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U-235 Sample-Mass Determinations and Intercomparisons

by

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I. Introduction

The neutron-induced fission cross section of U-235 is not only one of the most-frequently used references but is also of direct importance in reactor applications. As a consequence, knowledge of this cross section is required with ~1% uncertainty as reflected in corresponding entries in request lists (1,2), which have persisted since the last 10-15 years. Measurements to that level of accuracy require the investigation of the contributing components, one of which is the fission mass. The latter is most often determined by others than the experimenter who measures the differential cross sections or integral reaction-rate ratios in a reactor test facility. The isotopic composition and the sample mass are usually obtained from associated chemistry departments or standard laboratories, however, the experimentor has still the responsibility to assure that the values he uses are adequately described by the quoted uncertainties. This can be achieved by comparing samples from different origins. It was in this spirit that an intercomparison of fission samples obtained from different US laboratories, which were involved in cross section measurements, was carried out in 1979 (3). The notable outcome of this effort was that a bias of~0.7% was found between the standard laboratory and other contributing laboratories (which was, however, within the stated uncertainty). The National Bureau of Standards (NBS) has since then worked on a redefinition of the mass asignments of its reference samples, has revised its mass scale by 0.8%, and has reduced its uncertainty by a factor of two (to about ± 0.5%). However, this new mass scale includes values relative to others. In the present work these have been removed in order to compare mass scales as independent from one another as possible. Independence already appears hard to come by. For example, the Los Alamos National Laboratory (LANL) sample mass specifications are mainly determined by the highest-weight entries which are for the isotopic composition from NBS, and for the specific activity (determined by isotopic dilution) from the Central Bureau for Nuclear Measurements (CBNM). The same material, INS-1, is apparently used by LANL and NBS, and samples obtained from the Centre D'Etudes de Bruyeres le Chatel (BRC) and CBNM for the present work were made with the same material originating from NBS.

One of the interesting developements in recent times in the area of precision measurements has been the 14-MeV-neutron fission cross sections of U-235. The praise has surely to go to Cance and Grenier (4) who first observed and reported values which were substantially lower than the data accepted at that time. These new values were subsequently confirmed by Arlt et al. (5) and later by others (6-10).

The 14-MeV values are not of great interest in applied areas at the present time, however, they have substantial importance because of their impact on the normalization of the evaluated U-235 cross section, as will be discussed in Section V. The very precise 14-MeV values affect the evaluated cross sections at much lower neutron energies and as some inconsistencies appear to emerge it was considered interesting to assure that these inconsistencies are not due to discrepant mass scales used in the various experiments. Consequently, the present authors inquired at the 1979 Knoxville

conference whether one or two of the samples which were used at the Khlopin Radium Institute in Leningrad (KRI) and at the Technical University of Dresden (TUD) for 14-MeV measurements could be made available for an intercomparison. This sample transfer was subsequently arranged by the International Atomic Energy Agency (IAEA) and the present report describes the comparison which was made at Argonne National Laboratory (ANL).

In order to improve on the validity of possible conclusions, the authors had also asked BRC, the AERE Harwell, and CBNM for contributions to this intercomparison. Samples were obtained from these laboratories and included in the present measurements. All samples are described in Section II.

The measurements consisted of two parts. The first part was the determination of the alpha-decay rates of the samples and the derivation of the absolute sample masses from these data. This is described in Section III. The second part was a set of relative fission-rate measurements and is described in Section IV. Updated data from the 1979 measurements are included in Sections III and IV.

The intercomparison of all the mass scales could only be made after all the reference values became available. This exchange of data took place at the present meeting. The results of the intercomparison are discussed in Section V.

II. Sample Descriptions

Six different fissile materials were involved in the 1979 and 1982/83 intercoparisons discussed here. The isotopic compositions and data on the specific activities which were made available or derived in the present work are given in Table I. 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 2. The laboratories which contributed the samples may have used different half-lives. The half-lives given in Table II were mainly from the recent evaluation by Holden (11), however, for the so important U-234, his downweigthing of the latest measurement by Geidel'man et al.(12) was not accepted, and the value obtained by Meadows (13) was not used. This, however, changed the result only from 2.455 to $2.456 \cdot 10^5$ ys. 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 determination by Meadows) 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 isotopic compositions given in Table 1 are as reported, or averages were several values were available. The IC values for the KRI material are as given by KRI. A value for the U-234 content derived from present alpha spectroscopy is in good agreement with the coresponding value from KRI. A material which appears rather similar has been defined in Ref. 10. The coresponding values for the isotopic composition are given in Table 1 in brackets. These values lead to a specific activity which differs by 0.2% from our determination.

The physical descriptions of the samples are sumarized in Table 3. 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. Knowledge of the diameter of the fissile deposit and the material, diameter and thickness of the backing is required for the calculation of the corrections for transmission and scattering effects. The values for the thickness of the deposits given in Table 3 (in $\mu g/cm^2$) are approximate values used for the calculation of the fission-fragment absorption. Most sample backings were plain discs, exceptions were the BRC and the KRI samples. The BRC sample backing was a 0.05-cm thick Ta disc with the thickness under the fissile deposit reduced to 0.03cm. The information on the KRI samples given in Table 3 is from Ref. 5, and as obtained during the present meeting. The Cr-Ni ratio and the density of the backing material is unknown. A 50-50% ratio and a density of 7.9g/cm was assumed. The KRI samples were (apparently by soldering) mounted in carrier rings as indicated in Fig. 1. The additional amount of solder was unknown and has been neglected. The material of the moumting ring is brass. The mounting procedure had apparently positioned 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.

III. Alpha Counting

The alpha-decay rates of all samples were determined with a low-geometry surface-barrier detector. Samples obtained for the present intercomparison were counted before and after the fission ratio experiments. The ANL samples have been counted repeatedly during the last 10 years. Samples with low decay rates (NBS, KRI, ANL-R5,N3, LANL-S1) and the samples from CBNM were counted with a geometry factor of ~1/220. Other samples were also counted with a geometry factor of ~1/1000 (ANL-5-1, 5-2, SST5, LANL-S3, and BRC). Some of the samples were counted in addition 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 with Monte-Carlo simulations and with a series-expansion approximation. Backgrounds of typically less than 0.3% was subtracted. Decay rates from 1979 were slightly revised for a redetermination of the counter geometry.

The accuracy of the present LG alpha counting has been tested: a) the comparison with the second LG counter shows agreement within 0.1%, b) this second LG counter has been compared with another LG counter at ANL-Idaho (agreement within 0.1%), and c) various uranium, plutonium and neptunium sample counts on different shelves have been compared (1st. shelf/2nd. +0.04%, 2nd./3rd. -0.05%, 1st./3rd. -.16%, and 1st./5th. -0.07%).

Representative alpha spectra obtained with the low-geometry counter are shown in Figs. 2 and 3. The spectra obtained for the KRI samples were used to obtain the contribution from the U-234 decay. A fraction of 3.1±0.1% of the total count rate was found. The AERE samples show an 0.8% count-rate contribution from impurities with energies above the U-234-decay alpha energy for one sample and 1.1% for the other. Decay assignments of these impurities indicate the Th-228 decay chain. It is unknown whether additional contributions from impurities are in the U-235 - U-236 - U-234 alpha-energy range.

The ANL samples were also 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. Samples of the same material and on identical backings were also counted with this 2π counter in order to determine the ratios with negligible statistical uncertainties.

