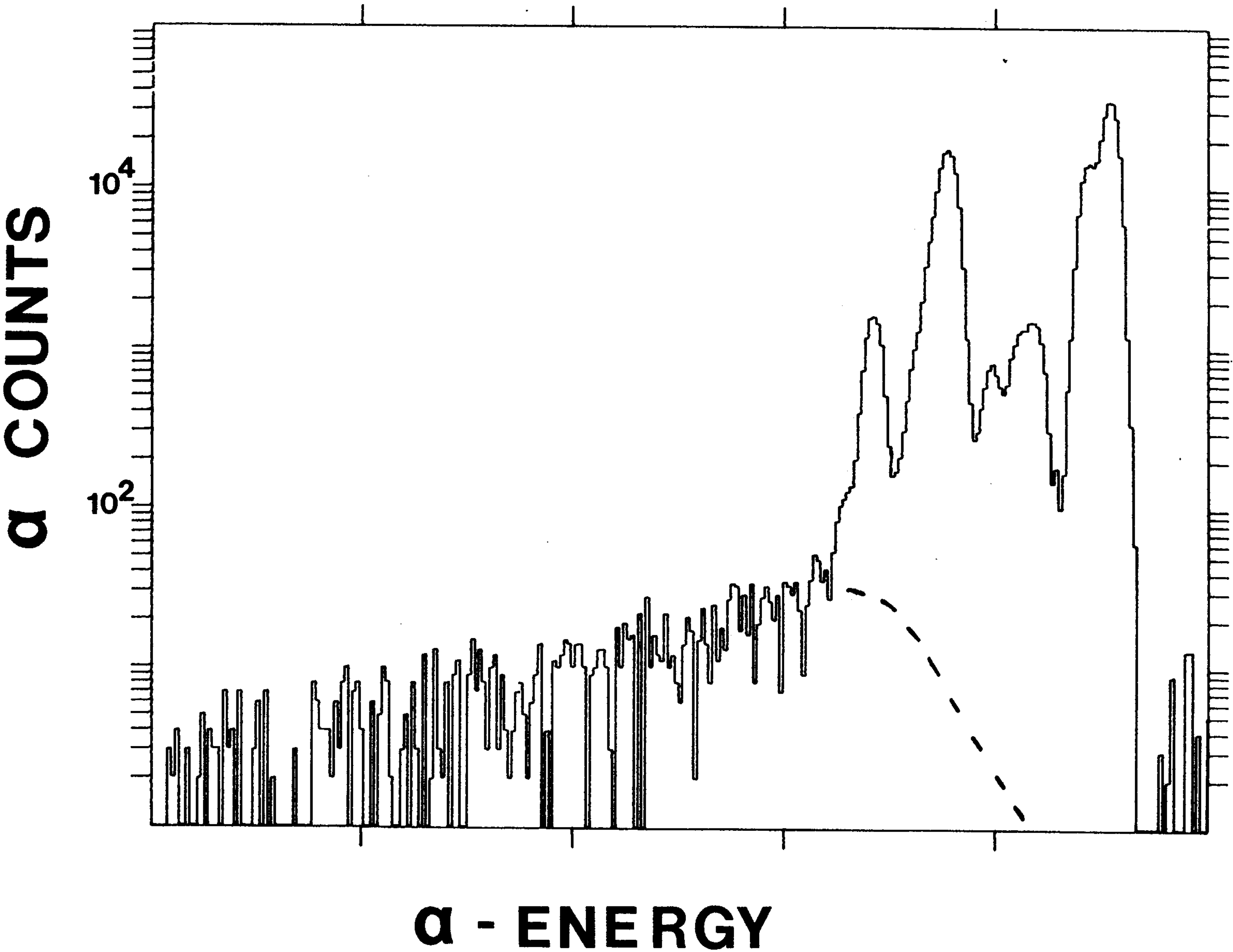

Fig. 2. α-Spectrum Obtained with the Low-Geometry α-Counter for Sample LASL-S1

TABLE 3. Results from Alpha-Counting

No.	Sample	LG/αps[a,d]	LG/αps[b,d]	2πPC/αps[c,d]	WA/αps[e]
1	ANL R-5	194.5 ± 0.7	194.1 ± 0.7	193.4 ± 0.9	194.1 ± 0.7
2	ANL N-U5-3	127.3 ± 0.5	-	126.9 ± 0.5	127.1 ± 0.5
3	ANL M-5-1	2594 ± 10	2593 ± 6	-	2593 ± 6
4	ANL M-5-2	2030 ± 8	2029 ± 5	-	2029 ± 5
5	ANL M-SST-5	845.7 ± 3.2	846.1 ± 2.4	-	846.0 ± 2.4
6	LASL S1	66.50 ± 0.26	-	66.65 ± 0.26	66.58 ± 0.26
7	LASL S3	375.7 ± 1.7	374.7 ± 1.3	373.9 ± 1.5	374.7 ± 1.3
8	NBS 25S-5-2	51.01 ± 0.21	-	51.03 ± 0.20	51.02 ± 0.20

[a] Low-geometry alpha-counting before the fission ratio experiment.

[b] Low-geometry alpha-counting after the fission ratio experiment.

[c] 2π proportional counter measurement after the fission ratio experiment.

[d] 0.2% subtracted from the total count rate and added to the uncertainty (see text).

