

NUCLEAR DATA AND MEASUREMENTS SERIES

ANL/NDM-84

^{235}U and ^{239}Pu Sample-Mass Determinations and Intercomparisons

by

W.P. Poenitz and J.W. Meadows

November 1983

**ARGONNE NATIONAL LABORATORY,
ARGONNE, ILLINOIS 60439, U.S.A.**

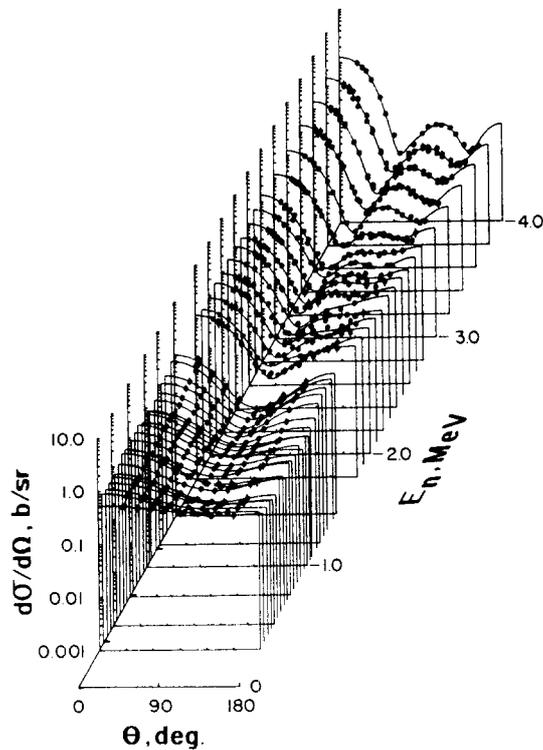
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E ^{235}U Fission Mass Standards, Absolute Alpha Counting, Fission
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^{235}U and ^{239}Pu Sample-Mass Determinations and Intercomparisons

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ABSTRACT

The masses of fifteen ^{235}U samples obtained from seven laboratories were determined and intercompared. The present results indicate that sample masses are well enough known ($\pm 0.3\%$) for the required high-accuracy-fission cross-section measurements. A comparison of four ^{239}Pu samples indicates problems in the order of 1-3% which are more likely, to be related to counting efficiencies than to sample masses. The half-life of ^{234}U was determined as $(2.457 \pm 0.005) \cdot 10^5$ yrs.

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

I. Introduction

The neutron-induced-fission cross sections of ^{235}U and of ^{239}Pu are of direct importance in reactor applications. The neutron-source term in neutronics calculations contains the fission cross sections of the fissile materials as a major factor, resulting in large sensitivities to fission-cross-section uncertainties. The $^{235}\text{U}(n,f)$ reaction is also used very frequently as a reference in fission, capture and dosimetry cross-section measurements. As a consequence, knowledge of these cross sections is required with 1% uncertainties which is reflected in corresponding entries in nuclear-data request lists (1,2). These requests have persisted for the last 10-15 years, indicating that the required low uncertainties have not yet been obtained. Measurements to that level of accuracy require careful investigation of the components which contribute to the determination of the cross section, one of which is the mass of the fissile sample. The latter is also required to be known for the measurements of reaction-rate ratios in reactor test facilities.

The required data for a specific sample is most often determined by others than the experimenter who measures the cross sections or reaction-rate ratios. The isotopic composition and the sample mass are usually obtained from associated chemistry departments or standard laboratories; however, the experimenter still has the responsibility to assure that the values he uses are adequately described by the quoted uncertainties. A variety of problems indicates the need to investigate the degree to which fission masses are known, and this has prompted the present study:

- (a) Many highly-accurate $^{235}\text{U}(n,f)$ cross sections at a 14-MeV-neutron energy have been reported in recent years (3-9) which are in good agreement, but $^{239}\text{Pu}(n,f)$ cross sections obtained in some of the same experiments are discrepant. These 14-MeV cross sections are not of great interest for fast-reactor applications, however, they significantly affect the evaluated cross sections in the lower energy region, which is important. Therefore, one would like to know whether the uncertainties quoted for the results from these measurements adequately include the sample-mass uncertainties.
- (b) The evaluation of the extensive body of existing cross-section data utilizing least-squares techniques results in very low uncertainties of 0.5 - 1.0% (10,11) which are difficult to accept because values measured with such high accuracy are, with the exception of the 14-MeV values, not found in the data base. Thus, measurements with accuracies better than or at about 1% are required in order to confirm or reject the evaluated data. This again indicates the need to know the sample masses to much better than 1%.
- (c) A recent intercomparison of reaction-rate measurements in a reactor test facility resulted in discrepancies which appear to be related to ^{239}Pu sample masses (12). This problem is related to long standing differences in the C/E discrepancies of C28/F49 found for critical assemblies operated in the U.S. and in the U.K.
- (d) Substantial and uncertain changes are indicated by recent evaluations

for the thermal-neutron fission cross sections, specifically for ^{239}Pu , which were believed to be very well known (13,14). Again, the question of the accuracy of the sample masses arises.

In order to resolve some of these problems, an intercomparison of ^{235}U masses was started in 1979 (15) and continued in 1982 (16). Several ^{239}Pu samples were included in the 1982 intercomparison. The present report contains updated values for the samples involved in the 1979 intercomparison, and presents the final data from the recent measurements which were carried out at Argonne National Laboratory. An uncertainty analysis has been carried out and is also presented.

Samples were obtained from the laboratories listed in Table 1. The fissile materials and the samples are described in Section II. Absolute alpha-decay rates were measured and masses determined, as is described in Section III. The measurements of relative fission-rate ratios are described in Section IV. "Best values" were derived for the sample masses, and they are compared with the values quoted by the various laboratories in Section V.

II. Sample Descriptions

Six different fissile materials of ^{235}U and four of ^{239}Pu were involved in the 1979 and 1982 intercomparisons which are discussed in the present report. The isotopic compositions and data on the specific activities, which were made available by the laboratories contributing the samples or which were derived in the present work, are given in Tables 2 and 3. Values given for the specific activities based upon the isotopic compositions (IC) and half-lives were derived with the reported IC's and the half-lives given in Table 4. The laboratories which contributed the samples may have used different half-lives. The half-lives given in Table 4 for the uranium isotopes were mainly from the recent evaluation by Holden (17); however, for the most important ^{234}U , his downweighting of the latest measurement by Geidelman et al. (18) was not accepted, and the value obtained by Meadows (19) was not used. This, however, changed the result only from 2.455 to $2.456 \cdot 10^5$ yrs. The reason for leaving out the value reported by Meadows is that it was concluded that the material, M-TH, (which figured prominently in the $T_{1/2}$ determination) was too uncertain to be used further as a reference. It was excluded in the present work, and all data measured with the corresponding sample, SST5, were made relative to the first ANL mass scale, U5-S-U4. The half-lives given in Table 4 for the plutonium isotopes were obtained from various sources as referenced in the table.

The isotopic compositions given in Tables 2 and 3 are as reported, or represent averages where several sets of values were available. A value obtained for the ^{234}U content of the KRI material was obtained from the

Table 1. Contributing Laboratories

Laboratory	Samples Provided by	²³⁵ U	²³⁹ Pu
AERE Atomic Energy Research Establishment, Harwell, UK	R. Wiltshire	X	
ANL Argonne National Laboratory Argonne, Illinois, USA Argonne, Idaho, USA	R. Armani	X	X
	J. Meadows	X	X
	S. Carpenter	X	X
BRC Centre d'Etudes de Bruyeres- le-Chatel, Montrouge Cedex, France	G. Grenier	X	
CBNM Central Bureau of Nuclear Measurements, Geel, Belgium, EURATOM	J. Pauwels	X	
KRI Klopkin Radium Institute, Leningrad, USSR	V. Shpakov	X	
LANL Los Alamos National Lab., New Mexico, USA	D. Barton	X	
NBS National Bureau of Standards, Washington, D.C., USA	D. Gilliam	X	X

Table 2. Isotopic Compositions and Specific Activities of the ^{235}U Samples

Material	Isotopic Compositions/wt.%				Specific Activities μmug				
	^{234}U	^{235}U	^{236}U	^{238}U	Isotopic Dilution	Isot. Comp. Half-L. (b)	Colorim.	Others	Average (c)
LANL (a) INS-1	0.0607	99.7457	0.0655	0.1277	13.338 $\pm .024$	13.26 $\pm .13$		13.30 $\pm .08$	13.33 $\pm .02$
NBS INS-1					13.412 $\pm .067$	13.26 $\pm .13$		13.38 $\pm .16$	13.38 $\pm .07$
ANL U5-S-U4	1.027	98.397	0.450	0.125	146.24 $\pm .25$	147.2 $\pm .7$	146.1 $\pm .9$		146.3 $\pm .3$
ANL M-Th	0.852	93.244	0.334	5.570		122.6 (e) $\pm .7$	124.1 (e) $\pm .7$		
KRI U5-P	0.00111 0.00111 (d)	99.9972	0.0017			4.954 $\pm .015$			
AERE U5-UK	1.1104	92.409	0.315	6.165		158.3 $\pm .5$			
CBNM/ BRC U5-NBS	1.6582	97.663	0.1497	0.5296	234.5 ± 2.0	233.9 $\pm .7$			234.0 ± 0.7

(a) Isotopic composition is an average of CBNM, NBS and LANL determinations.

(b) Present values.

(c) Uncertainty limited to lowest uncertainty of individual values.

(d) From present alpha spectroscopy.

(e) Values not used. Mass defined relative to ANL U5-S-U4.

Table 3. Isotopic Compositions and Specific Activities of the ^{239}Pu Samples

Sample	Isotopic Composition /wt.%				Specific Activities/aps μg				
	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	Isot. Comp.(a) & Half-lives	Isotopic Dilution	Rel. U5(b) Thermal	Others	Average(c)
ANL-FNG	99.952	0.048			2297.2 ± 2.3		2317.8 ± 19.2		2297.5 ± 2.3
ANL-Id	94.414	5.264	0.307	0.016	2607.9 ± 2.6		2597.8 ± 36.4		2607.8 ± 2.6
ANL-ZPR	98.939	1.012	0.0491		2358.6 ± 2.4	2359.3 ± 11.8			2358.6 ± 2.4
NBS	99.10	0.883	0.010	0.006	2347.7 ± 2.3	2345.3 ± 9.4		2341.9 ± 19.4	2347.5 ± 2.3

(a) Present values.

