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Light Ternary Fission Products: Probabilities and Charge Distributions

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LIGHT TERNARY FISSION PRODUCTS: PROBABILITIES AND CHARGE DISTRIBUTIONS

by

David G. Madland and Leona Stewart

ABSTRACT

A survey has been made of experimental information pertinent to the probability of ternary fission and the charge distribution of the light ternary fission products. A new prescription is presented for the ternary fission probability as a function of incident particle energy and certain compound nucleus properties. Based upon systematics, a method for obtaining charge distributions of the light ternary products is presented.

I. INTRODUCTION

The following is a presentation of certain systematics found in ternary fission data together with a number of phenomenological prescriptions by which these data can be extended to unexplored regions. The information presented here can be of use, for example, in areas such as the absolute normalization of binary fission theories, normalization and charge conservation in phenomenological yield models, or tritium production estimates from fission in a reactor. The reader is cautioned that this is an interim summary[†] based upon an as yet incomplete study of existing pertinent experimental and theoretical work in ternary fission.

The probability of emission of light ternary fission products is discussed in Sec. II. In Sec. III information is presented on the charge distribution of the light products. Recommendations for users and a summary are contained in Sec. IV.

[†]Performed at the request of the Fission Product and Actinide Data Subcommittee of the Cross Section Evaluation Working Group (CSEWG).

II. PROBABILITY OF LIGHT TERNARY FISSION PRODUCTS

Consider first the probability of emission of light ternary fission products of all species for which $(Z,A) \leqslant (4,10)$.[†] While no detailed, accurate, empirical relationships seem to exist, experiments do indicate approximately valid trends. Let N = # of light charged particles released per 1000 fissions and consider the following:

A. The work summarized by Thomas and Whetstone,[1] on compound fissioning systems of ^{236}U, ^{240}Pu, and ^{239}Np, can be represented by

$$N = 1.355 + 0.047E^{*} \quad , \tag{1}$$

where E^{*} is the excitation energy of the compound system. This expression gives N to within approximately ± 25% for $15 \leqslant E^{*} \leqslant 38$ MeV.

For $E^{*} < 15$ MeV, however, they find the excitation energy dependence to be more complex. N decreases from $E^{*} = 0$ (spontaneous fission) to $E^{*} \sim 6$–7 MeV (thermal neutron fission), which implies existence of a minimum in N for some value of E^{*} between 8 and 14 MeV. One finds $N_{avg} = 3.3 \pm 0.5$ for spontaneous fission of $^{240,242}Pu$, $^{242,244}Cm$, and ^{252}Cf, and $N_{avg} = 2.3 \pm 0.2$ for thermal fission of $^{233,235}U$ and $^{239,241}Pu$.

Table I contains neutron binding energies, B_n, for the compound system, which are useful in Eq. (1), that is, $E^{*} = B_n + E_{inc}$, where E_{inc} is the incident neutron energy.[††]

B. Similar work by Nobles,[2] together with data of Nagy[3] and Loveland,[4] can be represented by

$$N = -18.299 + 0.561X \quad , \tag{2}$$

[†] We exclude pre-scission and scission neutrons because their number has yet to be quantified to any reasonable degree of accuracy.

[††] This expression neglects the recoil energy correction which is typically ~60 keV for 14-MeV neutrons incident on uranium.

where X = fissility parameter = Z^2/A, and (A,Z) characterize the compound nucleus. This equation gives N to within approximately ± 30% for low excitation (spontaneous and thermal) fission data upon which it is based. Thus Eq. (2) is valid in regions of excitation where Eq. (1) is not. Values of X are tabulated in Table I for several actinides of interest.

C. Halpern[5] studied the ternary alpha data of Refs. 2, 3, 6 and considered the yield, N, to be a slowly varying function of A and Z for fixed excitation energy of the compound fissioning system (A,Z). A first-order Taylor expansion of N(A,Z) about some arbitrary (A_o,Z_o) demonstrates that N is a linear function of the variable $w = \beta Z + A$, where $\beta = (\partial N/\partial Z)/(\partial N/\partial A)$ evaluated at (A_o,Z_o). Halpern obtained fits linear in w for data of fixed neutron energy (and for spontaneous fission), which is approximately the same as fixed excitation energy. The best results were achieved for $w = 4Z - A$, that is, $\beta = -4$, for which the coefficient of w was ≃0.125 for all neutron energies considered.

An attempt has been made to improve upon the above prescriptions for the case of particle-induced ternary fission. The data used in the analysis are given in Table II.† These data are plotted in Fig. 1 as a function of the excitation energy, E^*, of the compound system (a portion of the data, at E^* ≃6-8 MeV, was deleted from the figure for lack of space). Included in the figure is a least squares linear fit to that portion of the data for which $E^* \geqslant 15$ MeV, that is, a fit similar to that described in paragraph A, except with a more extensive data set. One obtains

$$N = 0.508 + 0.070E^* \quad , \tag{3}$$

which is to be compared with Eq. (1). As Fig. 1 indicates, a linear fit in the variable E^* is still rather unsatisfactory, giving agreement to within approximately ± 25% at best. In fact, the computed value of chi-square is

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†Data points published without uncertainty estimates were not included in the table. In two instances, where three or more independent measurements were made of the same N, a datum was deleted because it was five or more standard deviations away from the average of the others. Note also that most experimental N values represent lower limits because of the difficulties in collecting light ternary products of low kinetic energy.

χ^2 = 22.8/degree of freedom (17 data points, 2 degrees of freedom).

It appears that parameterizations of the form of Eqs. (1), (2), and (3) do not contain enough information to be of any precise use. What may be lacking is more information about the compound fissioning species (E^* essentially conveys the bombarding energy of the incoming particle inducing the fission). The ternary fission probability perhaps depends as much, or more, upon details of the compound fissioning system than it does upon the excitation energy or incoming particle energy. Thus, Halpern's view, as described in paragraph C, is probably the most appropriate starting point. The work of Halpern[5] may be summarized by

$$N = a_1 + a_2(4Z-A)_{comp} \quad , \tag{4}$$

where "comp" refers to the compound system and a_1 and a_2 are to be determined for data sets obtained at fixed incident neutron energy.