The results from the present alpha counting and their total uncertainties are given in Table 4. Statistical uncertainties were 0.2% or less. The systematic uncertainties are determined by 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 U-235 - U-236 - U-234 alpha-energy range. Some corrections were applied for sample warping based on measurements with a microscope (BRC, NBS, KRI) and estimates have been made on these

uncertainties and included in the given systematic uncertainties.

The present values for the ANL samples are identical with the quoted values as they include previous counts. The only other direct alpha-decay rates reported so far are those by AERE and KRI. The average difference of between the present values and the AERE decay rates is 0.2%. The values given in Table 4 for the NBS samples were derived from the value quoted by NBS for the alpha-decay rate of its standard-reference sample and relative measurements by NBS between its reference and the sample NBS 25-S-5-2 used in the present experiments. These ratios were obtained by alpha counting (1X) and by fission counting (2X). Our value agrees with NBS within 0.1%. The values quoted for CBNM were derived from the given masses based on alpha counting and the slightly different $T_{1/2}$ used. Agreement between the present counts and those from CBNM is within 0.06%.

The values for the absolute uranium masses given in Table 4 are based on the present alpha-decay rates and the specific activities given in Table 1. Also listed in Table 4 are the values quoted by the owners of the samples. The agreement between the values from ANL and LANL is within 0.13% implying agreement between the alpha counting at both laboratories within that uncertainty. The LANL values were recently revised by a minor amount ($<0.1\%$). Agreement with NBS is very good after the aforementioned revision of the NBS mass scale by 0.8%. The value given in the Table for NBS is as quoted, thus includes measurements relative to LANL samples. The bias of 0.3% between the present values and those quoted by CBNM is due to the high weight of the values based on isotopic dilution. Agreement with the values based on alpha counting is within 0.14%, the difference being mainly due to the different U-234 half-life values used. The value quoted by BRC is based on the U-234 half life of $2.446\ 10^5$ ys., thus 0.4% of the difference with the present value can be understood with the different half-life values.

VI. Fission Ratio Measurements

It should be clear from the outset that in comparing sample masses of different material and with different backings by fission ratio measurements, one compares a variety of other features of fission-rate measurements besides fission masses. The measured fission rates are proportional to the sample masses, but also to the counting efficiency, e. g. the total fission-fragment absorption is involved.

The present fission-ratio measurements were carried out in a back-to-back ionization chamber (14). Measurements were made at 600±100-keV-neutron energy utilizing the $^{7}Li(p,n)$-source reaction and a pulsed and bunched proton beam. The samples were located at a distance of 5 cm from the neutron source. A random-pulser signal which was time correlated with the accelerator pulse was split on an odd-even basis and added to the two preamplifiers. These 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 ($\sim$0.2%), which did, however, not affect the ratio results. Different choices of background ranges in the TOF spectra did not affect the result either. Various test measurements (interchange of detector electronics, measurements at different distances from the target, interchange of detectors, proof of reproducebility) were described in the previous report (3).

Measurements were carried out for each of the ratios in two steps: once with one sample facing the target, then with the other sample facing the target. These two sets were obtained with approximately the same statistical uncertainties of typically 0.3%.

Corrections were applied for:

1. Sample distances 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 substantially larger where the KRI samples were involved (8-10%). However, by averageing the results from the measurements for the two directions of the fission chamber, the uncertainty for this correction becomes negligible.

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 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. The combined effect of transmission losses and scattering gains on the measured ratios was typically less then 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 asymmetric samples was typically ~ 0.0-0.3% and largest for measurements between the ANL 5-2, SST5 and the KRI samples (0.5%), because the large diameter ANL sample deposits overlap the brass mounting rings of the KRI samples. The uncertainty of the corrections for transmission and scattering was assumed to be 50% of the residual corrections.

3. Detection losses below the electronic threshold.

The threshold for the detection of fission events was set close to the alpha (pile-up) pulses in the pulse-height spectrum. The fission-pulse losses below this threshold were determined based upon a linear extrapolation from the pulses above the threshold to zero-pulse height. Though this is probably a good approximation, it is not quite correct as Monte-Carlo calculations for thicker samples show a none-linear shape (15). However, the possible error should be substantially reduced in a ratio measurement and should be negligible if both samples have similar thicknesses.

4. Fission events from isotopes other than U-235

The present measurements were interpreted to yield total uranium-mass ratios. The primary neutron energy was choosen to result in only small contributions from fission in isotopes other than U-235. Thus, the correction depends mainly on the U-235-wt fractions of the materials involved and results in a negligible contribution to the uncertainty of the ratio result.

5. Angular distribution of the sorce neutrons.

A correction was applied for the measurements of ratios between samples of different diameter. The evaluation by Liskin et al. (16) was used for the anisotropy of the $^{7}Li(p,n)$ reaction. This correction was most frequently 1.5% but 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 the most 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 several of the sample materials (ANL 5-2, 5-1, SST5, NBS). Values can also be calculated if the chemical composition is known (e. g. R=6.6 mg U/cm² for UO_2, 4.7 for UF_4, 5.9 for U_3O_8). However, the material of the KRI samples was unknown by the present authors until the present meeting took place. 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 U-235 should indicate to some extend the energy loss of charged particles in an unknown material. The energy loss of these alphas, determined from the energy spead (detector resolution subtracted), in the low-geometry alpha spectra is proportional to the sample thickness, δ, (for thin samples), 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, searching for an empirical relationship with the help of the many other samples for which the range was known. Fig. 3 shows that the relationship appears to be linear and clearly indicates that the assumption of a range of 7.5 mg U/cm^2 for the KRI samples was wrong. The fission fragment absorption losses of the KRI samples were finally determined based on the FF ranges which follow from the straight line in Fig. 3. This may not have been the best choice, the dashed line in Fig. 3 represents the majority of the data better and the consequent failure to explain the heaviest sample S3 could be accepted based on the energy dependence of dE_α/ds.

The fission-fragment range alone is not what determines the total absorption. The structure or smoothness of the backing affects in addition the total absorption to be accounted for. Consideration of the geometry of the ionization chamber leads to the understanding of the observed pulse-height spectrum: the sharp drop from the maximum in the pulse-height spectrum toward lower pulse heights comes from a "geometrical" cut-off of the FF due to the collector plate. Smaller pulses are from FF's emmitted with angles close to 90°, thus loosing most of their energy. Because total FF losses are caused by those emmitted extremely close to 90°, one would expect that the number of pulses below the geometric cut-off are in first order proportional to the total FF losses -- for a perfect backing. However, an imperfect 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 specifically expected 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 materials is shown in Fig. 4. Some features are as expected, for example, the KRI samples appear to have the best polished backing (based on qualitative inspection under a microscope) and the ratio in Fig. 4 is consequently low. The backings of the ANL samples SST5 and 5-2 had not been polished but appear to be smooth though a few larger 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 as to required additional corrections. No further action was taken, but measurements are planned for the ANL samples in which the 2π-ionization-chamber count rates will be compared with LG-FF counting.

V. Results and Discussion

Fifteen samples were involved in the present intercomparison, thus measurements of 14 ratios would be sufficient to obtain the ratio between any two sample masses. A sensible 105 ratios could be measured between the 15 samples, however, one of the ratio measurements took about an average of 6 hours and a total of 28 ratios was measured. This overdetermines the number of unknowns by a factor of 2. A consistent set of 14 unknowns can be derived with 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. This has been simlified with C - I, the identity matrix, thus neglecting the correlations:

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

The corresponding results are given in Table 6. Measured values are identified by the % difference between the measured and the consistent value. Besides the 28 fission ratio measurements (round brackets) additional 10 ratios derived from the alpha counting were included in the consistency fit (winged brackets). The latter were confined to ratios between samples of the same material with the exception of two ratios where materials were involved for which the isotopic composition was exceptionally well known.