[e] Weighted average, but uncertainty limited to the smallest uncertainty of the individual measurement.

TABLE 4. Uranium Mass from Alpha-Counting

No.	Sample	αps	Spec. Activity αpm/μg	Mass/μg[a]	Mass/μg[b]	Mass/μg[c]
1	ANL R-5	194.1 ± 0.7	146.4 ± 0.3	79.55 ± 0.33	79.8 ± 0.3[d]	78.2 ± 0.6
2	ANL N-U5-3	127.1 ± 0.5	146.4 ± 0.3	52.09 ± 0.23	52.09 ± 0.25[e]	52.67 ± 0.3
3	ANL M-5-1	2593 ± 6	146.4 ± 0.3	1063 ± 3	1066 ± 5	1049
4	ANL M-5-2	2029 ± 5	146.4 ± 0.3	831.6 ± 2.7	831.8 ± 3.5	819.8
5	ANL M-SST-5	846.0 ± 2.4	123.3 ± 0.3	411.7 ± 1.5	411.5 ± 1.5	411.6
6	LASL S1	66.58 ± 0.26	13.33 ± 0.01	299.7 ± 1.2	298.8 ± 0.2	298.8 ± 0.2
7	LASL S3	374.7 ± 1.3	13.33 ± 0.01	1687 ± 6	1689.4 ± 4.1	1689.4 ± 4.1
8	NBS 25S-2-5	51.02 ± 0.20	13.33 ± 0.01	229.6 ± 0.9	230.6 ± 3.0	230.6 ± 3.0

[a]Result from present measurements and compilation.

[b]Values presently quoted by the owners of these samples.

[c]Values previously used for these samples.

[d]Adjusted for improved values of the specific activity.

[e]Adjusted for a 1.1% loss of material.