A consistent, although not unique, manner by which additional information on the particle energy as well as the compound nucleus can be brought to bear in Eq. (4) is to parameterize the coefficients a_1 and a_2 as follows.

$$a_1 = b_1 + b_2\varepsilon \tag{5}$$

and

$$a_2 = c_1 + c_2\varepsilon \quad , \tag{6}$$

where the b_i and c_i are constants and ε is the available excitation energy of the compound system with respect to its outer fission barrier in the two-humped fission barrier model.[7] That is,

$$\varepsilon = E^* - \langle E_B \rangle \tag{7}$$

and

$$E^* = B_n + E_{inc} \quad , \tag{8}$$

where E^* is the excitation energy of the compound system (as already noted), $\langle E_B \rangle$ is the average value of the height of the outer fission barrier, tabulated[8] in Table I, B_n is the binding energy of the last neutron of the compound nucleus ground state, also tabulated[9] in Table I, and E_{inc} is the incident neutron energy.† An energy parameterization, ε, with respect to a fission barrier is more appropriate than, say, E^* because ε is an energy pertinent to the compound system having chosen the fission channel for de-excitation whereas E^* refers to the initial condition. Moreover, ε is computed with respect to the outer fission barrier because there is some evidence that the mass split decision is made near the outer barrier, namely that the potential energy surface is reflection asymmetric at (and beyond) this point.[10,11] Thus, ε is an appropriate energy parameter for the fission channel, and whether or not it is useful to describe ternary fission probabilities will be seen below. Note that Eqs. (5) and (6) are only linear in ε. As will be shown, the quality and paucity of the data (Table II) admit only to terms linear in ε.

The data of Table II have been divided into six bins in ε which are 1 MeV wide at low ε (thermal and fast neutron fission) and 2.5 MeV wide at high ε (14-MeV neutron fission and above). Least squares linear fits to Eq. (4) were obtained for the data of each bin. The results are

$$N = \left\{ \begin{array}{ll} -24.777 + 0.2014(4Z-A)_{comp} \text{ for } 0.5 \leqslant \varepsilon \leqslant 1.5 \text{ MeV} & , \\ -16.954 + 0.1425(4Z-A)_{comp} \text{ for } 1.5 < \varepsilon \leqslant 2.5 \text{ MeV} & , \\ -8.513 + 0.0815(4Z-A)_{comp} \text{ for } 2.5 < \varepsilon \leqslant 3.5 \text{ MeV} & , \\ -22.741 + 0.1860(4Z-A)_{comp} \text{ for } 12.5 \leqslant \varepsilon \leqslant 15 \text{ MeV} & , \\ -37.494 + 0.2985(4Z-A)_{comp} \text{ for } 17 \leqslant \varepsilon \leqslant 19.5 \text{ MeV} & , \\ -50.347 + 0.4020(4Z-A)_{comp} \text{ for } 30 \leqslant \varepsilon \leqslant 32.5 \text{ MeV} & . \end{array} \right\} \quad (9)$$

These results are illustrated in Figs. 2-7 together with the averaged data upon which they are based.

Least squares linear fits to Eqs. (5) and (6) were then obtained for coefficients a_1 and a_2 of Eq. (9). This was done for two broad regions in ε:

†See footnote to paragraph A with respect to Eq. (8).

(1) $0.5 \leqslant \varepsilon \leqslant 3.5$ MeV, corresponding to thermal and fast neutron fission, and (2) $12.5 \leqslant \varepsilon \leqslant 32.5$ MeV, corresponding to 14-MeV neutron fission and above. The results are

$$a_1 = \left\{ \begin{array}{l} -33.395 + 8.295\,\varepsilon \\ \qquad 0.5 \leqslant \varepsilon \leqslant 3.5 \text{ MeV} \quad , \\ -6.935 - 1.410\,\varepsilon \\ \qquad 12.5 \leqslant \varepsilon \leqslant 32.5 \text{ MeV} \quad , \end{array} \right\} \tag{10}$$

and

$$a_2 = \left\{ \begin{array}{l} 0.263 - 0.0613\,\varepsilon \\ \qquad 0.5 \leqslant \varepsilon \leqslant 3.5 \text{ MeV} \quad , \\ 0.0604 + 0.0111\,\varepsilon \\ \qquad 12.5 \leqslant \varepsilon \leqslant 32.5 \text{ MeV} \quad . \end{array} \right\} \tag{11}$$

These results are illustrated in Figs. 8 and 9 together with the "data" [coefficients of Eq. (9)] upon which they are based. The number of Table II data points utilized in each region of ε is also indicated in the figures. As the figures show, no derived ternary fission probabilities are produced for $3.5 < \varepsilon < 12.5$ MeV. There are two reasons for this: (1) no experimental data exists for this region in ε, and (2) the behavior of a_1 and a_2 with respect to ε is changing rapidly in this region as Figs. 8 and 9 show, that is, a_1 and a_2 are linearly decreasing with ε for low ε and are linearly (or quadratically) increasing with ε for high ε. Thus, more data are needed in order to determine the behavior in what appears to be a minimum region.

Collecting the above results, the final expression for N, the number of light charged particles released per 1000 fissions, is given by

$$\begin{aligned} N = (-33.395 + 8.295\,\varepsilon) \\ + (0.263 - 0.0613\,\varepsilon)(4Z-A)_{\text{comp}} \end{aligned} \tag{12-A}$$

$$\text{for } 0.5 \leqslant \varepsilon \leqslant 3.5 \text{ MeV}$$

and

$$N = (-6.935 - 1.410\varepsilon) + (0.0604 + 0.0111\varepsilon)(4Z-A)_{comp} \quad \text{for } 12.5 \leqslant \varepsilon \leqslant 32.5 \text{ MeV} \quad . \tag{12-B}$$

Comparing Eq. (12-A) to the data of Table II, one finds $\chi^2 = 11.9$/degree of freedom (20 data points, 4 degrees for freedom), which would be reduced to $\chi^2 = 5.6$/degree of freedom if the 2.5-MeV point for ^{232}Th + n were deleted. For Eq. (12-B) one finds $\chi^2 = 4.9$/degree of freedom (17 data points, 4 degrees of freedom), which should be compared to the value of 22.8 obtained by using Eq. (3).

III. CHARGE DISTRIBUTION OF LIGHT TERNARY FISSION PRODUCTS

There exists even less experimental information on the charge distribution of light ternary fission products than on the probability of emission of the light products (Table II). Data are compiled in Table III for the charge and mass distributions of the light products produced in the thermal neutron fission of ^{233}U and ^{235}U and the spontaneous fission of ^{252}Cf. Relative data (in % of alpha particle yield) were taken from the compilation by Halpern[5] and, in the case of ^{233,235}U thermal fission, have been normalized using the thermal N values of Table II. The ^{252}Cf data have been normalized by using the N value of Appendix I of Ref. 1, namely, N = 3.46 ± 0.19. Estimated uncertainties are contained in the footnotes to Table III. As with the N values of Table II all entries should be considered as tending toward lower limits.