(b) Using $(\sigma\text{g})_{\text{U5}}/(\sigma\text{g})_{\text{Pu9}} = 0.7197$.

(c) Uncertainty limited to lowest uncertainty of contributing values.

Table 4. Constants used for the Determination of the Specific Activities

Isotopes	Half-life, Y	Atomic weight, g/mol	Reference
^{234}U	$(2.456 \pm 0.005) \cdot 10^5$	234.0409	See text
^{235}U	$(7.037 \pm 0.011) \cdot 10^8$	235.0439	17
^{236}U	$(2.342 \pm 0.003) \cdot 10^9$	236.0456	17
^{238}U	$(4.468 \pm 0.005) \cdot 10^9$	238.0507	17
^{239}Pu	$(2.4119 \pm 0.0026) \cdot 10^4$	239.0521	20
^{240}Pu	$(6.569 \pm 0.006) \cdot 10^3$	240.0538	20
^{241}Pu	$(1.47 \pm 0.04) \cdot 10^1$	241.0589	20
^{242}Pu	$(3.76 \pm 0.02) \cdot 10^5$	242.0593	20

1 year = 365.25 days

1 mol = $0.60225 \cdot 10^{24}$ atoms

present alpha spectroscopy, and it was found to be in excellent agreement with the values specified by KRI.

The physical descriptions of the samples are summarized in Tables 5 and 6. Knowledge of the chemical compound and approximate thickness of the fissile deposit is required for the calculation of corrections for the total fission-fragment absorption. It should be stressed that the values for the thicknesses of the deposits given in the tables are approximate values (in $\mu\text{g}/\text{cm}^2$), used only for the calculation of the total fission-fragment absorption. Knowledge of the diameter of the fissile deposit, of the material, and of the diameter and the thickness of the backing is required for the calculation of the corrections for transmission and scattering effects. Most sample backings were plain discs; exceptions were the BRC, the KRI U-235-samples, and some ANL samples. The BRC sample backing was a 0.05-cm thick Ta disc with the thickness under the fissile deposit reduced to 0.03 cm. The Cr-Ni ratio and the density of the backing material of the KRI samples is unknown. A 50-50% ratio and a density of $7.9 \text{ g}/\text{cm}^3$ were assumed. The KRI samples were mounted in carrier rings (apparently by means of soldering) as indicated in Fig. 1. The additional amount of solder was unknown and it has been neglected in the calculation of the scattering corrections. The material of the ring was unknown so brass was assumed. The mounting procedure had apparently resulted in positioning the samples slightly and unevenly above and below the top surface of the mounting ring, which was important for determining the alpha-counting-geometry factors. Several of the ANL samples had three 0.35-cm-diameter mounting holes close to the outer edge of the sample backings.

III. Alpha Counting

The alpha-decay rates of all samples were determined with a low-geometry counter. This detector consists of a silicon surface-barrier diode positioned behind an aperture with a diameter of 1.27 cm. Samples were mounted on shelves, with the first shelf at a distance of 4.57 cm from the aperture. The distances of subsequent shelves from the aperture increases this base distance in increments of 5.08 cm. The geometry of this counter (aperture-"lip" thickness, diameter and ovality) was measured repeatedly by different means. Samples obtained for the present intercomparison were counted before and after the fission-ratio experiments. The full-energy spectra were recorded with a 1024-channel analyser, and count rates were obtained by integrating over the $^{238}\text{U} - ^{235}\text{U} - ^{236}\text{U}$ and ^{234}U and the $^{239}\text{Pu} - ^{240}\text{Pu} - ^{241}\text{Pu}$ and ^{242}Pu alpha-energy ranges. Backgrounds that were typically less than 0.3% were subtracted. The ANL samples have been counted repeatedly during the last 10 years. ^{235}U samples with low decay rates (NBS, KRI, ANL-R5, ANL-N3, LANL-S1) and the samples from CBNM were counted with a geometry factor $\sim 1/220$. The ^{235}U samples with sufficiently high decay rates were also counted with a geometry factor of $\sim 1/1000$ (ANL-5-1,

Table 5. ²³⁵U Sample Specifications.

Fissile Sample Deposit						Backing		
Sample	Material	Compound	Dep. Techn.	Diam. cm	Approx. Thickness $\mu\text{g}/\text{cm}^2$ (a)	Material	Thickness cm	Diameter cm
ANL-R5	U5-S-U4	U ₃ O ₈	EP	2.22	20.6	SS	0.0127	4.445
ANL-N3	U5-S-U4	U ₃ O ₈	EP	1.27	41.1	SS	0.0254	1.905
ANL-5-1	U5-S-U4	UO ₂ · H ₂ O	EP	2.49	210.4	SS	0.0254	6.985
ANL-5-2	U5-S-U4		EP	2.50	164.2	SS	0.0254	6.985
ANL-SST5	U5-Th	UF ₄	EV	2.54	81.2	SS	0.0254	6.985
LANL-S1	INS-1	U ₃ O ₈	EV	2.00	95.1	Pt	0.0127	4.763
LANL-S3	INS-1	U ₃ O ₈	EV	2.00	537.9	Pt	0.0127	4.763
NBS25-S-52	INS-1	UO ₂	EV	1.27	182.0	Pt	0.0127	1.905
KRI VI	U5-P	U ₃ O ₈	HFS	2.1	220.7	Cr-Ni	0.010+	2.100+
KRI XV	U5-P	U ₃ O ₈	HFS	2.1	260.2	Cr-Ni	0.010+	2.100+
BRC	U5-NBS	U ₃ O ₈	?	1.2945	85.8	Ta	0.03 (0.05)	2.771
AERE-A	U5-UK	U ₃ O ₈	EV	2.0	110.4	SS	0.0394	2.699
AERE-B	U5-UK	U ₃ O ₈	EV	2.0	110.6	SS	0.0394	2.699
CBMN-33	U5-NBS	UF ₄	EV	1.27	96.0	SS	0.015	1.905
CBMN-36	U5-NBS	UF ₄	EV	1.27	197.0	SS	0.015	1.905

EV = Evaporation, EP = Electroplating, HFS = High Frequency Sputtering,

SS = Stainless Steel

+ = Additional Material due to the Brass Mounting Ring

(a) These are only approximate values used for the calculation of the total fission-fragment absorption.

Table 6. ^{239}Pu - Sample Specifications

Sample	Fissile Sample Deposit				Material	Backing	
	Compound	Dep. Techn.	Diameter cm	Approx. Thickness $\mu\text{g}/\text{cm}^2$		Thickness cm	Diameter cm
ANL-FNG	PuO_2	EP	2.54	25.7	SS	0.0254	6.985
ANL-Id.	PuO_2	EP	1.778	43.5	SS	0.0076	3.429
ANL-ZPR	PuO_2	EP	2.223	21.0	SS	0.0127	4.524
NBS	PuO_2		1.27	82.5	Pt	0.0127	1.905

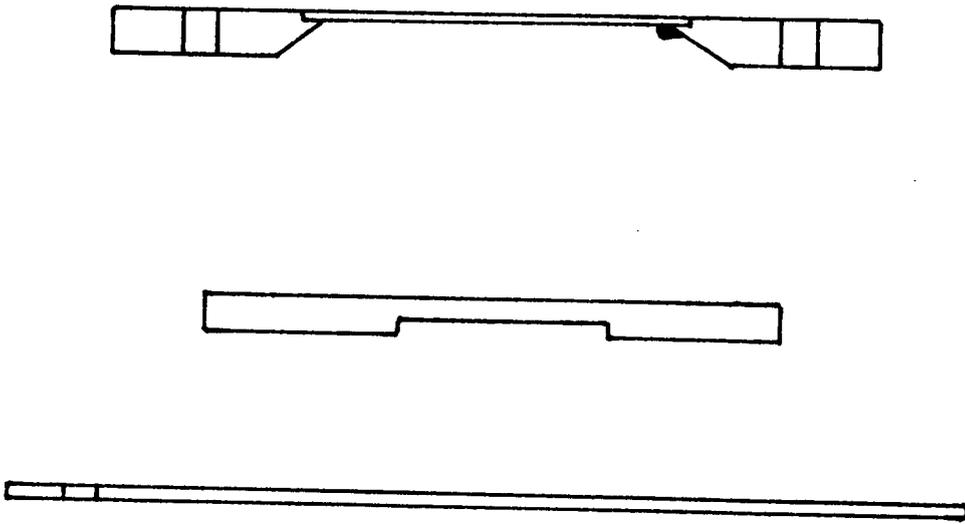


Fig. 1. Schematic of the KRI, BRC, ANL SST5+5-2 Samples.

-5-2 and SST5; LANL-S3, and BRC). The ^{239}Pu samples were counted with various geometry factors ranging from $\sim 1/220$ to $\sim 1/10000$. Some of the ^{235}U samples were also counted in a second low-geometry counter of similar design with a somewhat different aperture and geometry factor (LANL-S3, ANL-R5, -5-2, -SST5, AERE-B, KRI-VI). Geometry factors were determined by means of Monte-Carlo simulations (21) and with a series-expansion approximation (22). Corrections for the aperture "lip" thickness were obtained by Monte Carlo simulations. The decay rates obtained in 1979 were slightly revised for a redetermination of the counter geometry.

The accuracy of the present LG alpha counting has been tested by:

- (a) A comparison of ^{241}Am and ^{243}Am sources between the present LG counter and the second LG counter which resulted in agreement within 0.1%. The agreement for the alpha decay rates of ^{235}U samples involved in the present intercomparison which were counted with both detectors was of similar size.
- (b) The second LG counter was compared with another LG counter at ANL-Idaho and, again, agreement within 0.1% was found.
- (c) Various uranium, plutonium and neptunium samples were counted on different shelves resulting in the following biases for the decay rates determined from counting: 1st. shelf vs. 2nd. shelf of +0.04%; 2nd. vs. 3rd. of -.16%; and 1st vs. 5th of -0.07%.