TABLE 5. Masses Derived from Relative Alpha-Counting

Reference Sample	Reference Mass/µg	Derived Mass/µg						
		ANL R-5	N-U5-3	M-5-1	M-5-2	LASL S1	S3	NBS
ANL R-5	79.8 ± 0.3		52.25 ± 0.30	1066 ± 5	834.2 ± 4.0			
ANL N-U5-3	52.09 ± 0.25	79.5 ± 0.5		1063 ± 6	831.6 ± 4.9			
ANL M-5-1	1066 ± 5	79.8 ± 0.4	52.25 ± 0.30		834.1 ± 4.0			
ANL M-5-2	831.8 ± 3.5	79.6 ± 0.4	52.11 ± 0.28	1063 ± 5				
LASL S1	298.8 ± 0.2							229.0 ± 1.0
LASL S3	1689.4 ± 4.1							230.0 ± 1.1
NBS 25S-2-5	230.6 ± 3.0					300.9 ± 4.1	1694 ± 23	
Average Derived Mass/µg		79.7 ± 0.1	52.20 ± 0.05	1064 ± 1	833.5 ± 0.8	300.9 ± 4.1	1694 ± 23	229.4 ± 0.5
Reference Mass/µg		79.8 ± 0.3	52.09 ± 0.25	1066 ± 5	831.8 ± 3.5	298.8 ± 0.2	1689.4 ± 4.1	230.6 ± 3.0
Difference of Derived Mass from Reference Mass/%		-0.1	+0.2	-0.2	+0.2	+0.7	+0.3	-0.5

V. FAST NEUTRON FISSION RATIO MEASUREMENTS

V.1. Experimental Procedure

The present experimental procedure followed that employed for fast neutron fission cross section ratio measurements described in detail elsewhere.[10-12] The present measurements were carried out at an average energy of 600 ± 80 keV. The fissile deposits were located back-to-back at a distance of ~5 cm from the neutron source. Measurements were carried out with each sample alternately facing the neutron source or facing away from the source.

Special care was taken to assure identical signal processing. Pulses from a random source were split on a 1 : 1 basis and added to each fission chamber preamplifier. These events were found to be processed by the on-line computer system and the associated electronics with a better than 0.1% parity. An additional check was made by reversing the preamplifiers (and all subsequent electronics and computer software) in one of the ratio experiments. The ratio of the two measurements was found to be

0.9985 ± 0.0025.

V.2. Corrections

All corrections were applied to the two results for the different orientations of the fission chamber. However, some of these corrections cancel out when the average of the two values is formed and therefore systematic uncertainties are reduced. Corrections were applied for different distances of the fission samples from the source, transmission through the sample backings, scattering of neutrons within the chamber and the target assembly, neutron source angular anisotropy, isotopic composition, and fission fragment absorption, including momentum effect and angular distribution. Detailed descriptions of such corrections were previously given.[10-12,19,23] Fission fragment absorption was calculated using a range of 4.1 per mg U/cm^2 for the samples R-5, N-U5-3, M-5-1 and M-5-2, and 4.7 per mg U/cm^2 for M-SST-5. These values were obtained from measurements for these deposits.[19] A range of 6.5 per mg U/cm^2 was used for the samples LASL S1, S2 and NBS 25S-2-5. This value was concluded from measurements of the absorption of fission fragments in Pu-deposits for the NBS samples.[20]

V.3. Results from Fission Ratio Measurements

The corrected ratios measured for the two different directions of the fission chamber differed on the average by more than 1%. This was ascribed to inadequate corrections for the different distances of the fission deposits from the source. It appears difficult to mount fission samples with warped backings back-to-back in a lightweight fission chamber with sufficient pressure to eliminate any additional spacing between the samples. Measurements at different distances from the neutron source and different mountings of the samples, as well as the observation that the differences were largest for warped samples supported this interpretation. The averages of the ratios from the two directions of the counter were formed. This eliminates the error for the correction for the different distances of the samples from the source. It also cancels the correction for neutron transmission through the sample backings and reduces the effect of the corrections for scattering of neutrons and for momentum transfer to the fragments. Thus, the remaining major systematic uncertainties are due to the corrections for the neutron source anisotropy (which is zero for samples of equal size) and for the fission fragment absorption.

The ratio of two samples of different size was measured for varied conditions in order to check further the appropriate application of corrections. With the two samples mounted back-to-back at the standard distance of $\sim$5 cm from the source, the ratio

1.2982 (±0.21% statistics)

was obtained. At twice the distance from the source, the ratio

1.2976 (±0.28% statistics)

was determined. Finally, mounting the samples on a 0.127 mm molybdenum plate resulted in the value

1.2995 (±0.18% statistics).