Inspection of the table shows that generally the most frequently occurring ternary products are (in decreasing order) ^{4}He, ^{3}H, ^{6}He, ^{1}H, and ^{2}H. On the basis of three compound fissioning systems this ordering is independent of the compound species.† This is an important result because it allows the construction of approximately valid absolute charge distributions for other compound systems. For example, the number frequencies of Table III can be scaled according to the N values of Table II to produce absolute charge distributions. However, the validity of the scaling might very well diminish for increasing

†This statement presumes that were there a ^{1}H data point for ^{233}U, it would mesh properly with the sequence.

excitation energy of the compound system because the data of Table III are for thermal or spontaneous fission. Figure 10 illustrates the three charge distributions, n(Z), which have been delineated in the table. Each distribution has been arbitrarily normalized to 10^4 products of Z = 2. If one were to increase the n(Z=1) value for ^{233}U, to account for the lack of the ^{1}H datum in Table III, the shape of the distributions would become almost identical. Two other important features stand out in the figure: (1) to a good approximation the charge distribution is a δ-function at Z = 2, and (2) if one considers products of Z = 1 and 2 to be of first-order occurrence and products of Z = 3 and 4 to be of second-order occurrence, then for each order it is obvious that even-Z products are enhanced over odd-Z products. Assuming a pairing force to be operative the same effect should occur for neutrons and indeed, for fixed Z, even-N products are enhanced over odd-N products according to the data of Table III. Note, however, that these observations are confined to measurements on compound fissioning systems of even-Z even-N character.

IV. SUMMARY AND RECOMMENDATIONS

It is clear from the above that much experimental work could be done in the area of ternary fission probabilities. As indicated by Eqs. (12-A, 12-B) these probabilities should be measured for ε values between 3.5 and 12.5 MeV, which roughly corresponds to neutron energies ranging from 4 to 13 MeV. Such measurements would settle the question of the existence and location of a possible minimum in the ternary fission probability as a function of ε, as implied by Figs. 8 and 9.

Similarly, more detailed information is needed on the mass and charge distributions of the ternary fission products (as contained in Table III). In particular, the $^{235}U + n_{th}$ experiment, summarized in Table III, should be repeated at several higher neutron energies in order to extract the excitation energy dependence of the ternary charge and mass distributions. Moreover, it would be useful to measure such detail for a compound system that is odd Z - even N or odd Z - odd N to see if the even-odd Z character of the charge distribution (Fig. 10) remains the same or becomes washed out.

It would be pleasing if it could be stated that use of the variable ε was crucial to the formalism leading up to Eqs. (12-A, 12-B). We cannot make such a statement. In fact, the calculation was repeated by using the variable E^*, see Eq. (8), with only slightly worse overall results in which the greatest discrepancy occurred for the thermal region.

Users of this work requiring N values should first have recourse to the experimental values contained in Table II. Equations (12-A, 12-B) can be used to energy average as well as to calculate N values in unexplored regions. In case of the latter, additional fission barrier heights may be found in Ref. 9. If charge distributions are sought we recommend scaling the ^{235}U thermal data of Table III by the appropriate experimental N values of Table II or the calculated N values from Eqs. (12-A, 12-B).

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TABLE I

SOME PROPERTIES OF THE COMPOUND NUCLEUS IN FISSION

System	$(Z^2/A)_{comp}$	$(4Z-A)_{comp}$	B_n (MeV)	$\langle E_B \rangle$ (MeV)
^{229}Th + n	35.21739	130	6.790	6.15 ± 0.20
^{232}Th + n	34.76395	127	4.786	6.28 ± 0.20
^{233}U + n	36.17094	134	6.841	5.95 ± 0.25
^{234}U + n	36.01702	133	5.306	5.65 ± 0.30
^{235}U + n	35.86441	132	6.546	5.74 ± 0.20
^{236}U + n	35.71308	131	5.124	5.95 ± 0.30
^{237}U + n	35.56302	130	6.144	6.12 ± 0.20
^{238}U + n	35.41423	129	4.803	6.30 ± 0.30
^{238}Pu + n	36.97071	137	5.655	5.35 ± 0.30
^{239}Pu + n	36.81667	136	6.534	5.40 ± 0.20
^{240}Pu + n	36.66390	135	5.240	(5.50)[d]
^{241}Pu + n	36.51240	134	6.301	5.39 ± 0.20
^{242}Pu + n	36.36214	133	5.037	(5.60)[d]
^{244}Pu + n	36.06531	131	4.720	(5.45)[d]
^{241}Am + n	37.29339	138	5.528	4.90 ± 0.20
^{243}Am + n	36.98770	136	5.363	4.80 ± 0.25
^{242}Cm + n	37.92593	141	5.702	5.5 ± 0.3
^{244}Cm + n	37.61633	139	5.520	(4.2)[d]
^{240}Pu sf	36.81667	136		5.40 ± 0.20
^{242}Pu sf	36.51240	134		5.39 ± 0.20
^{242}Cm sf	38.08264	142		
^{244}Cm sf	37.77049	140		< 4.9
^{252}Cf sf	38.11111	140		
^{232}Th + α	35.86441	132	− 4.569[a]	5.74 ± 0.20
^{238}U + α	36.51240	134	− 4.982[b]	5.39 ± 0.20
^{238}U + p	36.18828	133	5.299[c]	5.5 ± 0.3

[a]Binding energy of last alpha particle in ^{236}U.

[b]Binding energy of last alpha particle in ^{242}Pu.

[c]Binding energy of last proton in ^{239}Np.

[d]Not well determined.

TABLE II

TERNARY FISSION DATA FOR PARTICLE-INDUCED FISSION

System	E_{inc} (MeV)	N
$^{232}Th + n$	2.5	0.840 ± 0.031[c]
"	14.0	0.958 ± 0.088[c]
$^{233}U + n$	0.0	2.427 ± 0.094[a]
"	0.33	2.985 ± 0.312[a]
"	0.69	2.257 ± 0.260[a]
"	1.17	2.160 ± 0.257[a]
"	1.99	2.551 ± 0.377[a]
$^{235}U + n$	0.0	2.137 ± 0.196[c,d]
"	0.33	2.016 ± 0.309[a]
"	1.0	1.873 ± 0.123[a]
"	1.17	1.653 ± 0.227[a]
"	2.5	2.174 ± 0.302[a]
"	3.0	1.678 ± 0.183[a]
"	14.0	2.016 ± 0.260[a]
"	14.0	1.456 ± 0.070[c]
$^{238}U + n$	2.5	0.907 ± 0.023[c]
"	2.5	1.667 ± 0.233[a]
"	14.0	0.976 ± 0.124[a]
"	14.0	1.258 ± 0.055[c]
$^{239}Pu + n$	0.0	2.326 ± 0.108[a]
"	0.33	2.439 ± 0.256[a]
"	0.69	2.092 ± 0.249[a]
"	1.0	2.481 ± 0.135[a]
"	1.99	2.500 ± 0.638[a]
$^{241}Pu + n$	0.0	2.273 ± 0.145[a]
$^{232}Th + \alpha$	29.5	1.961 ± 0.200[a]
"	42.0	2.717 ± 0.281[a]
$^{238}U + \alpha$	29.5	2.558 ± 0.288[a]
"	42.0	3.521 ± 0.285[a]
$^{238}U + p$	10.5	1.661 ± 0.201[a]
"	11.	2.25 ± 0.17 [b]
"	12.	1.99 ± 0.16 [b]
"	13.	2.16 ± 0.16 [b]
"	14.	2.31 ± 0.17 [b]
"	15.	2.33 ± 0.14 [b]
"	17.	2.42 ± 0.15 [b]
"	17.5	2.101 ± 0.199[a]

[a]See Appendix II of Ref. 1.
[b]See Table IV of Ref. 1.
[c]See Table I of Ref. 3.
[d]See Table 11.16 of Ref. 12.