The uncertainties of the input data were typically 0.3-0.5%. The uncertainties of the results from the present measurements given in Table 6 are typically 0.2-0.3%. The results from the present ratio measurements can be used to determine absolute sample masses either based upon the values derived from the present alpha counting or with masses quoted by the owners of the samples. Both types of data are given for each sample in the Appendix.

Comparison of all four values which can be obtained for the mass of each sample from:

1) The mass quoted by the owners of the sample,
2) The mass determined from the present alpha counting,
3) The mass determined from the present ratio measurements relative to all other sample masses and the masses determined by the present alpha counting
4) The mass determined as under 3), but using the masses quoted by the owners of the samples,

are typically within a range of ±0.3% or better, thus indicate a better knowledge of the sample masses than the quoted uncertainties. Knowledge of the U-235 sample mass in a cross section measurement or reaction-rate-ratio measurement in a reactor within 0.3% is considered sufficient.

One of the conclusions of the 1979 intercomparison was that the U-234 half life may be the source of some of the inconsistencies noted

at that time. Very accurate values were available for the isotopic compositions of two of the fissile materials involved in the present intercomparison (AERE, CBNM). The ANL, LANL, NBS, and CBNM mass scales are mainly determined by independent isotopic dilution measurements (though the isotopic dilution measurement for the LANL samples was done at CBNM, this was quite some time ago). Thus, the half life of U-234 can be determined from the present alpha decay rates for the AERE and CBNM samples and their masses based on the ratio measurements and the quoted masses for the ANL, LANL, NBS, and CBNM samples. The value is

$$T_{1/2}(\text{U-234}) = (2.4595 \pm 0.00) \times 10^{5} \text{ yrs.}$$

which is in very good agreement with the latest measurement by Geidel'man et al. (12):

$$T_{1/2}(\text{U-234}) = (2.459 \pm 0.007) \times 10^{5} \text{ yrs.}$$

It is concluded from the present investigations, that U-235 sample masses are well enough known for future measurements and have not been a source of errors in recent high accuracy measurements. However, corrections for total ff absorption may have been too low. The U-234 half life is now known with sufficient accuracy to determine sample masses of spiked U-235 material to within 0.3%.

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Table 1. Isotopic Compositions and Specific Activities

	Isotopic Compositions/wt%				Specific Activities/αpmpμg				
Material	U-234	U-235	U-236	U-238	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 ± .024	13.26 ± .13	-----	13.30 ± .08	13.33 ± .02
NBS INS-1					13.412 ± .067	13.26 ± .13	-----	13.38 ± .16	13.38 ± .07
ANL U5-S-U4	1.027	98.397	0.450	0.125	146.24 ± .25	147.2 ± .7	146.1 ± .9		146.3 ± .3
ANL ~~(e)~~ M-Th	0.852	93.244	0.334	5.570		122.6 (e) ± .7	124.1 (e) ± .7		
KRI ~~(d)~~ U5-P	0.00111 (d) (0.0010	99.99889 99.9955	0.0017 (?) 0.0035)(f)	?		4.952 ±.015 (4.941)			
AERE ~~UK~~ U5-92	1.1104	92.409	0.315	6.165		158.3 ± .5			
CBNM/ BRC U5-97	1.6582	97.663	0.1497	0.5296		233.9 ± .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.
(f) From Ref. 10. Wt% assumed. It is not sure that this is the same material.

Table 2. Constants used for the Present Specific-activity Determinations.

Isotope	Half-life, Y	Atomic weight, g/mol
U-234	$2.456 \cdot 10^5$	234.0409
U-235	$7.037 \cdot 10^8$	235.0439
U-236	$2.342 \cdot 10^7$	236.0456
U-238	$4.468 \cdot 10^9$	238.0507

1 mol = $0.60225 \cdot 10^{24}$
1 year = ~~235~~ 365.25 days

Table 3. Sample Specifications.

Fissile Sample Deposit						Backing		
Sample	Material	Compound	Dep. Techn.	Diam. cm	Approx. Thickness $\mu g/cm^2$	Material	Thickness Cm	Diameter Cm
ANL-R5	U5-S-U4	U_3O_8	EP	2.22	20.6	SS	0.0127	4.445
ANL-N3	U5-S-U4	U_3O_8	EP	1.27	41.1	SS	0.0254	1.905
ANL-5-1	U5-S-U4	$UO_2 \cdot H_2O$	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_3O_8	EV	2.00	95.1	Pt	0.0127	4.763
LANL-S3	INS-1	U_3O_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_3O_8 ~~UO_2 ?~~	HFS	2.1	220.7	Cr-Ni	0.010+	2.100+
KRI XV	U5-P	U_3O_8	HFS	2.1	260.2	Cr-Ni	0.010+	2.100+
BRC	U5-NBS	U_3O_8	?	1.2945	85.8	Ta	0.03 (0.05)	2.771
AERE-A ~~HAR-A~~	U5-UK	U_3O_8	EV	2.0	110.4	SS	0.0394	2.699
AERE-B ~~HAR-B~~	U5-UK	U_3O_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

cm

EV = Evaporation, EP = Electroplating, HFS = High Frequency Sputtering,
SS = Stainless Steel
+ = Additional Material due to the Brass Mounting Ring

Table 4. Results from the Present Alpha Counting.

Sample	Alpha Decay Rate, αps		Sample Masses, μg Uranium	
	Quoted	Present	Quoted	Present
ANL R5		194.1 ± 0.6		79.60 ± 0.29
ANL N3		127.2 ± 0.4		52.17 ± 0.19
ANL 5-1		2602 ± 6		1067 ± 4
ANL 5-2		2035 ± 5		834.6 ± 2.7
ANL SST5		847.8 ± 1.7		418.1 ± 1.6
LANL S1		66.52 ± 0.2	298.7 ± 0.3	299.4 ± 1.2
LANL S3		375.1 ± 1.1	1688.3 ± 3.0	1688.6 ± 5.7
NBS	50.66 ± .25 (a1) 51.00 ± .25 (a2)	50.97 ± .13	228.5 ± 1.2	228.6 ± 1.3
KRI VI	62.6 ± 2.0	62.94 ± 0.2 (b)	758 ± 25 757.9 ± 7.6 (e)	762.7 ± 3.3
KRI XV	74.4 ± 2.2	73.97 ± 0.2 (b)	901 ± 30 901.0 ± 9.0 (e)	896.2 ± 3.9
BRC		454.9 ± 1.4	115.6	116.7 ± 0.5
AERE A ~~HAR~~ A	971.2 ± 4.6	914.1 ± 3.2 (c)	343.4 ± 2.7 (d)	346.5 ± 1.6
AERE B ~~HAR B~~	915.6 ~~927.3~~ ± 4.6	914.9 ± 3.2 (c)	345.1 ± 2.8 (d)	346.8 ± 1.6
CBNM 33	476.3 ± 4.1	476.7 ± 1.2	122.1 ± 1.0 (1) ~~121.7~~ 121.9 ± 0.4 (2)	122.3 ± 0.5
CBNM 36	976.9 ± 8.3	977.3 ± 2.5	250.4 ± 2.1 (f) ~~249.6 ± 3.3 (f)(g)~~ 249.9 ± 0.9 (2)	250.7 ± 1.0

(a) Obtained from NBS ratio measurement relative to NBS standard by (1) alpha counting, (2) fission counting.
(b) Including the 3.1% contribution from U-234.
(c) Excluding contributions from impurities with alpha-energies above 4.77 MeV.
(d) Preliminary.
(e) Based on given areal density and total area.
(f) Based on alpha counting and specific activity.
(g) Based on isotopic dilution

Table 5. Results from the Present Ratio Measurements.