The measurement of seven ratios between the eight samples provided sufficient data to derive any desirable mass ratio. However, more than seven ratios were measured and therefore some measured values may be derived from others. This provides a check on the consistency of the present measurements. Table 6 contains those experimental quantities which represent an overdetermined system of values. Also given in this table are the values which may be derived from other measurements, and the weighted averages of the directly measured and the derived values. The latter were used as approximations in order to derive a consistant set of values in a fitting procedure. The results of the consistency fit are also given in Table 6. The consistent values of M1-M4 of Table 6 are listed together with the non-overdetermined values in Table 7 and represent the results from the present fission ratio measurements. Using any of the eight sample masses as reference, the values for the others were derived from the ratios in Table 7 and are given in Table 8.

VI. CONCLUSIONS AND RECOMMENDATIONS

The results given in Table 8 should be condensed to reflect mass scales instead of individual samples. This was done in Table 9. It can be seen that the NBS mass scale gives consistently high values and the ANL #2 mass scale gives consistently low values. Average ("unified") mass scale values for the eight samples are given as unweighted averages of the four contributing mass scales. The relative differences are shown in Fig. 3 in a schematic similar to Fig. 1. Also shown in this figure is the mass scale from the University of Michigan based on the thermal neutron fission ratio measurements (see Section II). The heavy lines (levels) in Fig. 3 represent the mass scales, the level-splits display the different

TABLE 6. Consistency of Overdetermined System

Measurement	Ratio	Measured Value	Derived Values	Average Value	Consistent Values
M 1	$\frac{\text{M-5-2}}{\text{M-SST-5}}$	2.001 (4)[a]	M4/M8 2.010 (5) M3/M5 1.999 (5)	2.003 (4)	2.004
M 2	$\frac{\text{R-5}}{\text{M-SST-5}}$	0.1902 (4)	M3/M6 0.1899 (5)	0.1901 (4)	0.1898
M 3	$\frac{\text{S1}}{\text{M-SST-5}}$	0.7170 (14)	M1*M5 0.7178 (19)	0.7173 (14)	0.7152
M 4	$\frac{\text{NBS}}{\text{M-SST-5}}$	0.5480 (10)	M3/M7 0.5522 (12)	0.5497 (20)	0.5492
M 5	$\frac{\text{S1}}{\text{M-5-2}}$	0.3587 (6)	M3/M1 0.3583 (10)	0.3586 (6)	(0.3569)[b]
M 6	$\frac{\text{S1}}{\text{R-5}}$	3.777 (7)	M3/M2 3.770 (11)	3.775 (7)	(3.768)
M 7	$\frac{\text{S1}}{\text{NBS}}$	1.298 (2)	M3/M4 1.308 (4) M5/M8 1.316 (3)	1.304 (6)	(1.302)
M 8	$\frac{\text{NBS}}{\text{M-5-2}}$	0.2726 (4)	M4/M1 0.2739 (6)	0.2730 (6)	(0.2741)

[a] The value in the bracket gives the statistical uncertainty of the last digit of the value.

[b] Values in this column with a bracket are calculated from those without a bracket.

TABLE 7. Results from Fission Ratio Measurements

Ratio	Value
$\frac{\text{ANL M 5-2}}{\text{ANL M-SST-5}}$	2.004
$\frac{\text{ANL R-5}}{\text{ANL M-SST-5}}$	0.1898
$\frac{\text{LASL S1}}{\text{ANL M-SST-5}}$	0.7152
$\frac{\text{NBS}}{\text{ANL M-SST-5}}$	0.5492
$\frac{\text{ANL N-3}}{\text{ANL M-SST-5}}$	0.1242
$\frac{\text{LASL S3}}{\text{ANL M SST-5}}$	4.038
$\frac{\text{ANL M 5-1}}{\text{ANL M 5-2}}$	1.2805