TABLE III

ABSOLUTE YIELDS OF LIGHT CHARGED TERNARY FISSION PRODUCTS PER 10^6 FISSIONS

Species	$^{233}U + n_{th}$ [a]	$^{235}U + n_{th}$ [b]	^{252}Cf sf [a]
^{1}H		23.4	50.0
^{2}H	9.1	8.8	18.4
^{3}H	104.3	121.9	214.6
^{3}He		< 0.1	< 30
^{4}He	2268.3	1950.4	3065.4
^{6}He	31.8	24.4	61.3
^{8}He	0.82	0.64	3.6
^{6}Li		< 0.01	5.1 (combined)
^{7}Li	0.84	0.70	
^{8}Li	0.45	0.27	
^{9}Li	0.82	0.21	
^{7}Be		$< 2 \times 10^{-5}$	10.8 (combined)
^{9}Be	0.84	0.39	
^{10}Be	9.8	5.8	

[a]Absolute errors are estimated at ± 20% of the quoted yield.

[b]Absolute errors are estimated at ± 25% of the quoted yield.

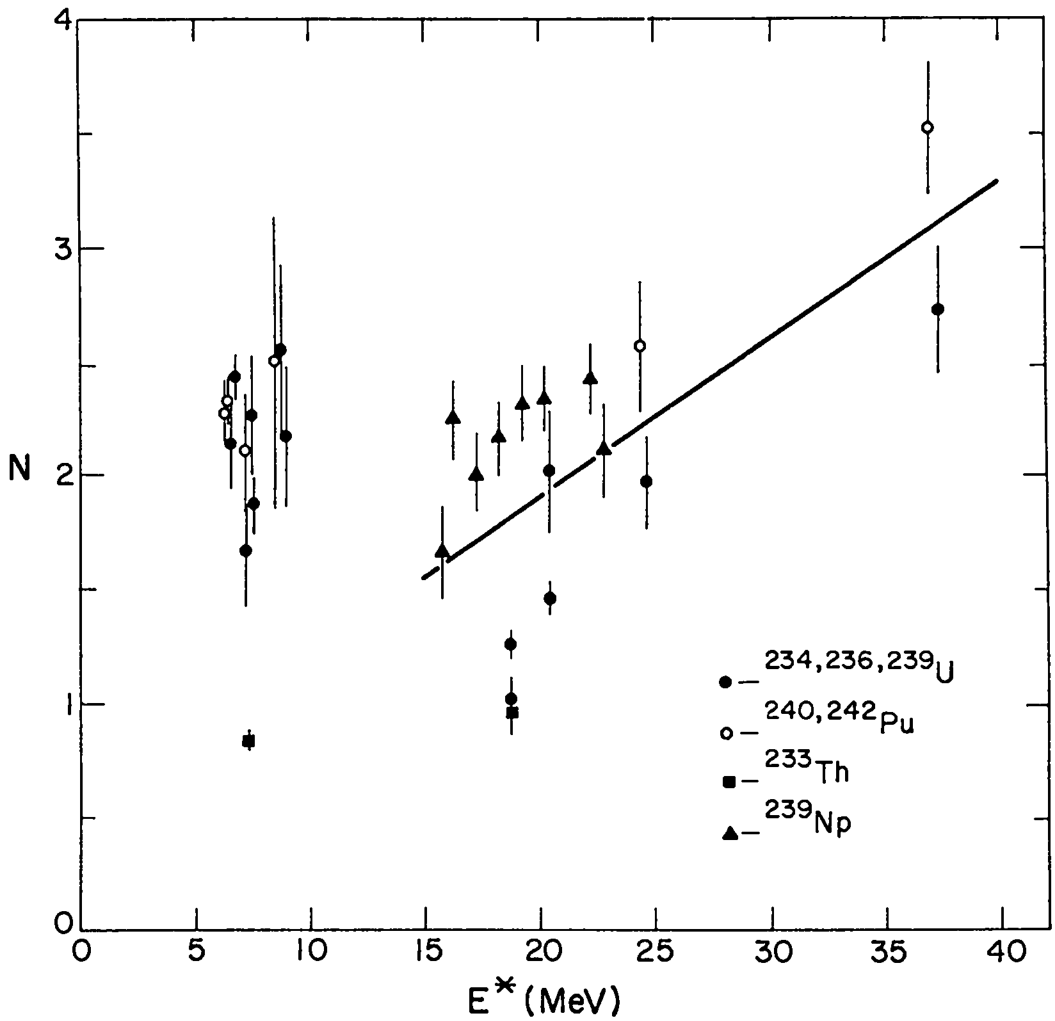

Fig. 1.

Light charged particle yield per 1000 fissions, N, vs the excitation energy, E^*, of the indicated compound systems. The linear fit is explained in the text (see Eq. 3).