Numinator

Denominator	R5	N3	5-1	5-2	SST5	S1	S3	NBS	KRI VI	KRI XV	BRC	HAR A	HAR B	CBNM33	CBNM36
ANL R5	-	0.6562	13.461	10.515	5.2576	3.765 (+.3)	21.231	2.8828	9.5904	11.249	1.4700	4.3725	4.3791	1.5421	3.1656
ANL N3	1.5240	-	20.53	16.033	8.0128	5.738	32.357	4.3934	14.616	17.143	2.2404	6.6627	6.6759	2.3502	4.8245
ANL 5-1	0.0743	0.0487 (+.4)	-	0.7816	0.3906	0.2797	1.5773	0.2142	0.7125	0.8357	0.1092	0.3248	0.3253	0.1146	0.2352
ANL 5-2	0.0951 (+.3)	0.0624	1.2795 (+.1)	-	0.4998	0.3579 (+.2)	2.0182	0.2740 (-.5)	0.9117	1.0693	0.1397	0.4156	0.4163	0.1466	0.3009
ANL SST5	0.1902 (0)	0.1248 (-.5)	2.5602	2.0009 (-.1)	-	0.7161 (+.1)	4.0382 (0)	0.5483 (-.1)	1.8241	2.1395 (+.2)	0.2796 (-.1)	0.8315	0.8329 (+.3)	0.2933	0.6021
LANL S1	0.2652	0.1743	3.5752	2.7941	1.3965	-	5.6402	0.7657	2.5473	2.9877	0.3904	1.1612	1.1631	0.4096	0.8408
LANL S3	0.0471	0.0309	0.6340	0.4955	0.2476	0.1773 (0)	-	0.1358	0.4517	0.5298	0.0692	0.2059	0.2063	0.0726	0.1491
NBS	0.3469	0.2276	4.6693	3.6496	1.8238	1.3060 (-.6) (+.1)	7.3649	-	3.3256	3.9021	0.5099	1.5165	1.5191	0.5349	1.0981
KRI VI	0.1043	0.0684	1.4035	1.0969 (+.1)	0.5482	0.3926	2.2138	0.3007 (-.6)	-	1.1729 (-.2) (+.3)	0.1533	0.4558	0.4566	0.1608	0.3306 (+.8)
KRI XV	0.0889	0.0583	1.1966	0.9352	0.4674	0.3347	1.8875	0.2563	0.8526	-	0.1307	0.3886	0.3893	0.1371	0.2814
BRC	0.6803	0.4464	9.1567	7.1563	3.5765	2.5612	14.443	1.9612 (0)	6.5247 (+.2)	7.6520	-	2.9742 (0)	2.9789	1.0490	2.1538
HAR A	0.2287 (-.1)	0.1501	3.0790	2.4063 (0)	1.2026	0.8612	4.8565	0.6594 (-.7)	2.1938 (-.1)	2.5731	0.3362	-	1.0017 (-.3) (+.1)	0.3527	0.7242
HAR B	0.2284	0.1498	3.0738	2.4023	1.2006	0.8598	4.8484	0.6583	2.1901	2.5687	0.3357	0.9983	-	0.3521	0.7229
CBNM 33	0.6485	0.4255	8.7289	6.8212 (+.3)	3.4095	2.4415	13.768	1.8694	6.2192	7.2946	0.9533	2.8350	2.8398	-	2.0527 (-.1)
CBNM 36	0.3159 (+.2)	0.2073	4.2521	3.3230 (+.3)	1.6609	1.1893	6.7069	0.9106	3.0248	3.5534	0.4643 (+.2)	1.3809 (-.4)	1.3833	0.4872	-

34135

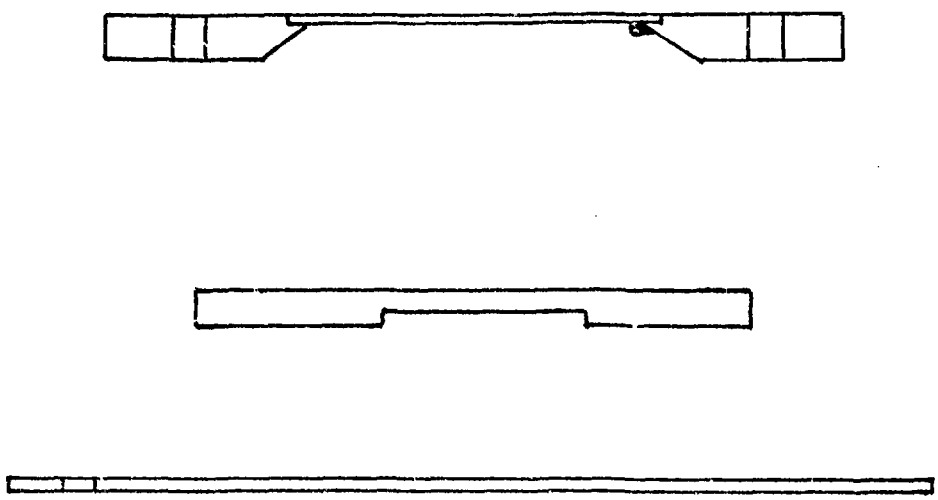

Fig. 1. Schematic of the KRI, BRC, ANL SST5+5-2 Samples.