Representative alpha spectra obtained with the low-geometry counter for the ^{235}U samples are shown in Figs. 2 and 3, and for the ^{239}Pu samples Fig. 4. The spectra for the KRI samples shows that $3.1 \pm 0.1\%$ of the total count rate was due to ^{234}U . This was used to estimate the amount of ^{234}U in these samples. The spectra of the AERE samples contained contributions from alpha emitters with energies above ^{234}U -alpha energies. Their energies and relative decay rates indicated that they were due to the ^{228}Th -decay chain. They contributed $\sim 0.8\%$ of the total count rate for one sample and $\sim 1.1\%$ for the other. One member of this chain, ^{212}Po , has an alpha energy that is outside the range of the present measurements but its contribution can be no more than $\sim 0.2\%$. These impurities required a correction for the measurements with the 2π counter discussed below but did not affect the present LG counting which was restricted to a window spanning the ^{238}U -, ^{235}U -, ^{236}U - and ^{234}U -alpha energy range. None of the spectra showed any alpha groups in this energy region that could not be attributed to the above uranium isotopes. All spectra of the ^{239}Pu samples showed some contributions from ^{241}Am and/or ^{238}Pu decay, both of which were excluded from the integration range.

The ^{235}U samples from ANL were, in addition, counted with a 2π counter for which the calibration factors were known for different thicknesses of uranium on SS backings, as determined with the second LG counter. ^{235}U samples of the same material, deposited on identical backings, were also counted with this 2π counter in order to determine the ratios with negligible statistical uncertainties. This required an additional correction to the count rates of the AERE samples for the impurities, which was based on the spectrum fractions determined with the LG counter.

The results from the present alpha counting and their uncertainties are given in Tables 7 and 8. Statistical uncertainties were 0.2% or less.

Table 7. Results from the Present Alpha Counting of the ^{235}U Samples

Sample	Alpha-decay rate, cps		Uncertainties, %			Present determination of mass, $\mu\text{g U}$
	Quoted (a)	Present	Stat.	Syst. 1 (a)	Syst. 2 (a)	
ANL R5		194.1 \pm 0.6	0.1	0.2	0.2	79.60 \pm 0.29
ANL N3		127.2 \pm 0.4	0.2	0.2	0.1	52.17 \pm 0.19
ANL 5-1		2602 \pm 6	0.1	0.1	0.2	1067 \pm 4
ANL 5-2		2035 \pm 5	0.1	0.1	0.2	834.6 \pm 2.7
ANL SST5		847.8 \pm 1.7	0.1	0.1	0.1	418.1 \pm 1.6
LANL S1		66.52 \pm 0.2	0.2	0.2	0.2	299.4 \pm 1.2
LANL S3		375.1 \pm 1.1	0.1	0.2	0.2	1688.6 \pm 5.7
NBS	50.82 \pm 0.25	50.97 \pm 0.13	0.1	0.2	0.1	228.6 \pm 1.3
KRI VI	62.6 \pm 2.0	62.94 \pm 2.0	0.2	0.2	0.1	762.7 \pm 3.3
KRI XV	74.4 \pm 2.2	73.97 \pm 0.2	0.2	0.2	0.1	896.2 \pm 3.9
BRC		454.9 \pm 1.4	0.2	0.2	0.1	116.7 \pm 0.5
AERE A	912.7 \pm 50 (b)	914.1 \pm 3.2 (b)	0.1	0.2	0.3	346.5 \pm 1.6
AERE B	917.3 \pm 4.6 (b)	914.9 \pm 3.2 (b)	0.1	0.2	0.3	346.8 \pm 1.6
CBNM 33	476.3 \pm 4.1	476.7 \pm 1.2	0.1	0.2	0.1	122.3 \pm 0.5
CBNM 36	976.9 \pm 8.3	977.3 \pm 2.5	0.1	0.2	0.1	250.7 \pm 1.0

(a) See text

(b) Excluding impurities

Table 8. Results from the Present Alpha Counting of the ^{239}Pu Samples

Sample	Alpha-decay rate, $10^5\alpha$ ps		Uncertainties, %			Present determination of mass, $\mu\text{g Pu}$
	Quoted	Present	Start	Syst. 1	Syst. 2	
ANL-FNG	2.981	2.989 ± 0.005	0.1	0.1	0.1	130.1 ± 0.3
ANL-Id.		2.829 ± 0.007	0.2	0.1	0.1	108.5 ± 0.3
ANL-ZPR		1.921 ± 0.003	0.1	0.1	0.1	81.45 ± 0.16
NBS	2.453	2.457 ± 0.004	0.1	0.1	0.1	104.7 ± 0.21

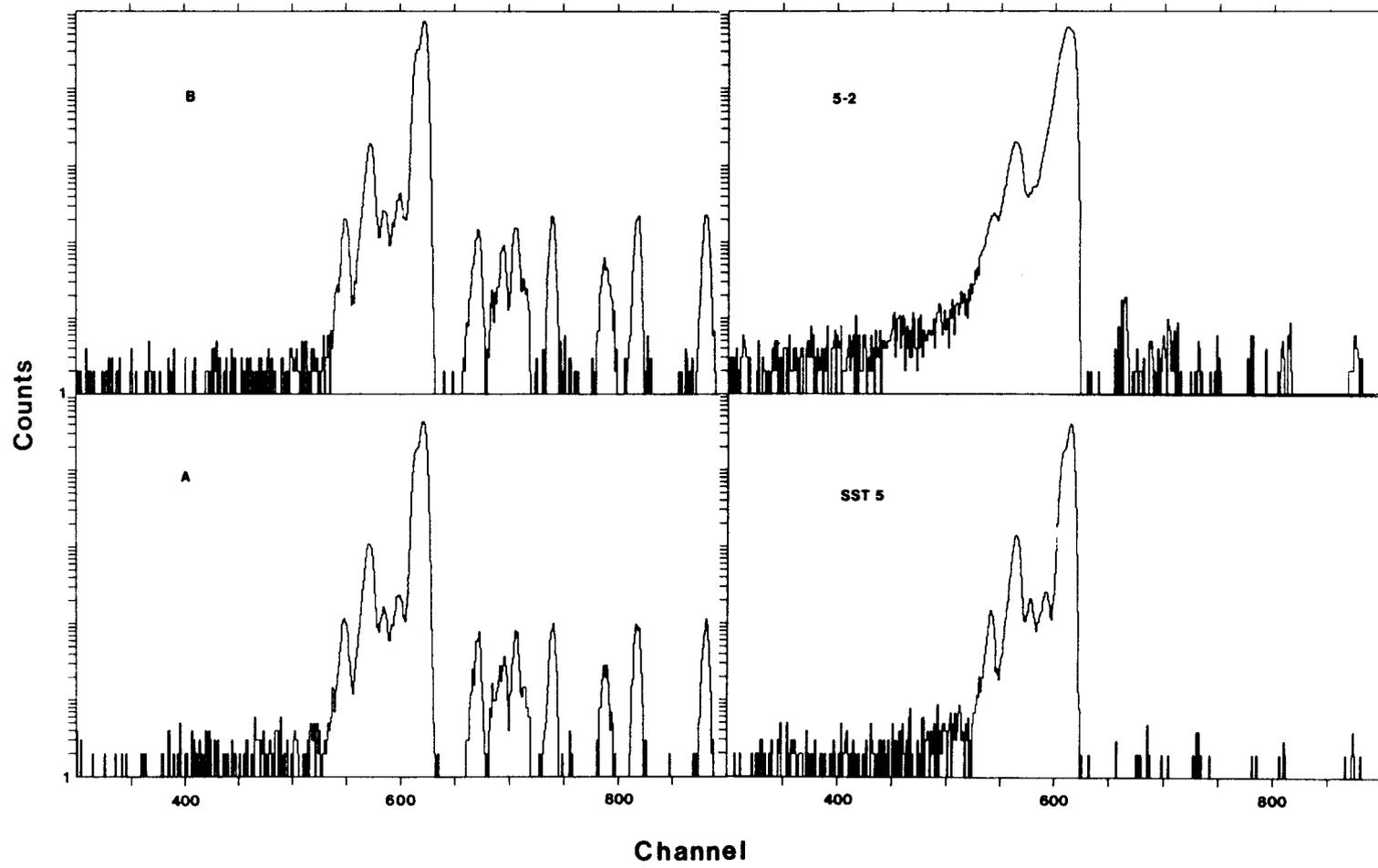


Fig. 2. Representative LG Alpha Spectra for ^{235}U Samples.