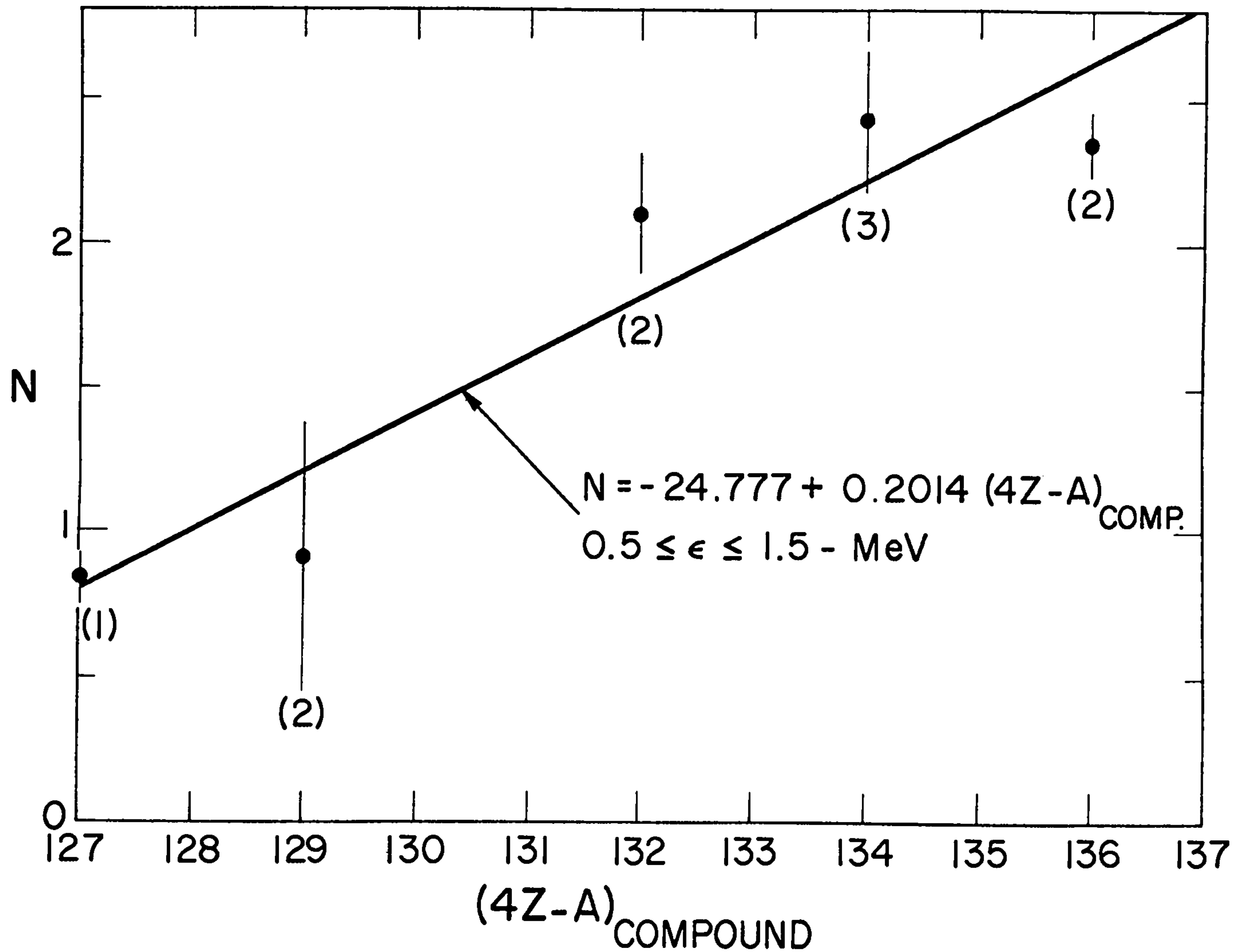

Fig. 2.

Least squares linear fit of N to $(4Z\text{-}A)_{comp}$ for data in the range $0.5 \leqslant \varepsilon \leqslant 1.5$ MeV. The illustrated points are weighted averages (inverse square error) of the number of measurements given in the parentheses.

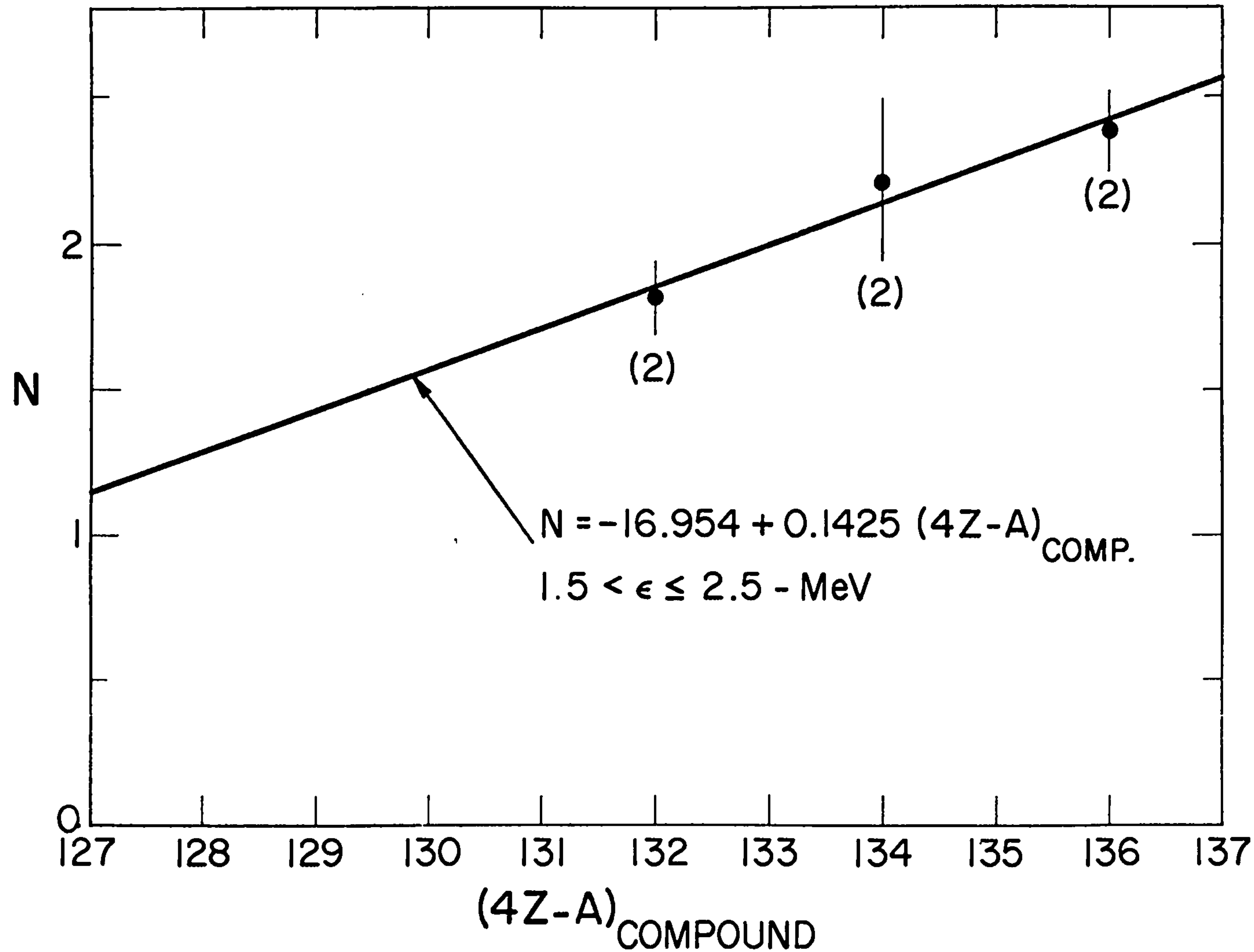

Fig. 3.

Least squares linear fit of N to $(4Z\text{-}A)_{comp}$ for data in the range $1.5 < \varepsilon \leqslant 2.5$ MeV. The illustrated points are weighted averages (inverse square error) of the number of measurements given in the parentheses.