TABLE 8. Masses Derived from Relative Fission Counting

Reference		Mass Derived from Fission Ratios and Reference							
Sample	Mass/μg	ANL R-5	ANL N3	ANL M-5-1	ANL M-5-2	ANL M-SST-5	LASL S1	LASL S3	NBS
ANL R-5	79.8	79.8	52.22	1079	842.6	420.4	300.7	1697.7	230.9
ANL N-U5-3	52.09	79.6	52.09	1076	840.5	419.4	300.0	1693.6	230.3
ANL M-5-1	1066	78.8	51.59	1066	832.5	415.4	297.1	1677.4	228.1
ANL M-5-2	831.8	78.8	51.55	1065	831.8	415.1	296.9	1676.1	228.0
ANL M-SST-5	411.5	78.1	51.11	1056	824.6	411.5	294.3	1661.6	226.0
LASL S1	298.8	79.3	51.89	1072	837.2	417.8	298.8	1687.0	229.4
LASL S3	1689.4	79.4	51.96	1074	838.4	418.4	299.2	1689.4	229.8
NBS 25S-5-2	230.6	79.7	52.15	1077	841.4	419.9	300.3	1695.5	230.6

TABLE 9. Masses Derived from Different Mass Scales/μg

Mass Scale	ANL R-5	ANL N-U5-3	ANL M-5-1	ANL M-5-2	ANL M-SST-5	LASL S1	LASL S3	NBS 25S-5-2
ANL #1	79.25	51.86	1071.5	836.9	417.6	298.7	1686.2	229.3
ANL #2	78.1	51.11	1056	824.6	411.5	294.3	1661.6	226.0
LASL	79.35	51.93	1073	837.8	419.1	299.0	1688.2	229.6
NBS	79.7	52.25	1077	841.4	419.9	300.3	1695.5	230.6
Average Fission Scale	79.10	51.76	1069	835.2	416.8	298.1	1683	228.9
Absolute Alpha Specific Activity Compilation	79.55	52.09	1063	831.6	411.7	299.7	1687	229.6
Result of Present Alpha and Fission Measurements	79.3	51.9	1066	833	414.3	298.9	1685	229.3
Quoted Mass	79.8	52.1	1066	832	411.5	298.8	1689.4	230.6
Difference/%	0.6	0.4	0.0	-0.1	-0.7	0.0	0.3	0.6