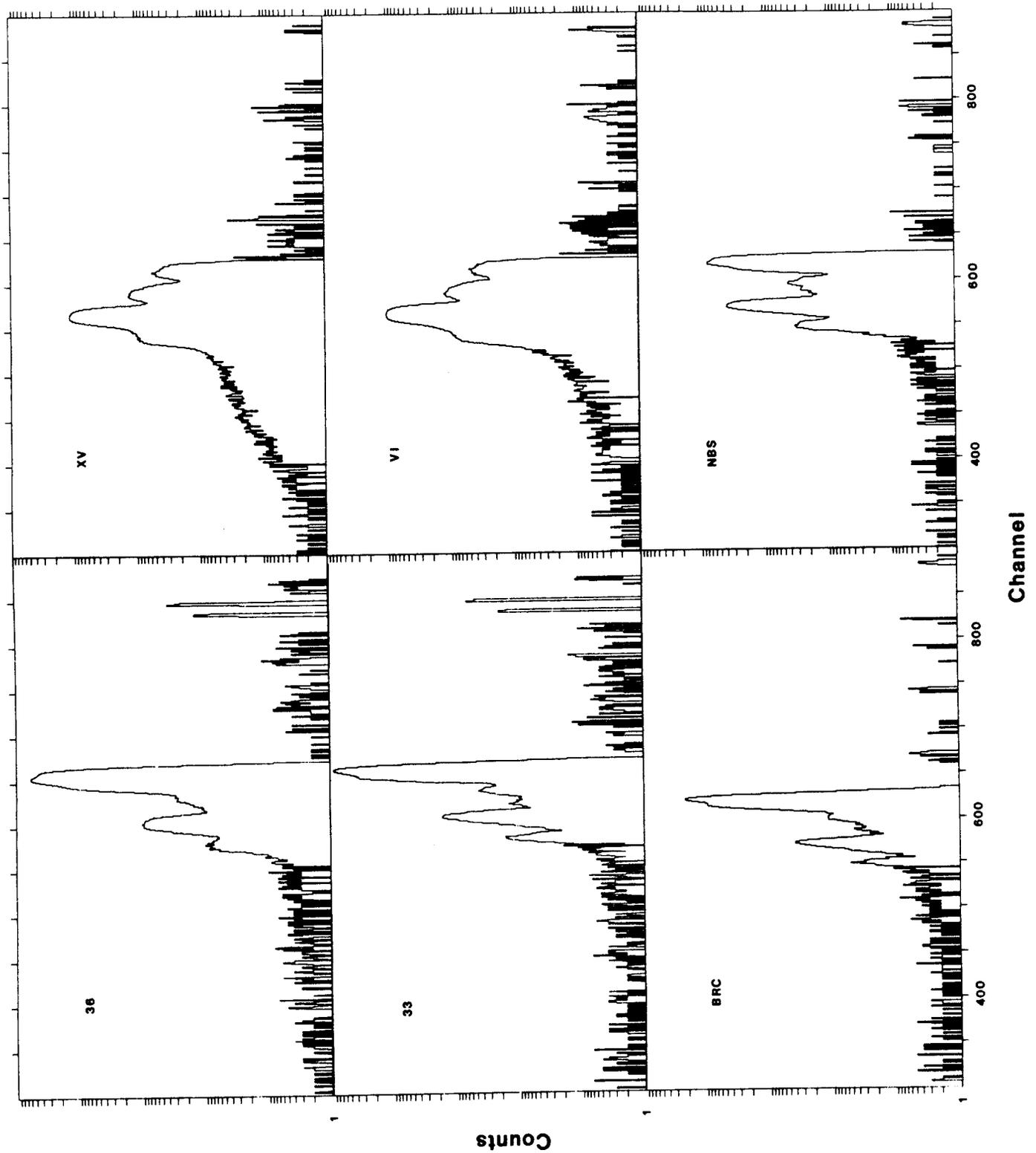


Fig. 3. Representative LG Alpha Spectra for ^{235}U Samples.

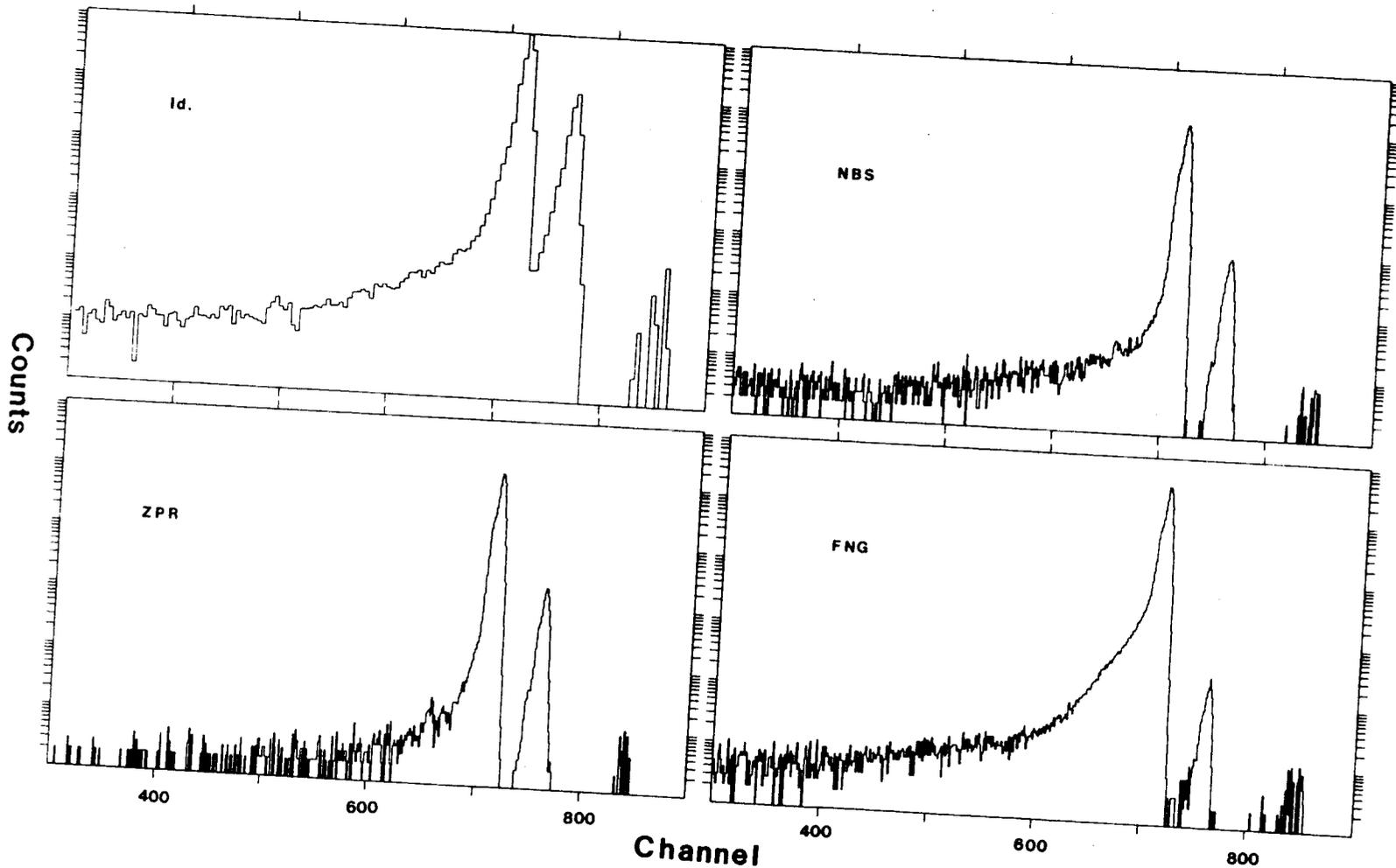


Fig. 4. Representative LG Alpha Spectra for ^{239}Pu Samples.

The systematic uncertainties are determined from the "known" uncertainties of the geometry factor (aperture and sample-deposit diameters, sample-to-aperture distance) and unknown components: 1) nonuniform area densities, which are probably negligible for all but the electroplated samples, 2) sample-backing warping which affects the sample-to-aperture distance, and 3) alpha impurities within the ^{238}U -, ^{235}U -, ^{236}U - and ^{234}U alpha-energy range. Some corrections were applied for sample warping based on measurements with a microscope (BRC, NBS, KRI). Estimates have been made of these uncertainties and included in the given systematic uncertainty components. The nonuniformity of the sample deposit affects mainly the first-shelf geometry factor and the corresponding uncertainties are limited to within the statistical uncertainties of the count rates determined on the first and the second shelves (in those cases where such counts could be obtained).

The present values for the decay rates of the ANL samples are identical with the quoted values, as they include all previous counts. The only other direct alpha-decay rates reported are those by AERE and KRI. The average difference between the present values and the decay rates determined by AERE is 0.2%. The values given in Tables 7 and 8 for the NBS samples were derived from the values quoted by NBS for the alpha-decay rates of its standard-reference samples, and relative measurements by NBS between its reference and the NBS samples used in the present experiments. These ratios were obtained by alpha counting (1X) and by fission counting (2X). Our result agrees with the values from NBS within 0.1% for ^{235}U and 0.2% for ^{239}Pu . The values quoted for CBNM were derived from the given masses based on the alpha counting and the slightly different $T_{1/2}$ used. Agreement between the present counts and those obtained by CBNM is within 0.06%. The values for the absolute uranium and plutonium masses given in Tables 7 and 8 are based on the present alpha-decay rates and the specific activities given in Tables 2 and 3.

IV. Fission-Ratio Measurements

The present fission-ratio measurements were carried out in a back-to-back ionization chamber (23). Measurements were made at (600 ± 100) -keV neutron energy utilizing the $^7\text{Li}(p,n)$ -source reaction and a pulsed and bunched proton beam with a repetition rate of 2 MHz. The samples were located at a distance of 5 cm from the neutron source. A random-pulsar signal was time correlated with the accelerator pulse by requiring a fast coincidence between the 500-nsec.-wide random pulse and the few-nsec.-wide accelerator pulse such that the timing of the resulting random coincidence signal was determined within 500 nsec. by the latter. These coincidence signals were split on an odd-even basis and added to the preamplifiers of the two halves of the ionization chamber. This procedure results in a peak in each of the time-of-flight spectra associated with the two fission samples. The corresponding events were found to be processed by the on-line computer and associated electronics with a better

than 0.1% parity. Identifying tags (pulser, detectors 1 and 2) were used to store 8 time-of-flight spectra (TOF) in the computer. Inspection of these TOF spectra showed some random-coincidence events (~ 0.2%). However, these did not affect the ratio results because they were similar fractions of the primary counts. Different choices of background ranges within the TOF spectra ranges affected the results by < 0.1%. Various test measurements (interchange of detector electronics, measurements at different distances from the target, interchange of detectors, proof of reproducibility) were described in a previous report (15).

Measurements of each of the ratios were carried out for two different orientations of the fission chamber: once with one sample facing the neutron target, then with the other sample facing the target. The midplane of the samples was kept at the same distance from the target. These two sets were obtained with approximately the same statistical uncertainties of typically 0.3%.

Corrections were applied for:

1. Sample distance from the target.

The two samples were separated by the sum of their backing thicknesses and, in some cases, by an additional 0.0127-cm-thick center mounting plate. The required corrections were typically 2-3%, but were substantially larger where the KRI samples were involved (8-10%) because of the increased separation caused by the in brass mounting rings. However, the uncertainty for this correction becomes negligible by averaging the results from the measurements for the two directions of the fission chamber.

2. Transmission losses and scattering gains.

Corrections were applied for the transmission losses which occur for the sample facing away from the target by area-weighting the neutron losses through the contributing structural components. Scattering gains for both samples were computed for the various scattering components with the Monte-Carlo technique, taking into account the angular distributions of the elastically-scattered neutrons and inelastic processes (24). The combined effect of transmission losses and scattering gains on the measured ratios was typically less than 1%. Averaging the measurements for the two directions of the fission chamber results in an effective correction factor of 1.0 for a completely symmetrical arrangement of identical samples. The "residual" correction for transmission and scattering effects for the more common case of asymmetrical samples was typically 0.0-0.3% and was largest for measurements between the ANL 5-2, SST-5 and the KRI samples (0.5%), because these large diameter ANL sample deposits overlap the brass mounting rings of the KRI samples. The uncertainty of the corrections for neutron transmission and scattering effects was assumed to be 50% of the residual corrections.