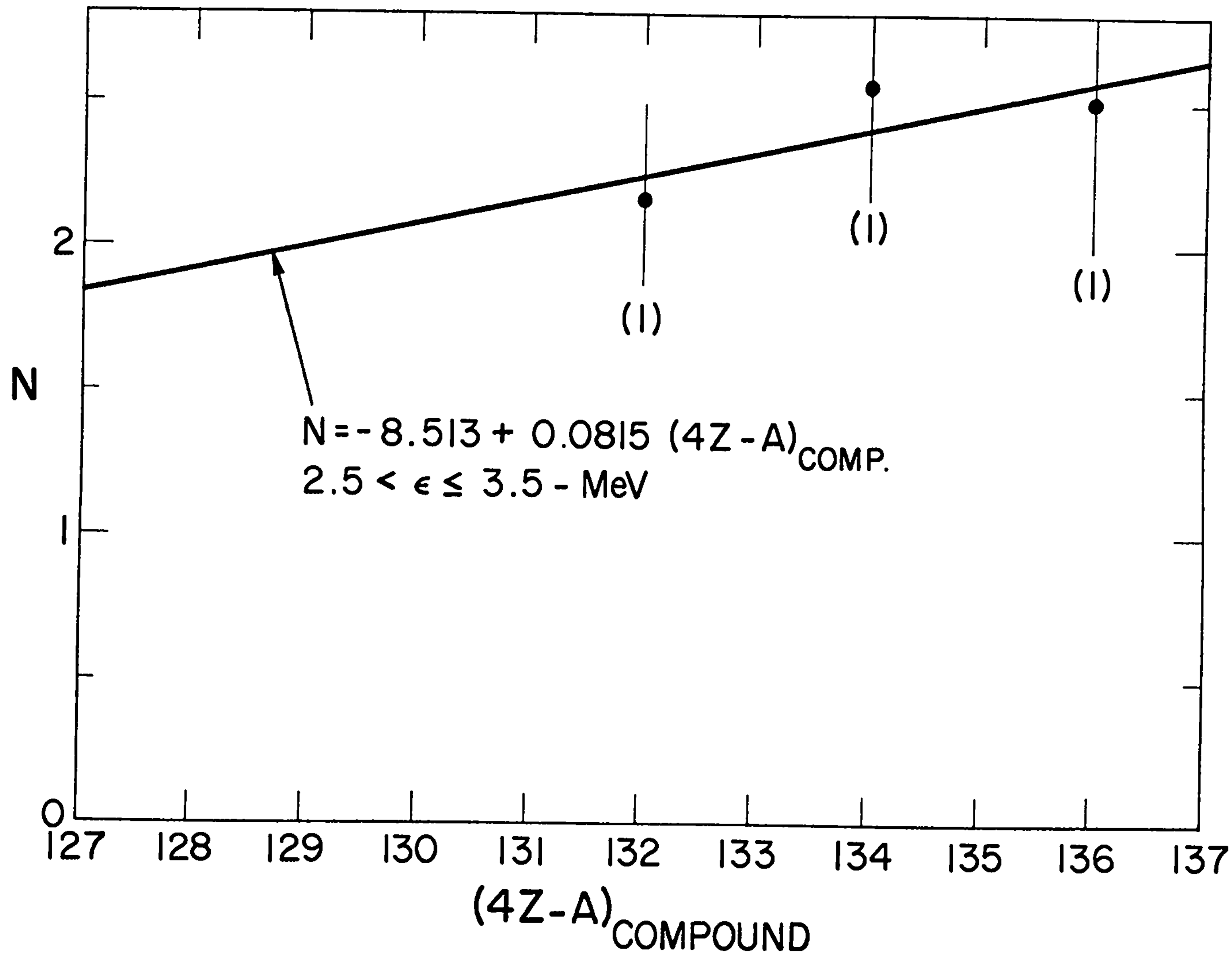

Fig. 4.

Least squares linear fit of N to $(4Z\text{-}A)_{comp}$ for data in the range $2.5 < \varepsilon \leqslant 3.5$ MeV. The number of measurements is given in the parentheses.