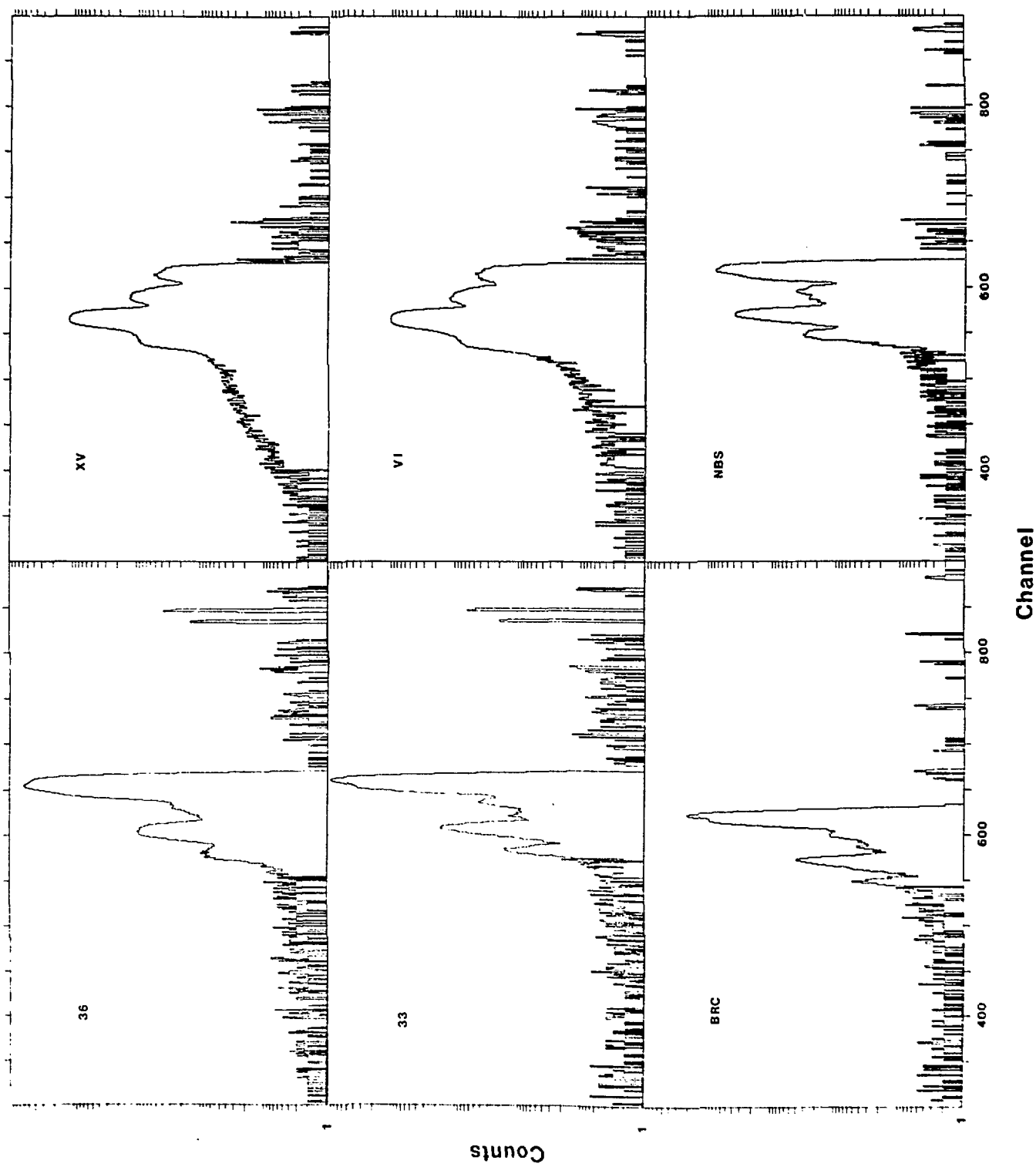

Fig. 2. Representative LG Alpha Spectra.

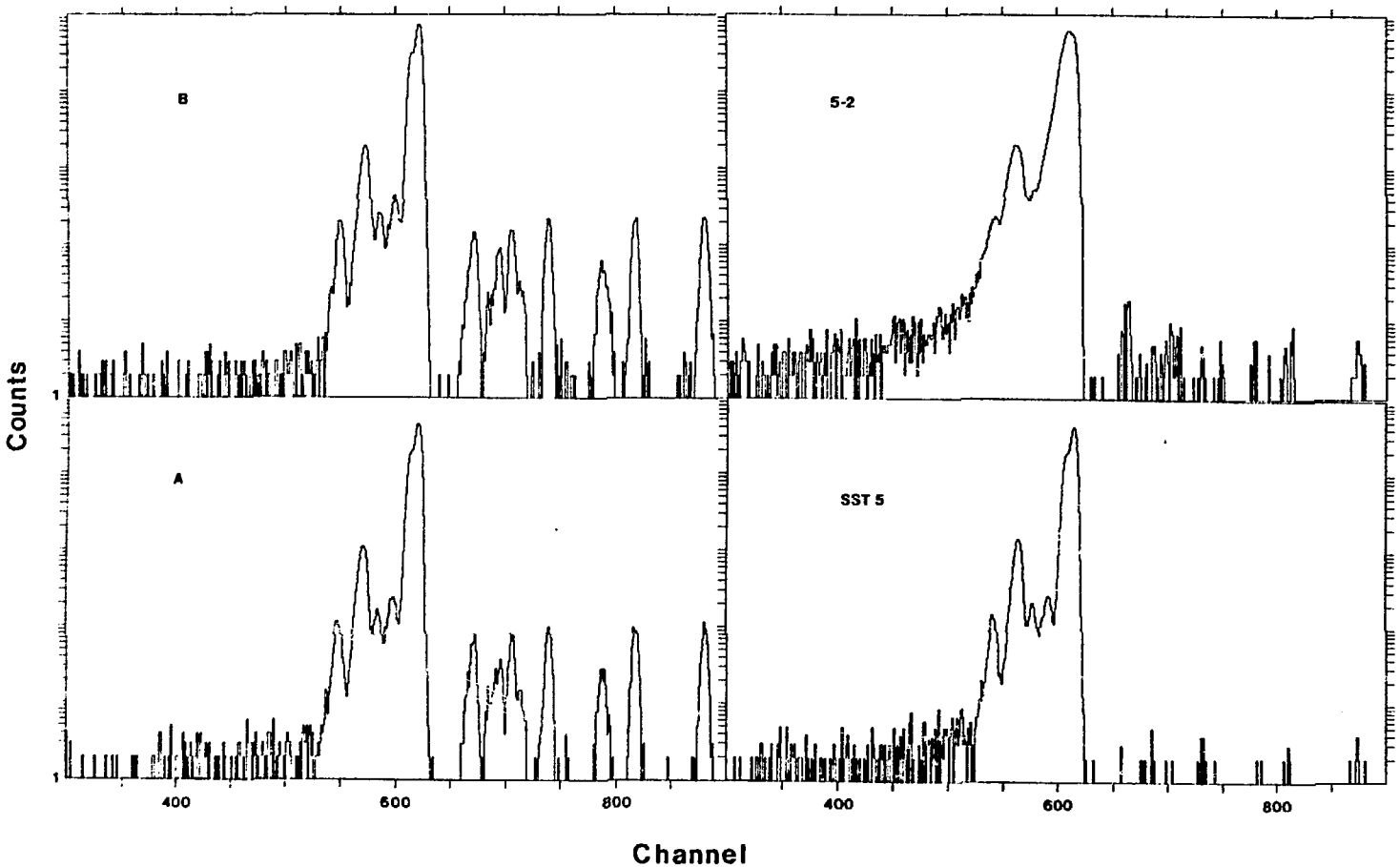

Fig. 3. Representative LG Alpha Spectra.