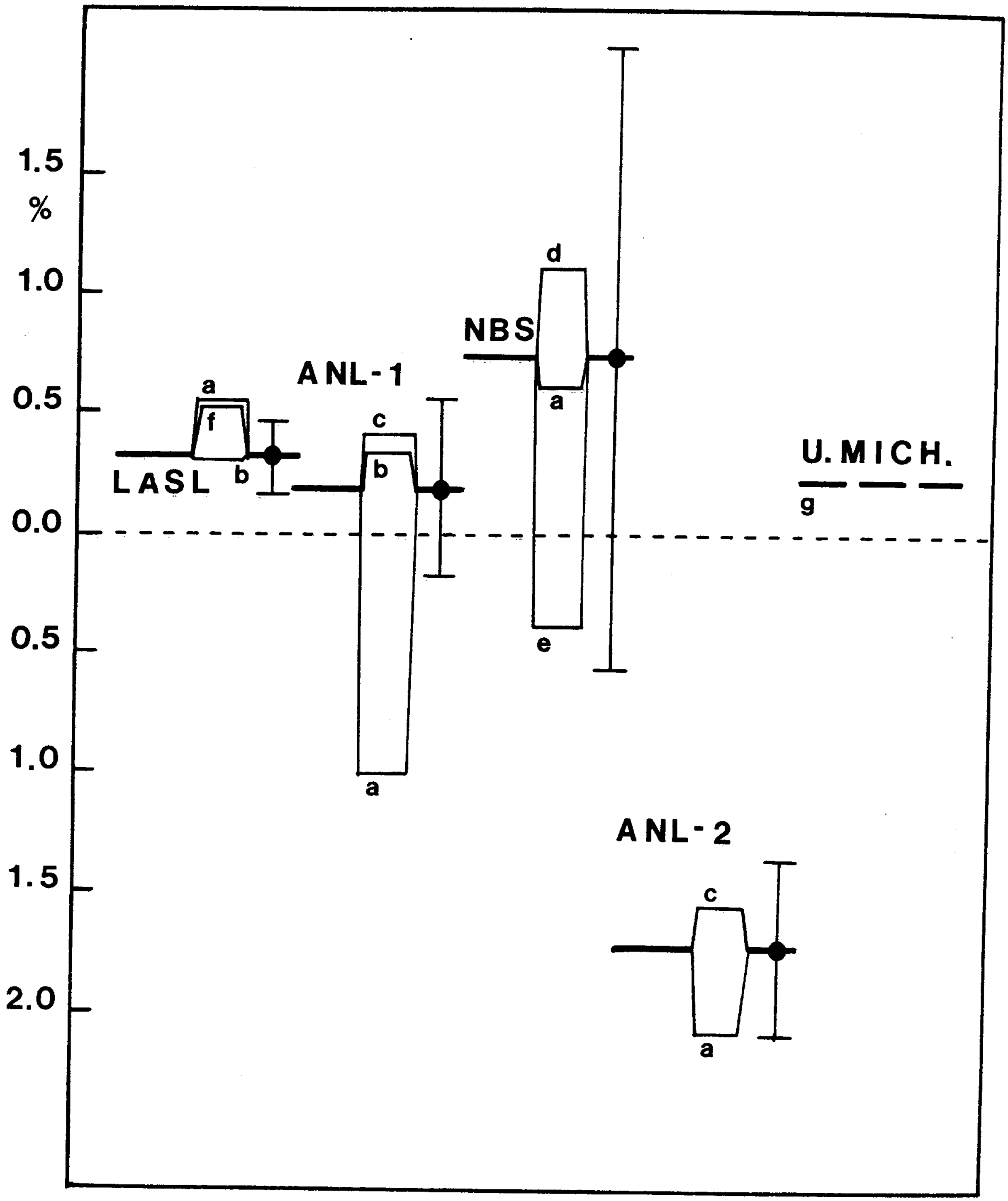

Fig. 3. Comparison of the Four Different Mass Scales Involed in the Present Intercomparison. Values are shown relative to a "unified mass scale" derived as an unweighted average. a = isotopic composition and half-life, b = isotopic dilution, c = colorimetric comparison with standard, d = thermal neutron comparison with quantitative deposition, e = thermal neutron comparison with ^{239}Pu sample, g = weighting.

components which contributed to the establishing of these mass scales (see Section III). The error bars indicate the uncertainty of the original mass values. Considerations of Fig. 3 and Table 9 suggests that

- Agreement is very good between the mass scales of LASL and ANL #1 (ANL-East and ANL-West), both of which are heavily weighted with the isotopic dilution results. These scales agree with the U. of Michigan scale which is based on weighing the deposit.

Comparison between Fig. 3 and Fig. 1 shows that the present measurements confirm the relative differences obtained with thermal neutron fission counting between the LASL, the ANL-East, the ANL-West, and the NBS mass scales. Larger differences were observed in the present measurements than in the thermal intercomparison with NBS by Meadows ($\sim$2.5% instead of 1.3% for the sample M-SST-5 and $\sim$1.7% vs 0.7% for the sample M-5-2).

It appears from Fig. 3 that much improvement could be obtained if the mass scale ANL #2 would be disregarded. However, the number of contributing mass scales is not large enough and the diversity and differences of the contributing components is too large to justify such selection at this point. A major argument against disregarding ANL #2 is that the ^{234}U half-life determined with this material is within 0.6% of the presently accepted value.[22] This value of the ^{234}U half-life is also supported by the result of a fit of the thermal neutron parameters.[21] However one might reverse this consideration and suggest:

- A substantial improvement between several mass scales would result from an increase of the half-life of ^{234}U by about 1½%. A measurement of $T_{1/2}$ (^{234}U) is recommended.