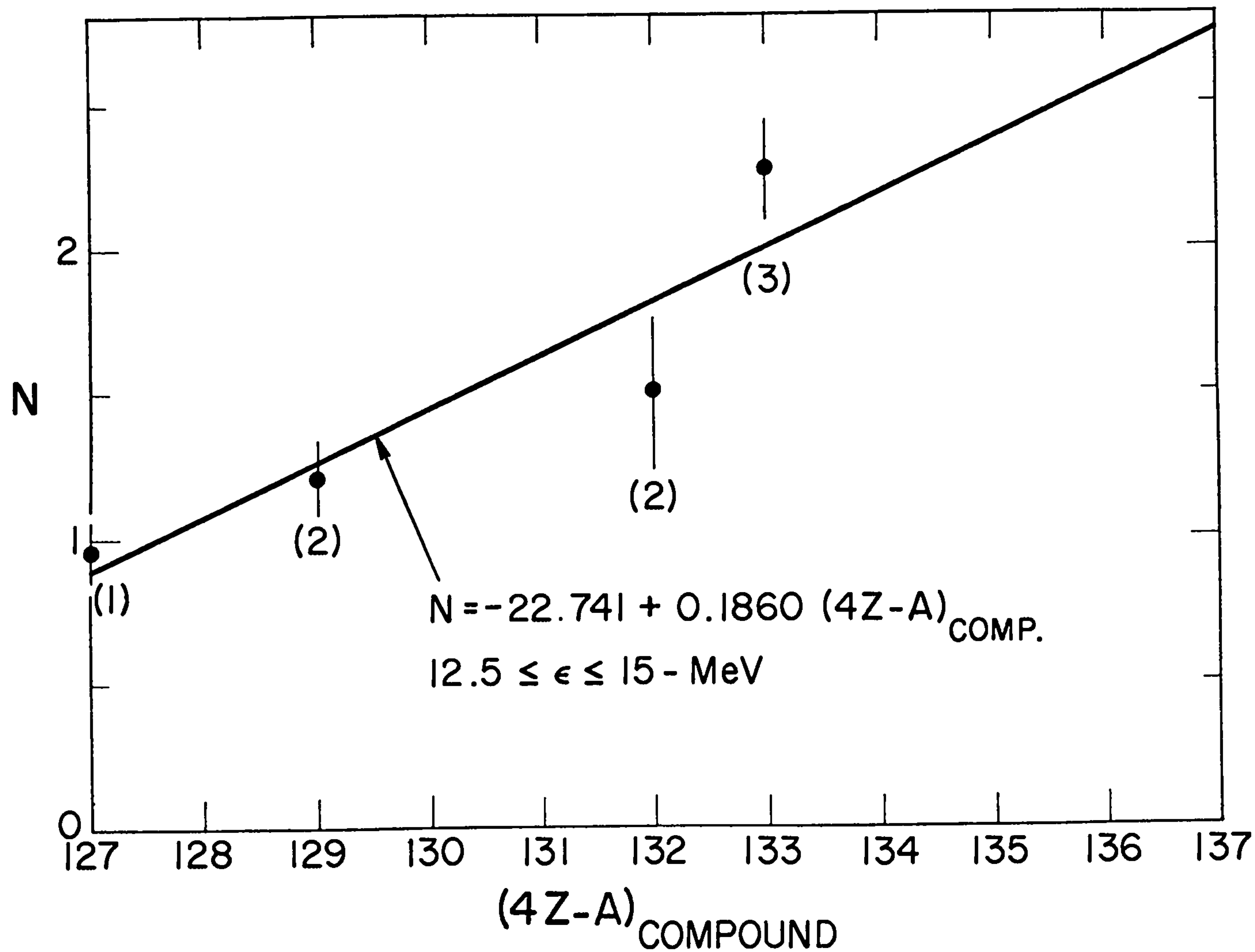

Fig. 5.

Least squares linear fit of N to $(4Z\text{-}A)_{comp}$ for data in the range $12.5 \leqslant \varepsilon \leqslant 15$ MeV. The illustrated points are weighted averages (inverse square error) of the number of measurements given in the parentheses.

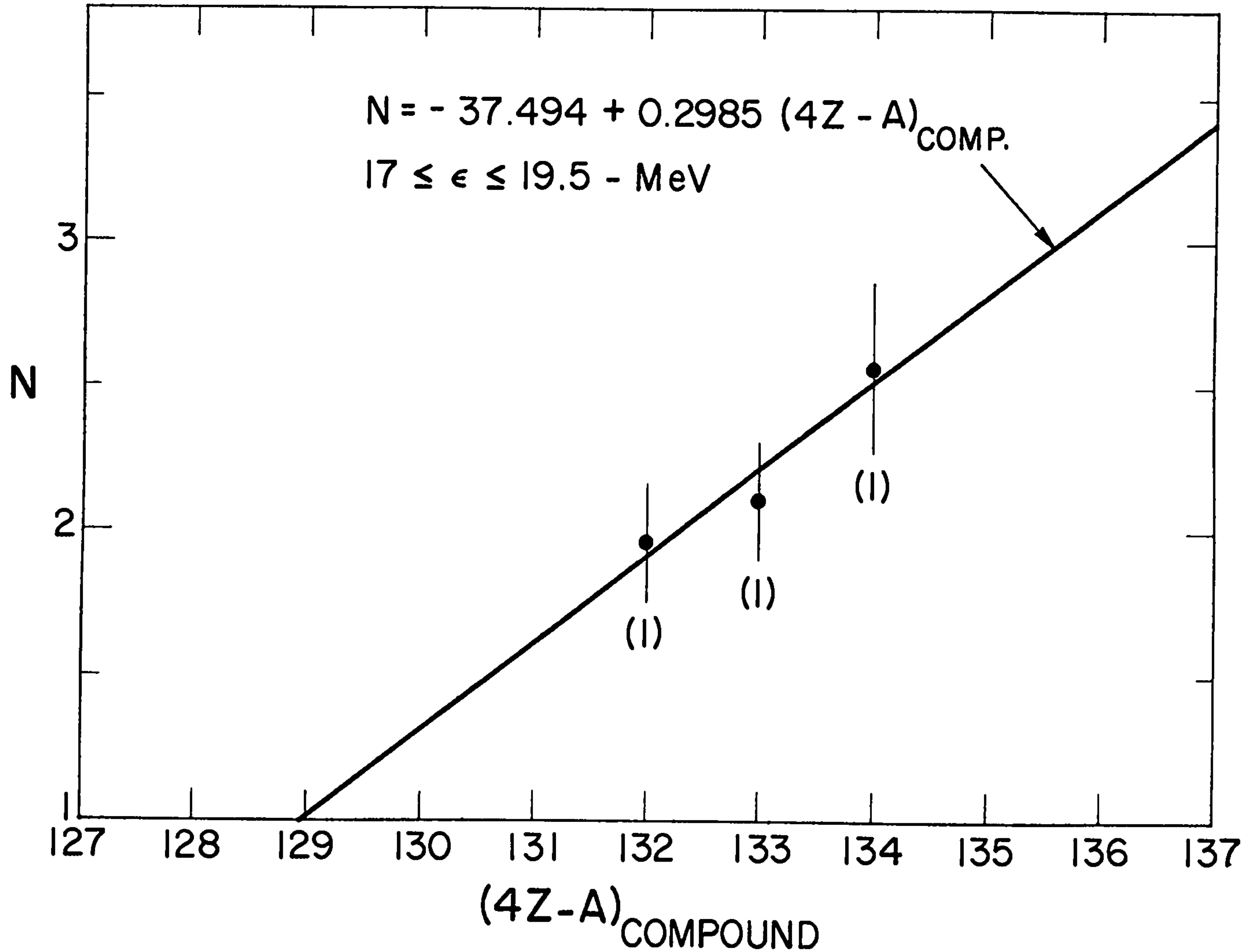

Fig. 6.

Least squares linear fit for $\bar{N}$ to $(4Z-A)_{comp}$ for data in the range $17 \leqslant \varepsilon \leqslant 19.5$ MeV. The number of measurements is given in the parentheses.