3. Detection losses below the electronic threshold.

A typical fission-fragment pulse-height spectrum obtained with the ionization chamber which was used in the present experiments is shown schematically in Fig. 5. The distance between a sample and the collector plate determines a geometrical cutoff for the FF-energy deposition. Pulses below this cutoff threshold are due to FF's emitted with large angles, thus losing a large fraction of their energy within the deposit. The threshold for the detection of fission events was set close to the alpha (pile-up) pulses in the pulse-height spectrum. The determination of the fission-pulse losses below this threshold were based upon a constant extrapolation from the pulses above the threshold to zero-pulse height. Though this is probably a good approximation based upon the observed spectra, it is not quite correct as Monte-Carlo calculations for thicker samples show a non-linear shape (25). However, the possible error should be substantially reduced in a ratio measurement and should be negligible if both samples have similar thicknesses. The fractions of pulses below the geometrical cutoff are shown in Figs. 6 and 7.

4. Fission events from isotopes other than ^{235}U .

The present measurements were interpreted to yield total uranium- or plutonium-mass ratios. The primary neutron energy was chosen to result in only small contributions from fission events in isotopes other than ^{235}U . Thus, the corrections depend mainly on the ^{235}U wt. fractions of the materials involved and result in negligible contributions to the uncertainties of the results. The ^{239}Pu wt. fractions were usually large enough to require only small corrections.

5. Angular distribution of the source neutrons.

A correction was applied for the measurements of ratios between samples of different diameters. The evaluation by Liskien et al. (26) was used for the anisotropy of the $\text{Li}(p,n)$ reaction. This correction was most frequently 1.5%, but amounted to 3.7% for ratios between samples with the smallest and largest deposit diameters.

6. Total fission-fragment absorption.

This correction is surely the most important as it is rather uncertain. The present procedure of measuring the ratio with the two directions of the fission chamber averages over the effect of the neutron momentum. The effect of the angular distribution of the fission fragments is small. The major remaining effect is determined by the range of the fission fragments, R , in a specific deposit material. Experimental values of R were known for some of the sample materials (ANL R5, N3, 5-1, 5-2, SST5). Values can also be calculated if the chemical composition is known (e.g. $R = 6.6 \text{ mg U/cm}^2$ for UO_2 , 4.7 for UF_4 , 5.9 for U_3O_8). However, the procedures used in the preparation of such deposits result in uncertainties of the chemical com-

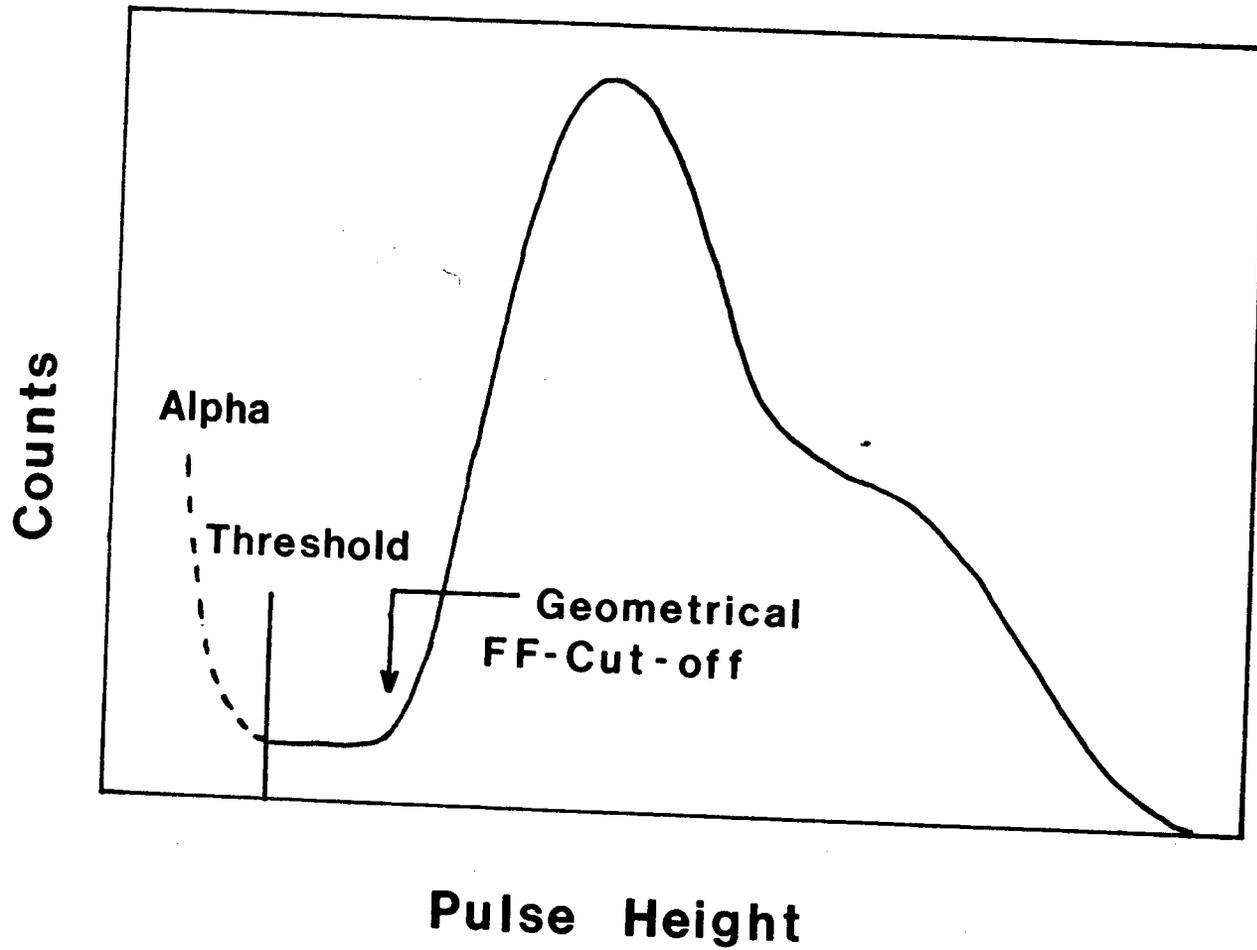


Fig. 5. Schematic of FF Pulse Height Spectrum.

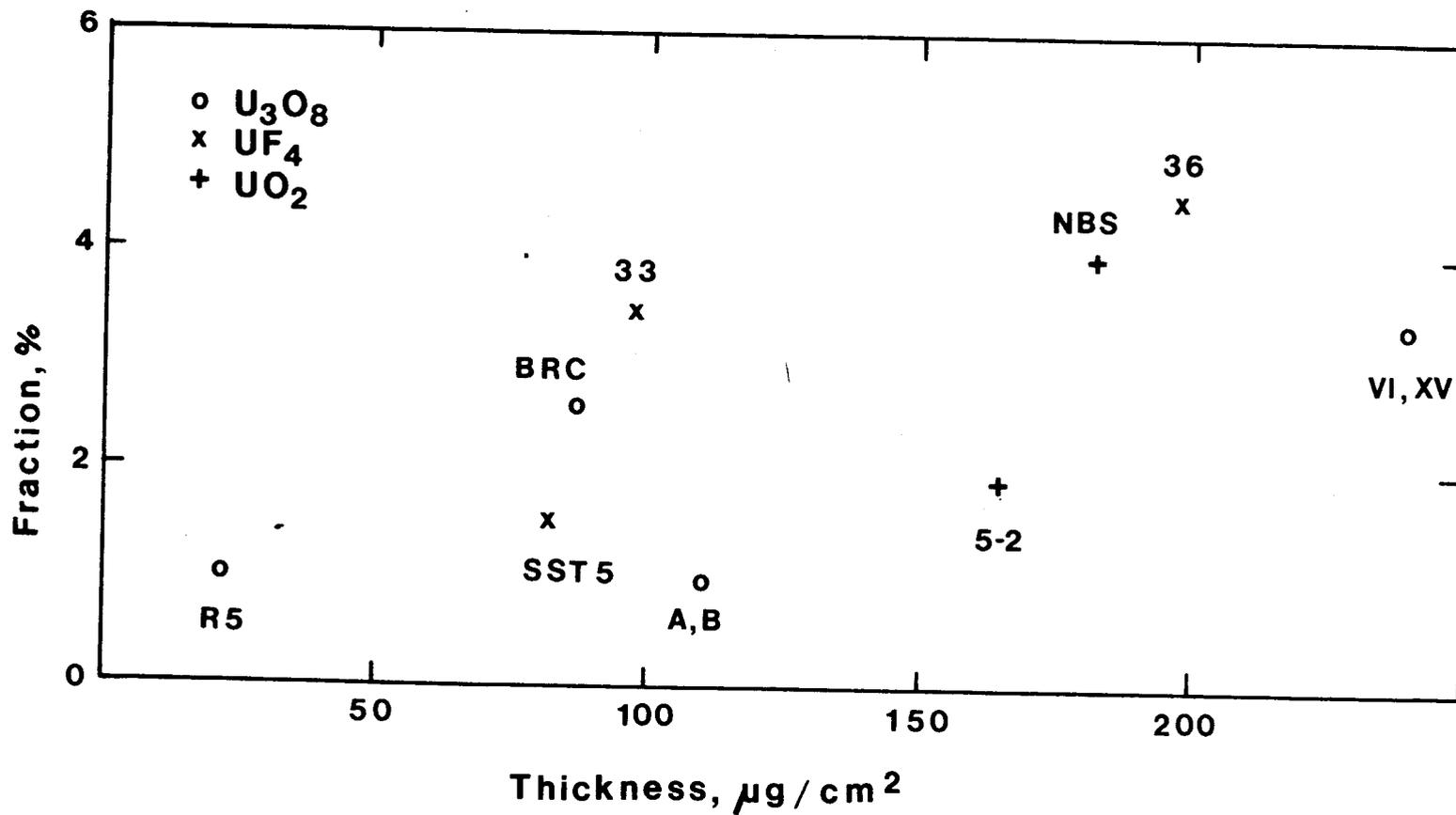


Fig. 6. Fraction of Pulses Below the Geometrical FF Energy Cut-off Threshold for the Uranium Samples.