A change of the ^{234}U half-life by $\sim$1½% would remove the present difference for the ANL #1 scale between the two values obtained by the isotopic dilution technique and colorimetric comparison with a standard on the one hand and the values obtained from the isotopic composition and the known half-lives on the other hand. This difference can hardly be understood by estimated uncertainties and appears quite troublesome because the isotopic composition was not only obtained by the conventional isotopic mass analysis but also from the original spiking procedure (see Section III). A change of $T_{1/2}$ (^{234}U) by 1½% would also bring the ANL #2 scale in a reasonable range of the other mass scales. It is interesting to observe that the half-life of ^{234}U was about 2% higher until a change occurred about 8 years ago due to newer experimental work.[22] A change of the ^{234}U half-life would also change one of the components which contributed to the LASL and the NBS mass scales, however, because this is a low ^{234}U content material, (both use INS -1) the change would be only 0.9%. The uncertainty of the ^{234}U content of INS-1 is rather large (because its content is low), thus the value obtained from the isotopic composition and the half-lives has negligible weight in the determination of the LASL mass scale.

Relative fission counting with 2π geometry ionization chambers requires corrections for the fission fragment absorption. The present corrections are based on experimental values for the range of fission fragments in the deposits (see Section V). It may be of interest to consider the ratios of the masses obtained from relative fission counting and from absolute alpha-counting as given in Table 9. Figure 4 shows these ratios vs the sample thickness. These curves may suggest that either the fission fragment absoption was over-corrected for heavier samples or under-corrected for thin samples. The latter would be expected if unevenness of the polished backings remains. In order to obtain some additional information on this question we compared the 2π ionization chamber count rate of a ^{252}Cf sample with its low geometry count rate. This sample was made by self-deposition of ^{252}Cf in a vacuum on a polished, nickel-plated stainless steel plate. The ^{252}Cf fission fragment spectrum obtained from this source was of excellent quality. However, the ratio of the 2π ionization chamber count-rate to that from the low-geometry counter was

$$0.994 \pm 0.003.$$

The result again suggests that the fission fragment absorption of thin deposits may be underestimated.

- Future improved mass scale intercomparisons should include low-geometry fission counting as well as 2π counting. The backings needs to be investigated, possibly with scanning electron microscopes.

Accepting the "unified" mass scale shown in Fig. 3 suggests the assignment of an uncertainty of $\sim$0.6%. This uncertainty would mean that seven components which contributed to the individual mass scales are within one standard deviation of the unified mass scale, two would be within one and two standard deviations, one between two and three, and one component would be outside of three standard deviations. This appears to be a satisfactory situation. All but one of the sample masses determined by the present absolute alpha counting are within one standard deviation of the masses based on this "unified" mass scale.

- A "unified" ^{235}U mass scale with an uncertainty of 0.6% is the result of the present measurements and compilation. This reference scale differs by $\sim$0.7% from the NBS mass scale (used in most previous intercomparisons) and reduces the uncertainty by a factor of 2.

Figure 3 might indicate that later improvements and resolution of the ^{234}U half-life problem might change this scale by 0.2 - 0.3% (up), however, this would be well within the estimated uncertainty of 0.6%.

- The good agreement (±0.1%) between the LASL, the ANL #1, and the U. of Michigan mass scales might suggest a 0.25% higher mass scale than the present unweighted average of all included mass scales. Such change would be unimportant for a new generation of cross section experiments.