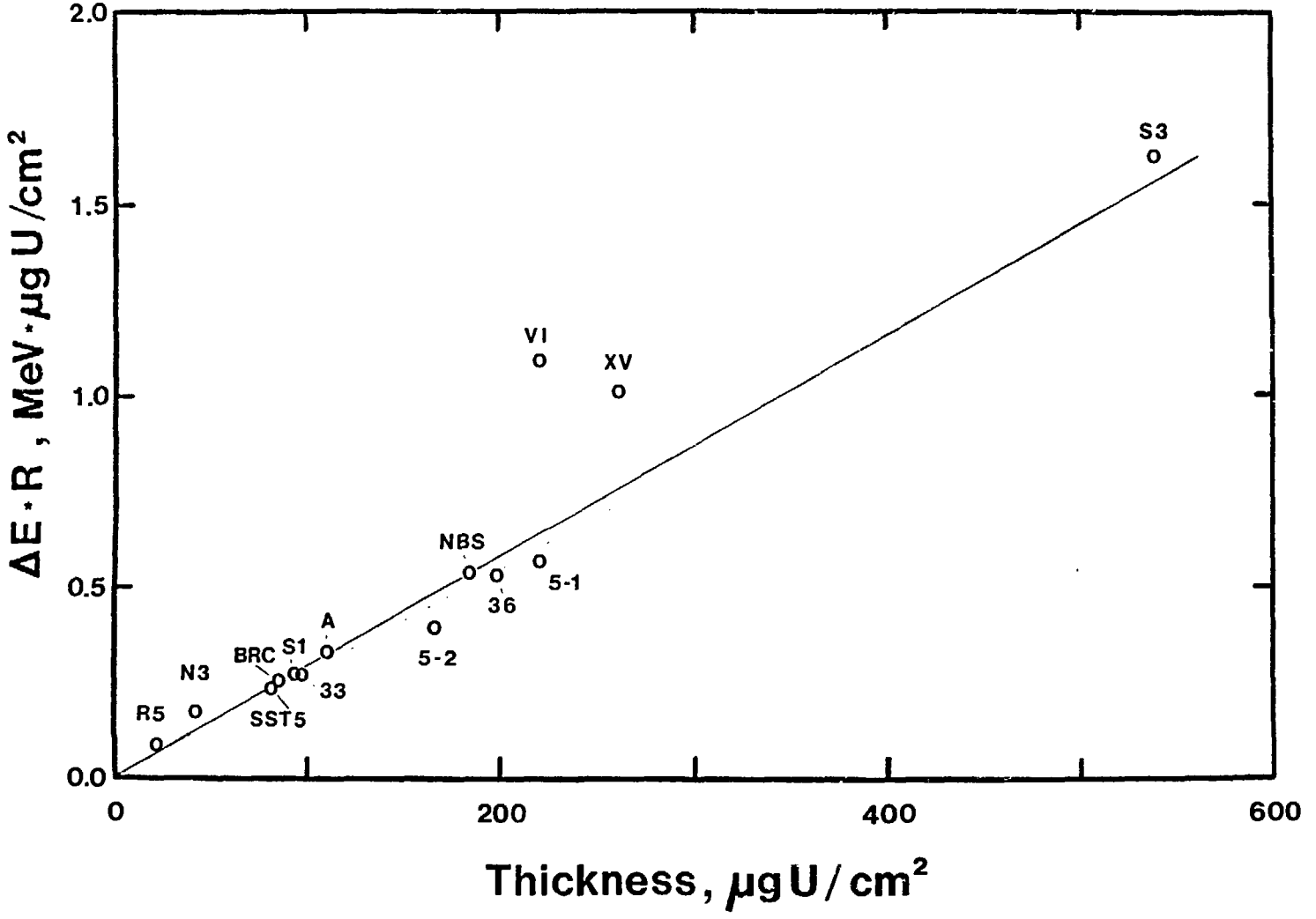

Fig. 4. Alpha Energy Loss Multiplied with the FF Range as a Function of the Sample Thickness. The Values for the KRI Samples is shown for an Assumed UO_2.

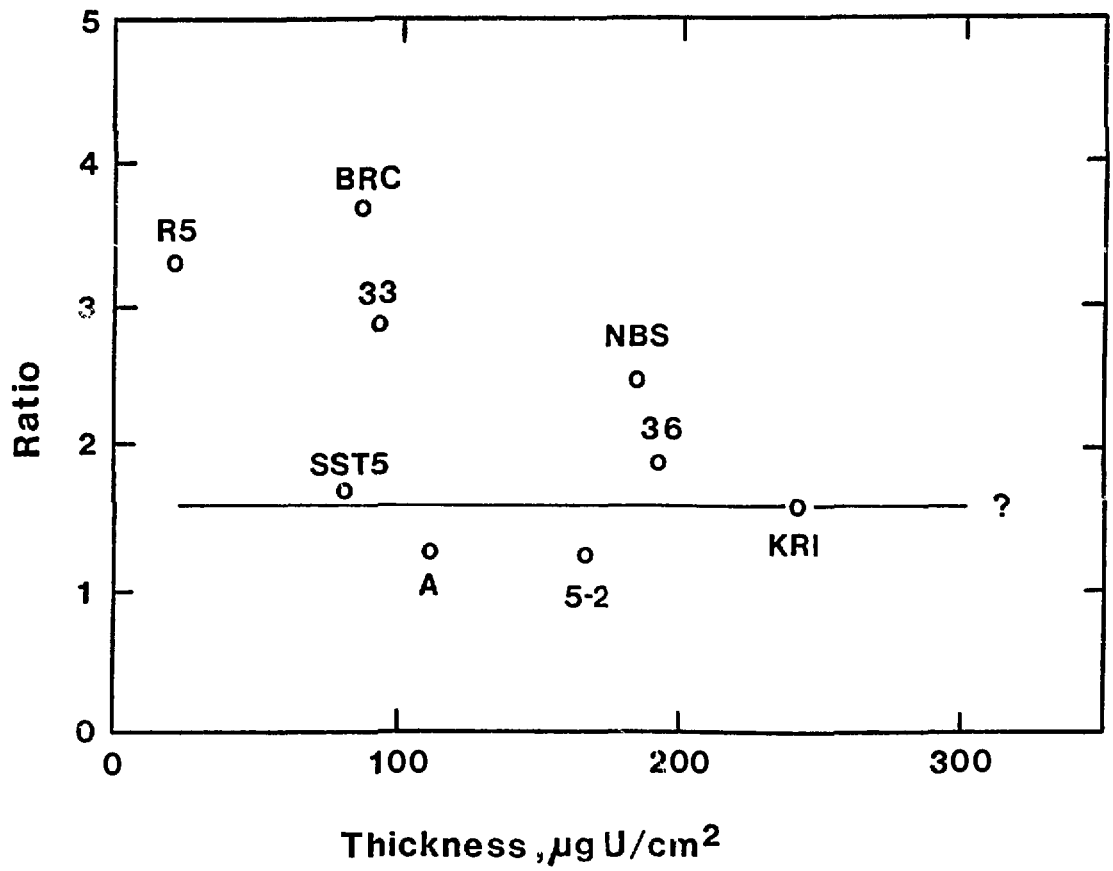

Fig. 5. The Ratios of the Fraction of Pulses Below the Geometrical Cut-off vs. the Fraction of Calculated FF Absorption Losses.

SAMPLE--ANL RS

MASS FROM FISSION RATIO MEASUREMTS//

REL. TO	MASS BASED ON ANL ALPHA COUNT		MASS BASED ON QUOTED MASSES	
ANL R5	.0		.0	
ANL R3	79.5		.0	
ANL S-1	79.3		.0	
ANL S-2	79.4		.0	
ANL SSTS	79.5		.0	
AV. ANL		79.4		79.4
LANL S1	79.5		79.3	
LANL S2	79.5		79.5	
AV. LANL		79.5		79.4
RS	79.3		79.3	
AV. NBS		79.3		79.3
NBL VL	79.5		79.0	
NBL XV	79.7		80.1	
AV. NBL		79.6		79.6
BNC	79.4		.0	
AV. BNC		79.4		.0
AERE A	79.3		78.5	
AERE B	79.2		78.8	
AV. AERE		79.2		78.7
CBNM 33	79.1		79.0	
CBNM 36	79.2		79.0	
AV. CBNM		79.3		79.0
AV. FROM RATIO	79.4			
ANL ALPHA COUNT	79.5			
AV. (RATIOS PLUS)	79.5			