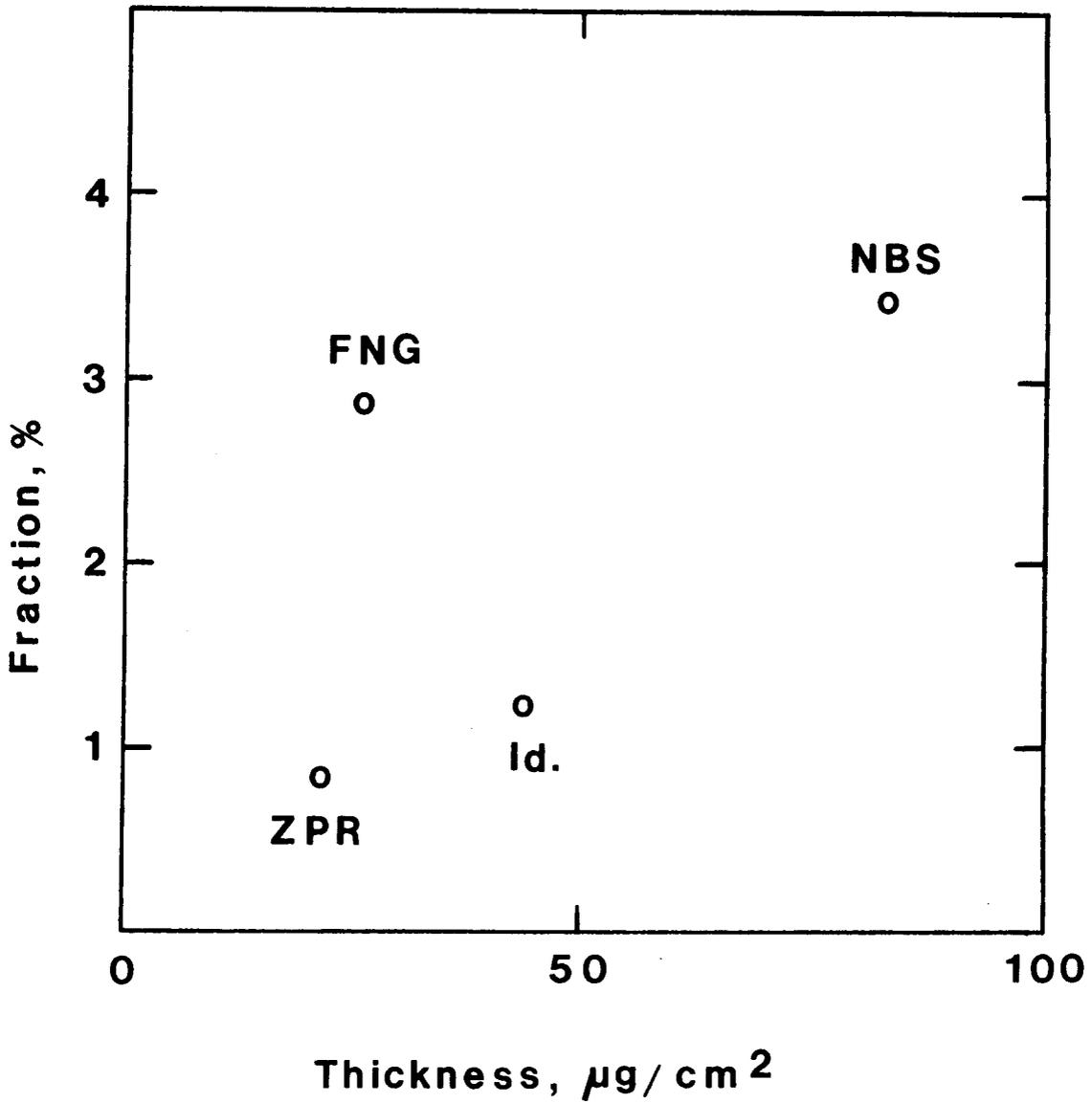


Fig. 7. Fraction of Pulses Below the Geometrical FF Energy Cut-off Threshold for the Plutonium Samples.

position, purity, and structure; and subsequently, the general applicability of unique FF-range values may be questioned. Thus, the following consideration was made: the average energy loss of the 4.397-MeV alpha which occurs with 57% probability in the decay of ^{235}U should indicate to some extent the energy loss of charged particles in an unknown material. The energy loss of these alphas, determined from the energy spread in the low-geometry alpha spectra (detector resolution subtracted), for thin samples is proportional to the sample thickness, δ , thus:

$$\Delta E_{\alpha} \sim \delta$$

The fission-fragment range would be expected to be, in some form, inversely related to the alpha energy loss, therefore

$$\Delta E_{\alpha} \cdot R \sim \delta$$

was considered as a possible empirical relationship. Fig. 8 shows that such a relationship appears to be confirmed for the uranium samples. A range of 7.5 mg U/cm^2 has been used in Fig. 8 for the KRI samples which seems to be identical with the value inferred from corrections applied for FF absorption in such samples (4). Consideration of Fig. 8 indicates that this is an overestimate of the range. The corrections applied in the present work for fission fragment absorption losses in the KRI samples were finally based on the FF ranges which follow from the straight line in Fig. 8. This may not have been the best choice, since the dashed line in Fig. 8 represents the majority of the data better and the consequent failure to explain the difference for the thickest sample, LANL-S3, could be explained with the energy dependence of dE_{α}/ds .

The fission-fragment range alone is not what determines the total FF absorption. The structure of the deposit material (e.g. clumping, flaking) and the smoothness of the backing also affects the total absorption. As pointed out above, pulses below the geometrical-cutoff energy threshold are caused by FF's which were emitted with angles close to 90° , and thus losing most of their energy. Because total FF losses are caused by those fragments which are emitted extremely close to 90° , one would expect that the number of pulses below the geometric cut-off are proportional in first order to the total FF losses -- for a perfect deposit and backing. However, an imperfect deposit and/or backing would cause additional pulses in the low-energy part of the spectrum and additional FF losses not explained by the FF range of the material. This would be expected specifically for thinner samples. The ratio between the fraction of pulses below the geometrical cut-off and the fraction of total FF losses calculated with the ranges for the various uranium materials is shown in Fig. 9. Some features are as expected, for example, the KRI samples appear to have

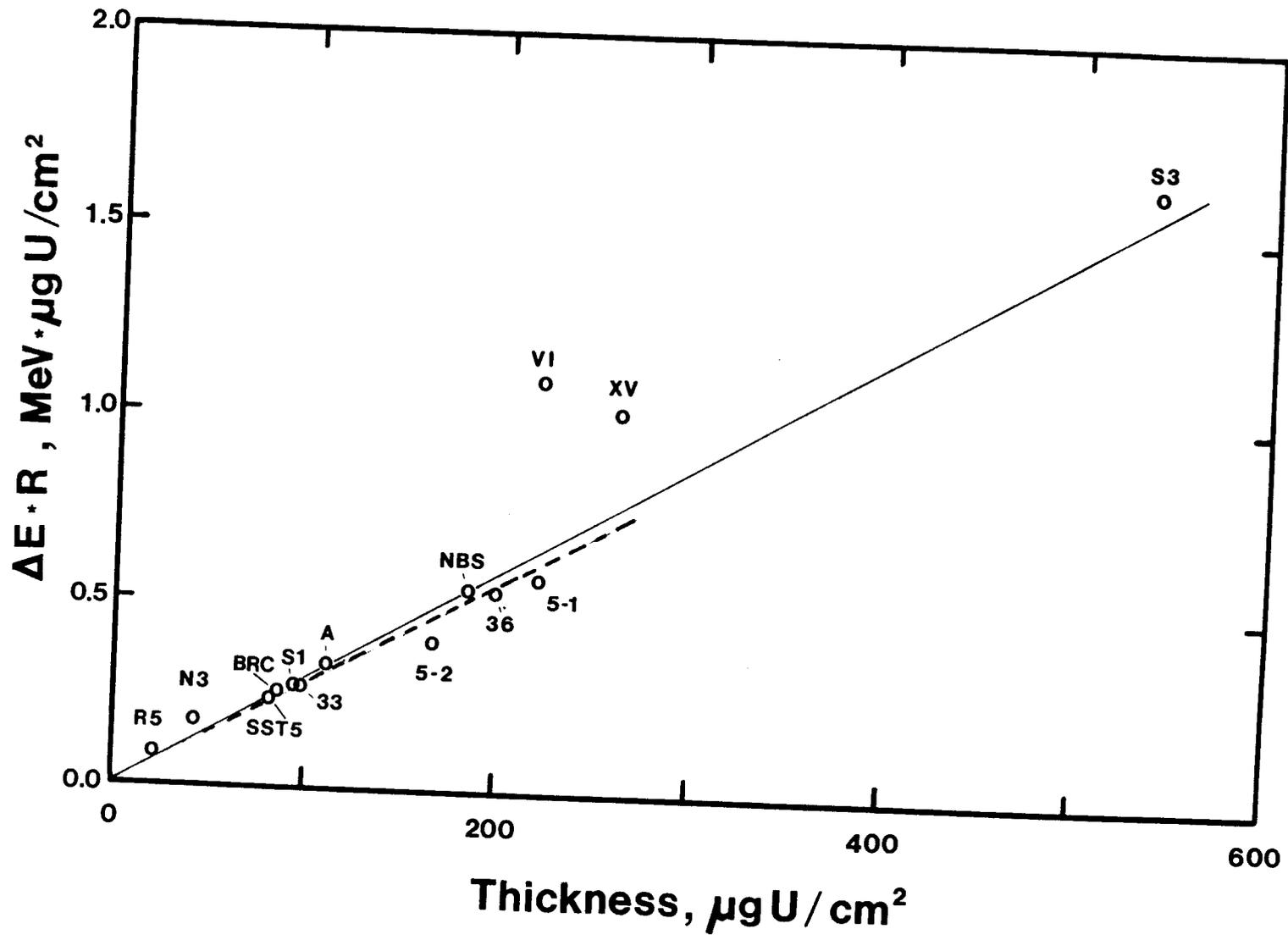


Fig. 8. Alpha Energy Loss Multiplied with the FF Range as a Function of the Sample Thickness. The Values for the KRI Samples is shown Assuming UO_2 .

the best polished backing (based on qualitative inspection under a microscope) and consequently the ratio in Fig. 9 is low. The backings of the ANL samples SST5 and 5-2 had not been polished but appeared to be smooth, though a few large scratches can be observed. The backing of the sample R5 has been polished, but polishing marks are visible, thus it is not surprising to find a high ratio as it is a very thin sample. In most other cases, however, the ratio does not clearly correspond to the merely qualitative nature of the microscope observation, and the figure seems to be inconclusive in regard to required additional corrections. No further action was taken, but measurements for the ANL samples are planned in which the 2π -ionization-chamber-count rates will be compared with LG-FF counting in order to determine the counting losses due to FF absorption.