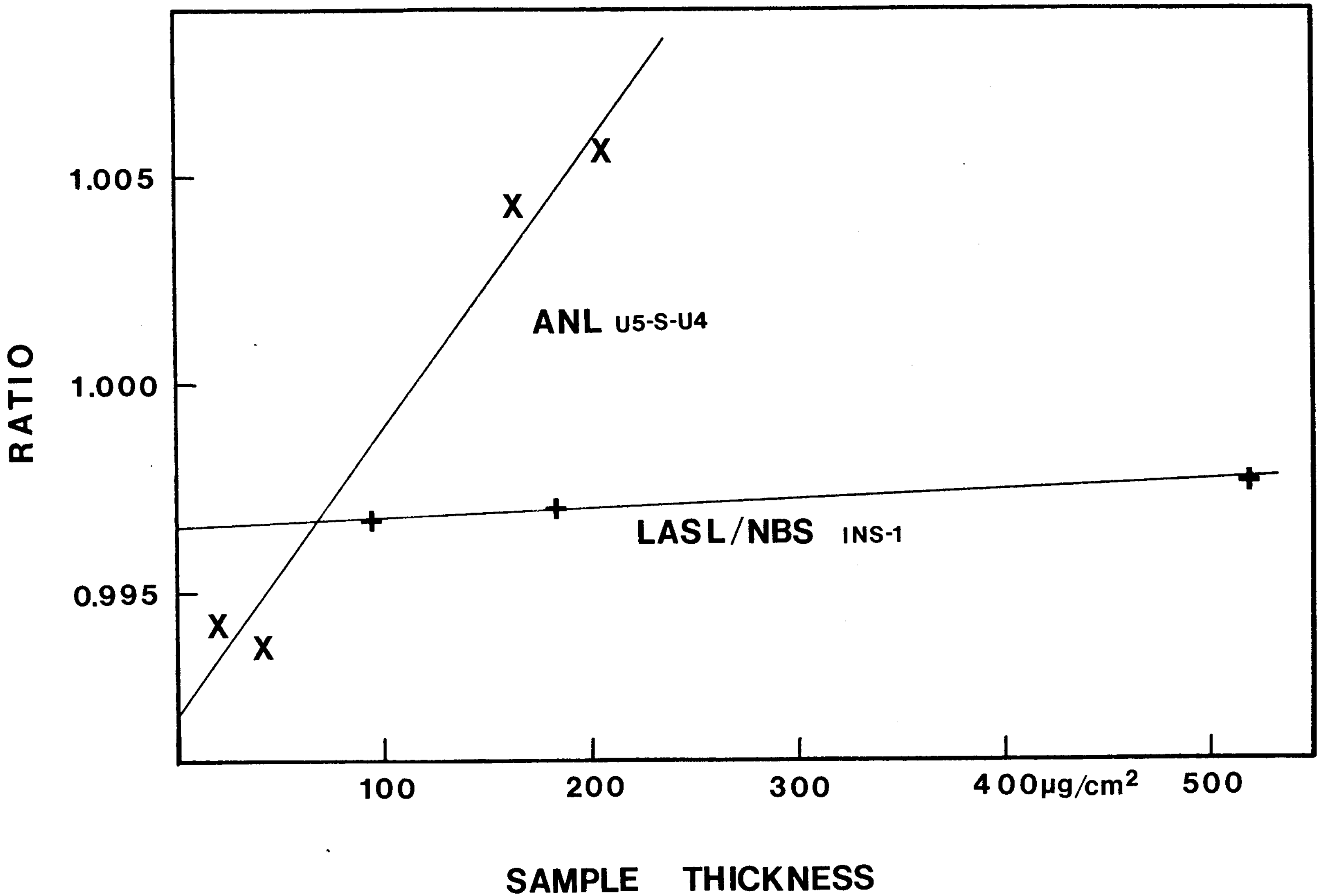

Fig. 4. Ratio of Fission-to-Alpha Counts vs. Sample Thickness for Two Different Fissile Materials. Samples with INS-1 were made by evaporation and samples with U5-S-U4 were made by electroplating.

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