SAMPLE--ANL 43

MASS FROM FISSION RATIO MEASUREMTS//

REL. TO	MASS BASED ON ANL ALPHA COUNT		MASS BASED ON QUOTED MASSES	
ANL R5	52.2		.0	
ANL 43	.0		.0	
ANL S-1	52.0		.0	
ANL S-2	52.1		.0	
ANL SSTS	52.2		.0	
AV. ANL		52.1		52.1
LANL S1	52.2		52.1	
LANL S2	52.2		52.2	
AV. LANL		52.2		52.1
HS	53.0		52.0	
AV. HHS		52.0		52.0
KRI VI	52.2		51.9	
KRI XV	52.3		52.6	
AV. KRI		52.2		52.2
KRC	52.1		.0	
AV. KRC		52.1		.0
AERE A	52.0		51.5	
AERE B	52.0		51.7	
AV. AERE		52.0		51.6
CBNM 33	52.1		51.9	
CBNM 36	52.0		51.8	
AV. CBNM		52.0		51.8
AV. ERDA RATIO	52.1			
ALL ALPHA COUNT	52.1			
AV. (RATIO+ALPHA)	52.1			

SAMPLE--ANL S-1

MASS FROM FISSION RATIO MEASUREMENTS//

REL. TO	ANL ALPHA COUNT	MASS BASED ON QUOTED MASSES	
ANL 82	1071.3	.0	
ANL 83	1071.3	.0	
ANL S-1	.0	.0	
ANL S-2	1067.8	.0	
ANL SSTS	1070.4	.0	
AV. ANL	1070.2		1070.2
LANL S1	1070.4	1067.9	
LANL S3	1070.6	1070.4	
AV. LANL	1070.5		1069.2
SS	1067.3	1066.8	
AV. SS	1067.2		1066.8
ART VI	1070.5	1063.7	
ART XV	1072.4	1078.1	
AV. ART	1071.4		1070.9
[illegible]	1063.7	.0	
AV. [illegible]	1068.7		.0
[illegible] A	1066.3	1057.3	
[illegible] B	1066.1	1060.9	
AV. APPL	1066.5		1059.1
[illegible] B5	1067.2	1063.7	
[illegible] B6	1066.9	1062.9	
AV. [illegible]	1066.5		1063.3
[illegible] FROM RATIO	1068.9		
[illegible]	1067.3		
[illegible]	1066.8		

SAMPLE--ANL S-2

MASS FROM FISSION RATIO MEASUREMTS//

REL. TO	MASS BASED ON ANL ALPHA COUNT		MASS BASED ON QUOTED MASSES	
ANL R5	837.0		.0	
ANL R3	836.1		.0	
ANL S-1	833.9		.0	
ANL S-2	.0		.0	
ANL SST5	836.5		.0	
AV. ANL		835.9		835.9
LANL S1	836.5		834.6	
LANL S3	836.7		836.5	
AV. LANL		836.6		835.6
NBS	834.3		833.9	
AV. NBS		834.3		833.9
KRI VI	836.6		831.3	
KRI XV	838.1		842.6	
AV. KRI		837.3		837.0
BRC	835.4		.0	
AV. BRC		835.4		.0
ALBE A	833.7		826.3	
ALBE B	833.1		829.0	
AV. ALBE		833.4		827.6
[illegible] 33	834.2		831.5	
[illegible] 36	833.2		830.8	
AV. [illegible]		833.7		831.2
AV. FROM RATIO	835.4			
ALL ALPHA COUNT	834.6			
AV. (RATIO+ALPHA)	835.0			

SAMPLE--ANL SSTS

MASS FROM FISSION RATIO MEASUREMTS//

REL. TO	MASS BASED ON ANL ALPHA COUNT		MASS BASED ON QUOTED MASSES	
ANL Q5	415.5		.0	
ANL Q3	418.0		.0	
ANL S-1	416.8		.0	
ANL S-2	417.1		.0	
ANL SSTS	.0		.0	
AV. ANL		417.6		417.6
LANL S1	418.1		417.1	
LANL S3	418.2		418.1	
AV. LANL		418.1		417.6
BBS	416.0		416.7	
AV. BBS		416.9		416.7
KRI V1	415.1		415.5	
KRI XV	418.0		421.1	
AV. KRI		418.5		418.3
HHC	417.4		.0	
AV. HHC		417.4		.0
HEHF A	416.7		413.0	
HEHF B	416.4		414.3	
AV. HEHF		416.5		413.7
CSIRO 33	417.1		415.6	
CSIRO 36	416.4		415.2	
AV. CSIRO		415.7		415.4
AV. FROM RATIO	417.5			
AV. ALPHA COUNT	417.1			
AV. (RATIO+ALPHA)	417.3			

SAMPLE--LANL S1

MASS FROM FISSION RATIO MEASUREMTS//

REL. TO	ANL ALPHA COUNT	MASS BASED ON	QUOTED MASSES	
ANL R5	300.2		.0	
ANL R3	299.3		.0	
ANL S-1	298.4		.0	
ANL S-2	298.7		.0	
ANL SSTS	299.4		.0	
AV. ANL		299.2		299.2
LANL S1	.0		.0	
LANL S3	299.4		299.3	
AV. LANL		299.4		299.3
MSS	298.6		298.4	
AV. MSS		298.6		298.4
NBL MT	299.4		297.5	
NBL AV	300.0		301.6	
AV. NBL		299.7		299.6
[illegible]	299.0		.0	
AV. BRC		298.9		.0
AERE A	298.4		295.7	
AERE B	298.2		296.7	
AV. AERE		298.3		296.2
[illegible]	298.6		297.6	
[illegible]	299.2		297.3	
AV. ORNL		298.4		297.5
AV. FROM RATIO	299.0			
AL ALPHA COUNT	299.4			
AV. (RATIO+ALPHA)	299.2			
QUOTED MASS		298.7		

SAMPLE--LANL S3

MASS FROM FISSION RATIO MEASUREMTS//

REL. TO	MASS BASED ON ANL ALPHA COUNT		MASS BASED ON QUOTED MASSES	
ANL S5	1690.0		.0	
ANL S3	1688.3		.0	
ANL S-1	1683.3		.0	
ANL S-2	1684.4		.0	
ANL SSTS	1688.6		.0	
AV. ANL		1686.9		1686.9
LANL S1	1688.7		1684.7	
LANL S3	.0		.0	
AV. LANL		1688.7		1684.7
NBS	1683.4		1682.6	
AV. NBS		1683.4		1682.6
KRI VI	1688.5		1677.9	
KRI XV	1691.6		1700.6	
AV. KRI		1690.0		1689.3
BRC	1686.4		.0	
AV. BRC		1686.4		.0
AERE A	1682.3		1667.8	
AERE B	1681.8		1672.8	
AV. AERE		1682.0		1670.3
CBNM 33	1686.6		1679.1	
CBNM 36	1681.4		1676.7	
AV. CBNM		1683.0		1677.9
AV. FROM RATIO	1685.9			
AVE ALPHA COUNT	1688.6			
AV. (RATIO+ALPHA)	1687.3			
WEIGHTED MASS		1688.3		

SAMPLE--NBS

MASS FROM FISSION RATIO MEASUREMTS//

REL. TO	ANL ALPHA COUNT	MASS BASED ON QUOTED MASSES	
ANL S5	229.5	.0	
ANL N3	229.2	.0	
ANL S-1	228.5	.0	
ANL S-2	228.7	.0	
ANL SSTS	229.2	.0	
AV. ANL	229.0		229.0
LANL S1	229.2	228.7	
LANL S3	229.3	229.2	
AV. LANL	229.3		229.0
NBS	.0	.0	
AV. NBS	.0		.0
KRI V1	229.3	227.9	
KRI XV	229.7	230.9	
AV. KRI	229.5		229.4
LRC	228.9	.0	
AV. LRC	228.9		.0
AERE A	228.5	226.4	
AERE B	228.3	227.2	
AV. AERE	228.4		226.8
CHEM 33	228.6	227.9	
CHEM 36	228.1	227.7	
AV. CHEM	228.5		227.8
AV. FROM RATIO	229.0		
ANL ALPHA COUNT	228.6		
AV. (RATIO+ALPHA)	228.8		
QUOTED MASS	228.5		