Fifteen ^{235}U and four ^{239}Pu samples were involved in the inter-comparison, thus measurements of 14 ratios for ^{235}U and 3 for ^{239}Pu would be sufficient to derive the ratio between any two sample masses. A sensible 105 ratios could be measured between the ^{235}U samples and 6 between the ^{239}Pu samples, however, one of the ratio measurements required an average exposure time of 6 hours. As a compromise, a total number of 28 ratios were measured for the ^{235}U samples and 5 for the ^{239}Pu samples. The results from these ratio measurements and their uncertainties are given in Tables 9 and 10.

V. Results and Discussion

V.1. ^{235}U Samples

Best values for the masses of the 15 uranium samples involved in the present intercomparison have been determined based upon the masses quoted by the contributing laboratories, the masses derived from the present absolute alpha counting, and the relative 2π alpha and fission ratio measurements. This overdetermination has been removed by least-squares adjustments

$$\delta = (A^T C^{-1} A)^{-1} A^T C^{-1} M,$$

where A is the coefficient matrix, and C is the variance-covariance matrix of the measurement vector M. Correlations arising from the use of the average specific-activity values of Table 2 for the present absolute mass determinations and from the application of various factors and corrections in the present alpha and fission measurements have been taken into account. The result of the LS analysis and its correlation matrix of them are given in Table 11. The masses quoted for the samples by the contributing laboratories are also given in Table 11. The average differences between the quoted masses and the results of the LS analysis are shown in Fig. 10 in percent. The agreement within $\lesssim \pm 0.3\%$ is very encouraging for future $^{235}\text{U}(n,f)$ -cross-section measurements. The differences are covered in all cases

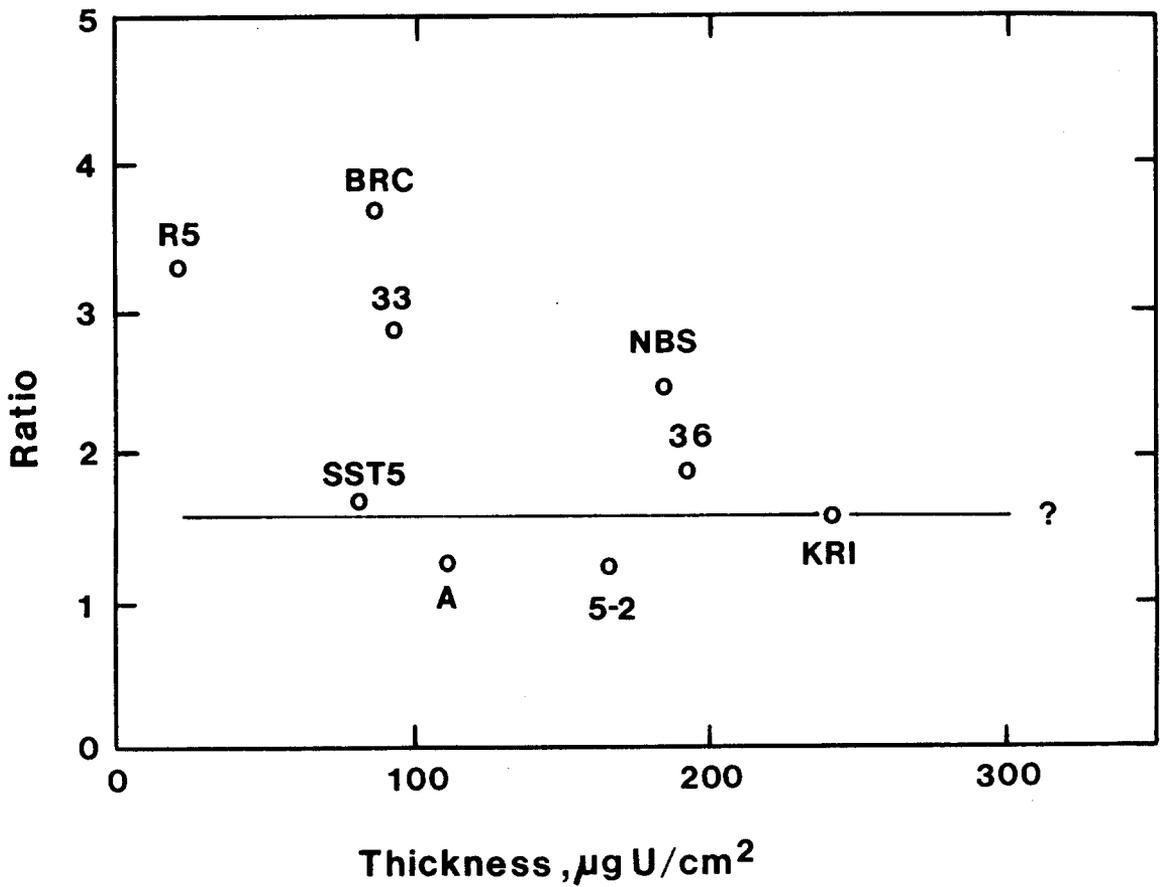


Fig. 9. The Ratios of the Fraction of Pulses Below the Geometrical Cut-off vs. the Fraction of Calculated FF Absorption Losses.

Table 9. Results of the ^{235}U Mass Ratio Measurements

U-Mass Ratio		Uncertainties, % (a)		FFA, Extr.	Source Ang.	Total
		Statistics	Transm., Scatt.			
5-2/SST-5	1.9983	0.2	0.1	0.2	0.0	0.32
R5 /SST-5	0.1902	0.2	0.2	0.2	0.1	0.37
S1 /SST-5	0.7170	0.2	0.2	0.2	0.1	0.37
NBS/SST-5	0.5480	0.2	0.2	0.2	0.3	0.47
N3 /SST-5	0.1242	0.2	0.2	0.2	0.3	0.47
S3 /SST-5	4.0378	0.2	0.2	0.2	0.2	0.41
S1 /5-2	0.3587	0.2	0.2	0.2	0.2	0.41
S1 /R5	3.7766	0.2	0.2	0.2	0.1	0.37
S1 /NBS	1.2984	0.2	0.2	0.2	0.1	0.37
NBS/5-2	0.2726	0.2	0.2	0.2	0.3	0.47
5-1/5-2	1.2805	0.1	0.1	0.1	0.0	0.20
5-2/V1	1.1033	0.2	0.3	0.2	0.1	0.44
NBS/UKA	0.6640	0.2	0.3	0.2	0.2	0.47
NBS/V1	0.3006	0.2	0.1	0.2	0.2	0.37
V1 /UKA	2.1821	0.2	0.1	0.3	0.0	0.39
V1 /BRC	6.5084	0.2	0.1	0.3	0.2	0.44
UKB/SST-5	0.8340	0.2	0.1	0.2	0.2	0.37
XV /SST-5	2.1531	0.2	0.3	0.3	0.1	0.49
R5 /UKA	0.2292	0.2	0.1	0.2	0.1	0.33
BRC/SST-5	0.2794	0.2	0.1	0.2	0.3	0.44
UKA/BRC	2.9681	0.3	0.1	0.2	0.2	0.44
UKA/36	1.3754	0.2	0.1	0.4	0.2	0.51

Table 9. Results of the ^{235}U Mass Ratio Measurements (Continued)

U-Mass Ratio		Uncertainties, % (a)					Total
		Statistics	Transm., Scatt.	FFA, Extr.	Source Ang.		
5-2/33	6.8354	0.2	0.1	0.3	0.3	0.49	
5-2/36	3.3326	0.2	0.1	0.2	0.3	0.44	
UKB/UKA	0.9990	0.3	0.1	0.1	0.0	0.35	
36 /V1	0.3357	0.2	0.2	0.3	0.2	0.48	
XV /V1	1.1771	0.2	0.1	0.2	0.0	0.32	
NBS/BRC	1.9590	0.2	0.2	0.3	0.0	0.42	

(a) The uncertainties for counting statistics, the corrections for neutron transmission and scattering effects, for FF absorption and fission spectra extrapolation, and for the $^7\text{Li}(p,n)$ neutron source anisotropy are given. The total uncertainty contains a 0.1% contribution for the uncertainty of the isotopic composition and fission events in isotopes other than ^{235}U .

Table 10. Results of the ^{239}Pu Mass Ratio Measurements

Pu-Mass Ratio		Uncertainties, %				Total
		Statistics	Transm., Scatt.	FFA, Extr.	Source Ang.	
ANL-Id./ANL-FNG	0.8270	0.3	0.3	0.5	0.3	0.73
NBS/ANL-FNG	0.8140	0.3	0.4	0.5	0.4	0.82
ANL-Id./NBS	1.0205	0.2	0.3	0.4	0.2	0.58
ANL/ZPR/ANL-FNG	0.6441	0.3	0.3	0.6	0.2	0.77
NBS/ANL-ZPR	1.2646	0.3	0.3	0.5	0.3	0.73

Table 11. Comparison of the Quoted ^{235}U Masses with the LS-Fit Results

Samples	U-Mass, μg Quoted	LS-Fit	R5	N3	5-1	5-2	SST5	S1	S3	Correlation Matrix								
										NBS	VI	XV	BRC	A	B	33	36	
ANL -R5	79.60 \pm 0.29	79.32 \pm 0.15	1.00															
-N3	52.17 \pm 0.19	52.03 \pm 0.13	0.39	1.00														
-5-1	1067 \pm 4	1066.4 \pm 2.1	0.45	0.41	1.00													
-5-2	834.6 \pm 2.7	833.7 \pm 1.3	0.51	0.43	0.74	1.00												
-SST5	418.1 \pm 1.6	417.3 \pm 0.7	0.57	0.41	0.47	0.68	1.00											
LANL-S1	298.7 \pm 0.3	298.7 \pm 0.3	0.26	0.15	0.20	0.25	0.21	1.00										
-S3	1688.3 \pm 3.0	1687.5 \pm 2.1	0.19	0.12	0.16	0.20	0.18	0.64	1.00									
NBS	228.5 \pm 1.2	228.6 \pm 0.4	0.29	0.19	0.25	0.31	0.28	0.46	0.31	1.00								
KRI -VI	757.9 \pm 7.6(a)	760.2 \pm 1.4	0.35	0.25	0.33	0.44	0.44	0.20	0.15	0.36	1.00							
-XV	901.0 \pm 9.0(a)	893.0 \pm 1.8	0.31	0.22	0.30	0.40	0.41	0.17	0.13	0.30	0.75	1.00						
BRC	116.1 \pm (b)	116.5 \pm 0.3	0.33	0.22	0.26	0.35	0.41	0.18	0.14	0.31	0.43	0.33	1.00					
AERE-A	345.9 \pm 2.2	346.4 \pm 0.6	0.47	0.27	0.31	0.42	0.50	0.20	0.16	0.32	0.48	0.36	0.53	1.00				
-B	347.7 \pm 2.2	347.2 \pm 0.7	0.43	0.26	0.30	0.41	0.49	0.19	0.15	0.30	0.45	0.35	0.53	0.88	1.00			
CBNM-33	121.9 \pm 0.4	122.2 \pm 0.2	0.21	0.16	0.23	0.31	0.28	0.10	0.08	0.16	0.28	0.24	0.18	0.25	0.23	1.00		
-36	250.0 \pm 0.9	250.7 \pm 0.4	0.24	0.18	0.25	0.34	0.31	0.11	0.09	0.19	0.33	0.28	0.22	0.29	0.27	0.76	1.00	

(a) based on given areal density and total area.

(b) restated for $T_{1/2} (^{234}\text{U}) = 2.456 \cdot 10^5$ yrs.

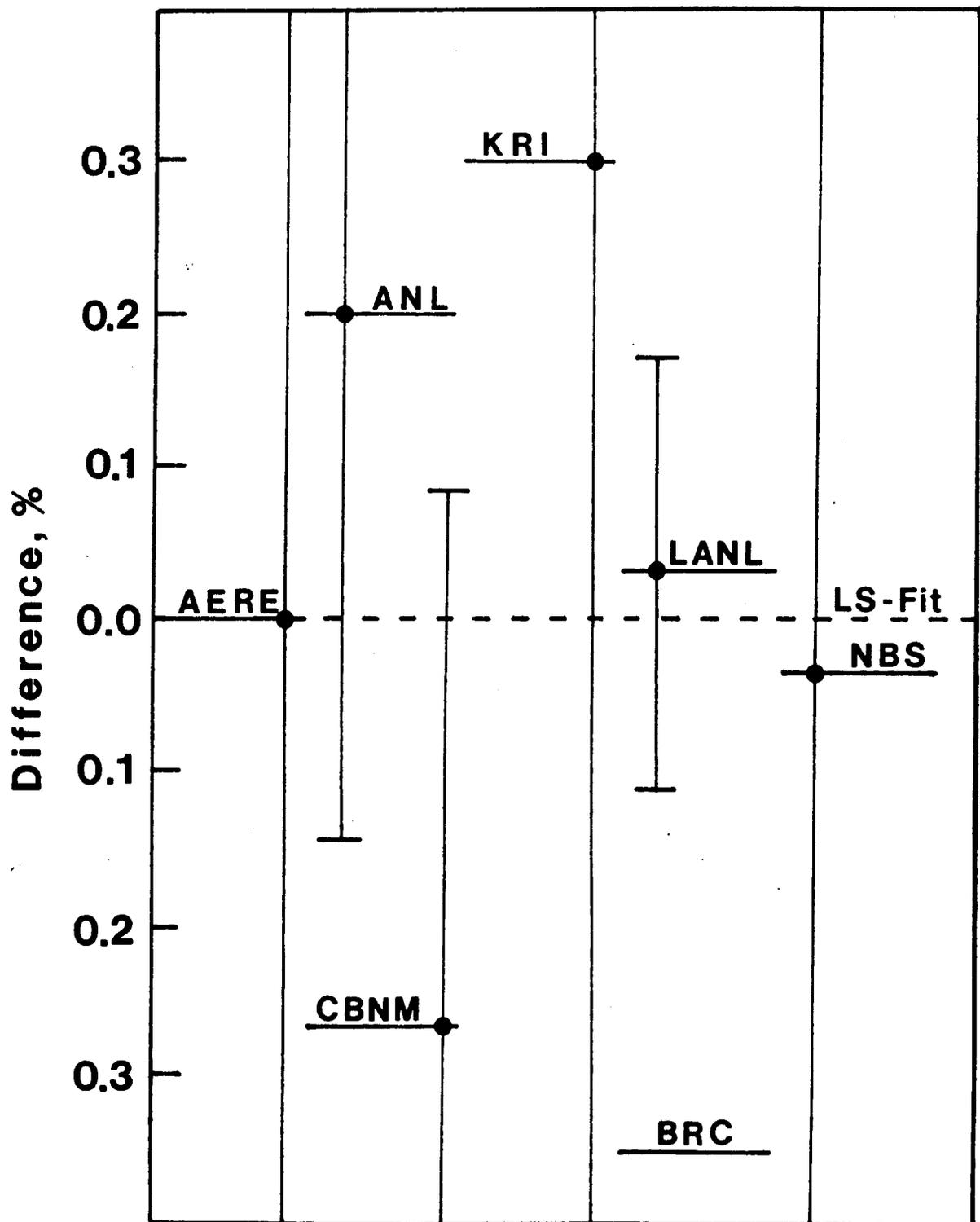


Fig. 10. The Comparison of the Quoted ^{235}U Masses with the Least Squares Fit "Best" Values.

by the quoted uncertainties, which, more often than not, appear to be quite conservative. It is therefore concluded, that ^{235}U masses and fission rate measurements are sufficiently accurate to permit reaction rate measurements in reactor test facilities or in cross sections experiments with the required accuracy.

V.2. The ^{234}U Half-Life.

The ^{234}U half-life is important for the mass assay of ^{235}U samples. Available experimental values are not consistent and a conflict exists with values obtained from fitting thermal parameters. Very accurate values are given for the isotopic compositions of two of the fissile materials involved in the present intercomparison (AERE and CBNM). The absolute alpha-decay rates and absolute masses of the corresponding samples can be used to determine the half-life of ^{234}U . For this purpose, all input data based upon the ^{234}U half-life was removed and the LS fit was repeated. The average value obtained for the half-life of ^{234}U from the four samples (AERE-A,-B; CBNM-33,-36) is

$$T_{1/2}(^{234}\text{U}) = (2.457 \pm 0.005) \cdot 10^5 \text{ yrs.}$$

This is in good agreement with the latest measurement of $2.459 \cdot 10^5$ yrs. by Geidelman et al. (18) as well as with the evaluated value of $2.455 \cdot 10^5$ yrs. by Holden (17), and its modification to $2.456 \cdot 10^5$ yrs. as used in the present work. Inclusion of the present value for the half-life of ^{234}U in the set of values revised by Holden (17), and the modifications discussed in Section II, result in a current value of

$$T_{1/2}(^{234}\text{U}) = (2.4566 + 0.0044) \cdot 10^5 \text{ yrs}$$

which is recommended as the best value to be used.

V.3. ^{239}Pu Samples.

The quoted Pu-sample masses are compared in Table 12 with the values determined from the present absolute alpha counting and values derived from the relative fission ratio measurements and the quoted masses. The uncertainty given for the latter is based upon the spread of the three contributing values, which was found to exceed the estimated uncertainties by a factor of 2. The agreement between the present mass determinations based upon absolute alpha counting and the quoted masses which are based upon a variety of techniques, including isotopic dilution (ANL-ZPR and NBS), is very good. These values probably define the sample masses very well. The poor agreement or the discrepancies for the values derived from the relative fission counting cannot be understood at present. The five measured

ratios given in Table 10 are consistent within $\sim 0.5\%$ or better as can be seen by forming a particular ratio from combinations of other measurements. The measurement of the ratio between the samples with the lowest (ANL-ZRP) and largest (ANL-FNG) masses was repeated with reversed electronic components, but agreement within 0.1% was found. Some correlation between the observed discrepancies and the alpha-decay rates was noted, but it was found to be inconclusive. The most disturbing observation is the rather large fraction of pulses in the FF pulse-height spectra below the geometrical cut-off (see Figs. 5,7) relative to these fractions for the ^{235}U samples (Fig. 6). This might indicate surface roughness which would result in uncorrected FF-detection losses. Indeed, three of the plutonium samples were prepared by electroplating from aqueous solutions whereas the uranium samples were obtained by evaporation or molecular plating from an organic solution. Electroplating of Pu from aqueous solutions appears to result rather frequently in flaking deposits.

It is concluded that ^{239}Pu sample masses can be very well defined by absolute alpha counting. However, ^{239}Pu fission-rate measurements are uncertain by 1-3% and require further investigations, specifically of FF losses in the deposits, and counting losses.

Table 12. ^{239}Pu Sample Masses, in $\mu\text{g Pu}$

	Quoted	Present α -Count.	Av. from Quoted + Rel. Fiss. Count.
ANL-FNG	130.1 \pm 0.5	130.1 \pm 0.3	128.6 \pm 2.4
ANL-Id	108.7 \pm	108.5 \pm 0.3	106.4 \pm 1.6
ANL-ZPR	81.47 \pm 0.4	81.45 \pm 0.16	83.7 \pm 1.0
NBS	104.5 \pm 0.5	104.7 \pm 0.21	105.3 \pm 1.9

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