SAMPLE--NBS VI

MASS FROM FISSION RATIO MEASUREMTS//

REL. TO	MASS BASED ON ANL ALPHA COUNT		MASS BASED ON QUOTED MASSES	
ANL S5	763.2		.0	
ANL S3	761.7		.0	
ANL S-1	760.2		.0	
ANL S-2	760.9		.0	
ANL SST5	763.7		.0	
AV. ANL		761.9		761.9
LANL S1	762.6		760.8	
LANL S3	762.8		762.6	
AV. LANL		762.7		761.7
NBS	760.2		759.9	
AV. NBS		760.2		759.9
NBL VI	.0		.0	
NBL XV	764.1		768.2	
AV. NBL		764.1		768.2
BRC	761.3		.0	
AV. BRC		761.3		.0
AERE A	760.2		753.4	
AERE B	759.5		755.9	
AV. AERE		759.9		754.6
CBNM 33	761.6		758.1	
CBNM 36	758.3		756.2	
AV. CBNM		759.4		757.1
AV. FROM RATIO	761.4			
ANL ALPHA COUNT	761.7			
AV. (RATIO+ALPHA)	762.1			
QUOTED MASS		757.9		

SAMPLE--ANL XV

MASS FROM FISSION RATIO MEASUREMTS//

REL. TO	MASS BASED ON ANL ALPHA COUNT		MASS BASED ON QUOTED MASSES	
ANL N5	895.4		.0	
ANL N3	894.9		.0	
ANL S-1	891.7		.0	
ANL S-2	892.4		.0	
ANL SSTS	894.5		.0	
AV. ANL		893.8		893.8
LANL S1	894.5		892.4	
LANL S3	894.6		894.5	
AV. LANL		894.6		893.5
NBS	891.9		891.5	
AV. NBS		891.9		891.5
KRI V1	894.6		888.9	
KRI XV	.0		.0	
AV. KRI		894.6		888.9
NRC	893.9		.0	
AV. NRC		892.9		.0
AERE A	891.7		883.7	
AERE B	890.8		886.5	
AV. AERE		891.2		885.1
CBNM 33	892.6		889.1	
CBNM 36	890.9		888.4	
AV. CBNM		891.5		888.8
AV. FROM RATIO	893.1			
ANL ALPHA COUNT	894.6			
AV. (RATIO+ALPHA)	893.8			
QUOTED MASS	891.3			

SAMPLE--BRC

MASS FROM FISSION RATIO MEASUREMTS//

REL. TO	MASS BASED ON ANL ALPHA COUNT		MASS BASED ON QUOTED MASSES	
ANL R5	117.0		.0	
ANL N3	116.9		.0	
ANL S-1	116.5		.0	
ANL S-2	116.6		.0	
ANL SST5	116.9		.0	
AV. ANL		116.8		116.8
LANL S1	116.9		116.6	
LANL S3	116.9		116.9	
AV. LANL		116.9		116.8
PBS	116.6		116.5	
AV. PBS		116.6		116.5
NBL VI	116.9		116.2	
NBL XV	117.1		117.7	
AV. NBL		117.0		117.0
BRC	.0		.0	
AV. BRC		.0		.0
AERE A	116.5		115.5	
AERE B	116.4		115.8	
AV. AERE		116.5		115.7
CBNM 33	116.6		116.2	
CBNM 36	116.4		116.1	
AV. CBNM		116.5		116.1
AV. FROM RATIO	116.7			
ANL ALPHA COUNT	116.7			
AV. (RATIO+ALPHA)	116.7			

SAMPLE--MURR A

MASS FROM FISSION RATIO MEASUREMTS//

REL. TO	ANL ALPHA COUNT	MASS BASED ON	QUOTED MASSES	
ANL 82	340.1		.0	
ANL 83	347.6		.0	
ANL 5-1	346.5		.0	
ANL 5-2	346.3		.0	
ANL SSTS	347.7		.0	
AV. ANL		347.3		347.3
LANL S1	347.7		346.8	
LANL S2	347.7		347.6	
AV. LANL		347.7		347.2
HRS	346.7		346.5	
AV. HRS		346.7		346.5
KRI VI	347.7		345.5	
KRI V	348.3		350.2	
AV. KRI		348.0		347.9
ORC	347.1		.0	
AV. ORC		347.1		.0
BEMF 3	.3		.0	
BEMF 4	346.2		344.5	
AV. BEMF		346.2		344.5
CBNM 33	346.3		345.6	
CBNM 36	346.2		345.2	
AV. CBNM		346.5		345.4
AV. FROM RATIO	347.2			
ANL ALPHA COUNT	346.5			
AV. (RATIO+ALPHA)	346.9			
QUOTED AVG		346.4		

SAMPLE--AERE B

MASS FROM FISSION RATIO MEASUREMTS//

REL. TO	ANL ALPHA COUNT	MASS BASED ON QUOTED MASSES	
ANL R^5	348.5	.0	
ANL R^3	349.3	.0	
ANL S-1	347.1	.0	
ANL S-2	347.4	.0	
ANL SSTS	348.2	.0	
AV. ANL	347.9		347.9
LANL S1	348.2	347.4	
LANL S3	348.3	348.2	
AV. LANL	348.3		347.8
NBS	347.3	347.1	
AV. NBS	347.3		347.1
NFT VI	349.9	346.1	
NFT XV	348.9	350.8	
AV. NFT	348.6		348.4
EBC	347.6	.0	
AV. EBC	347.6		.0
AERE A	347.1	344.0	
AERE B	.0	.0	
AV. AERE	347.1		344.0
CBNM 33	347.3	346.2	
CBNM 46	346.8	345.8	
AV. $CBNM^{235}$	347.1		346.0
AV. FROM RATIO	347.8		
ANL ALPHA COUNT	348.3		
AV. (RATIO+ALPHA)	347.8		
QUOTED MASS	345.1		

SAMPLE--CONN 33

MASS FROM FISSION RATIO MEASUREMTS//

REL. TO	MASS BASED ON ANL ALPHA COUNT		MASS BASED ON QUOTED MASSES	
ANL M5	122.7		.0	
ANL M3	122.6		.0	
ANL S-1	122.2		.0	
ANL S-2	122.4		.0	
ANL SST5	122.6		.0	
AV. ANL		122.5		122.5
LANL S1	122.6		122.3	
LANL S3	122.6		122.6	
AV. LANL		122.6		122.5
NBS	122.3		122.2	
AV. NBS		122.3		122.2
NRT VI	122.6		121.9	
NBL XV	122.9		123.5	
AV. NBL		122.7		122.7
BMC	122.4		.0	
AV. BMC		122.4		.0
AERE A	122.2		121.1	
AERE B	122.1		121.5	
AV. AERE		122.2		121.3
CBNM 33	.0		.0	
CBNM 36	122.1		121.8	
AV. CBNM		122.1		121.8
AV. FROM RATIO	122.5			
REL ALPHA COUNT	122.3			
AV. (RATIO*ALPHA)	122.4			
QUOTED MASS		121.9		