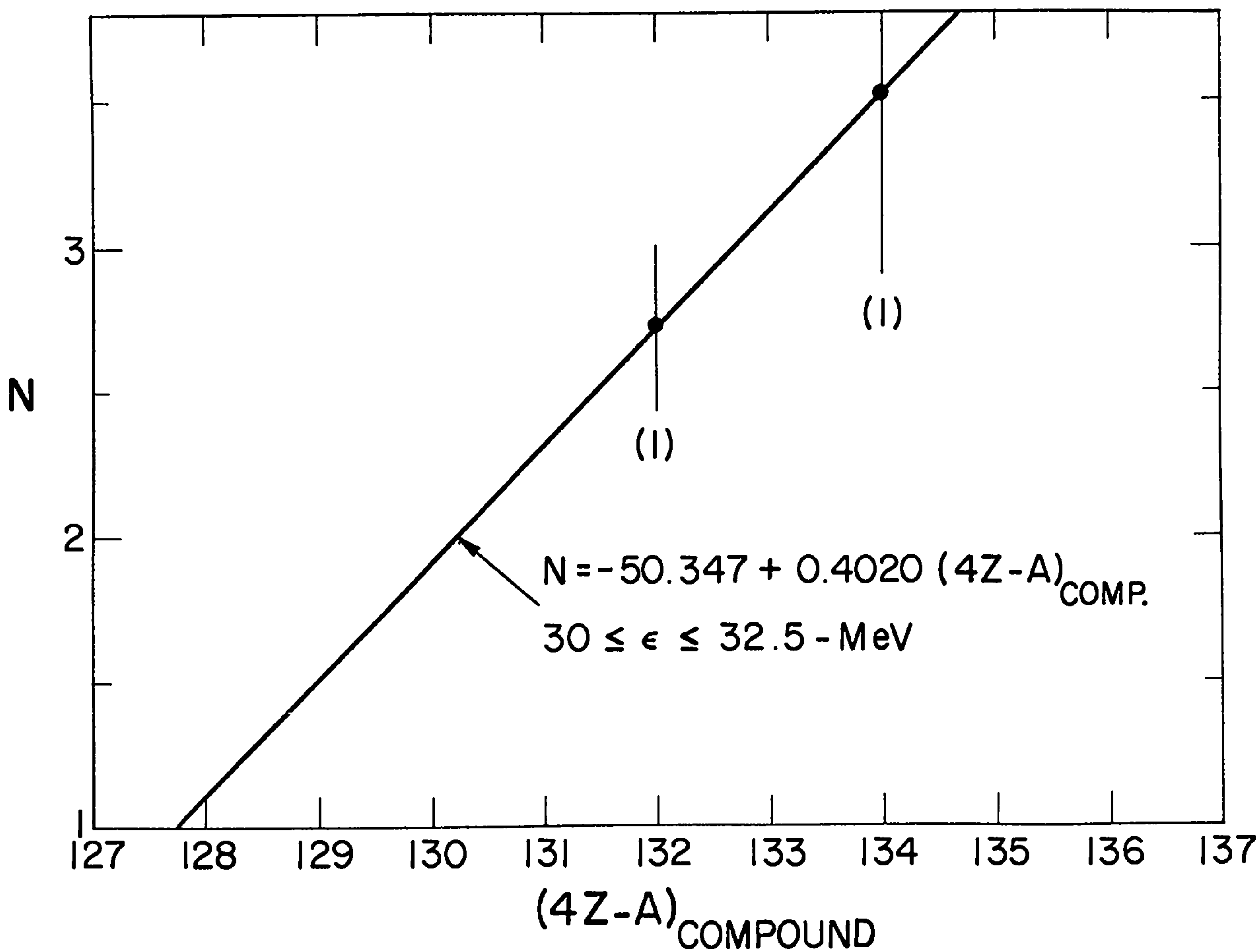

Fig. 7.

Least squares linear fit for N to $(4Z\text{-}A)_{comp}$ for data in the range $30 \leqslant \varepsilon \leqslant 32.5$ MeV). The number of measurements is given in the parentheses.

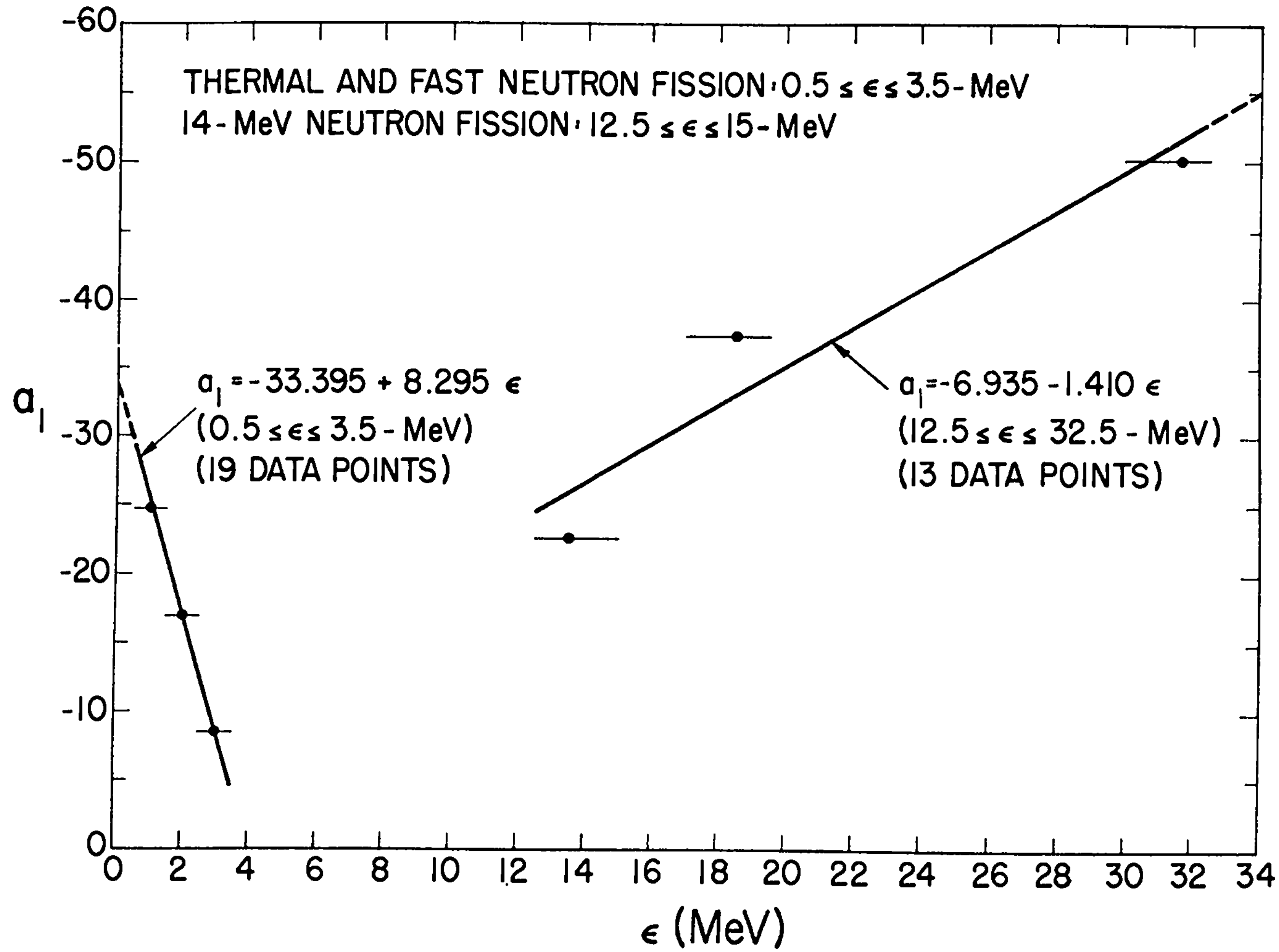

Fig. 8.

The parameter a_1 as a function of ε for two broad ranges in ε together with indicated least squares linear fits. The number of data points used for each range of ε is given. Dashed lines represent extrapolations into regions devoid of experimental measurements.

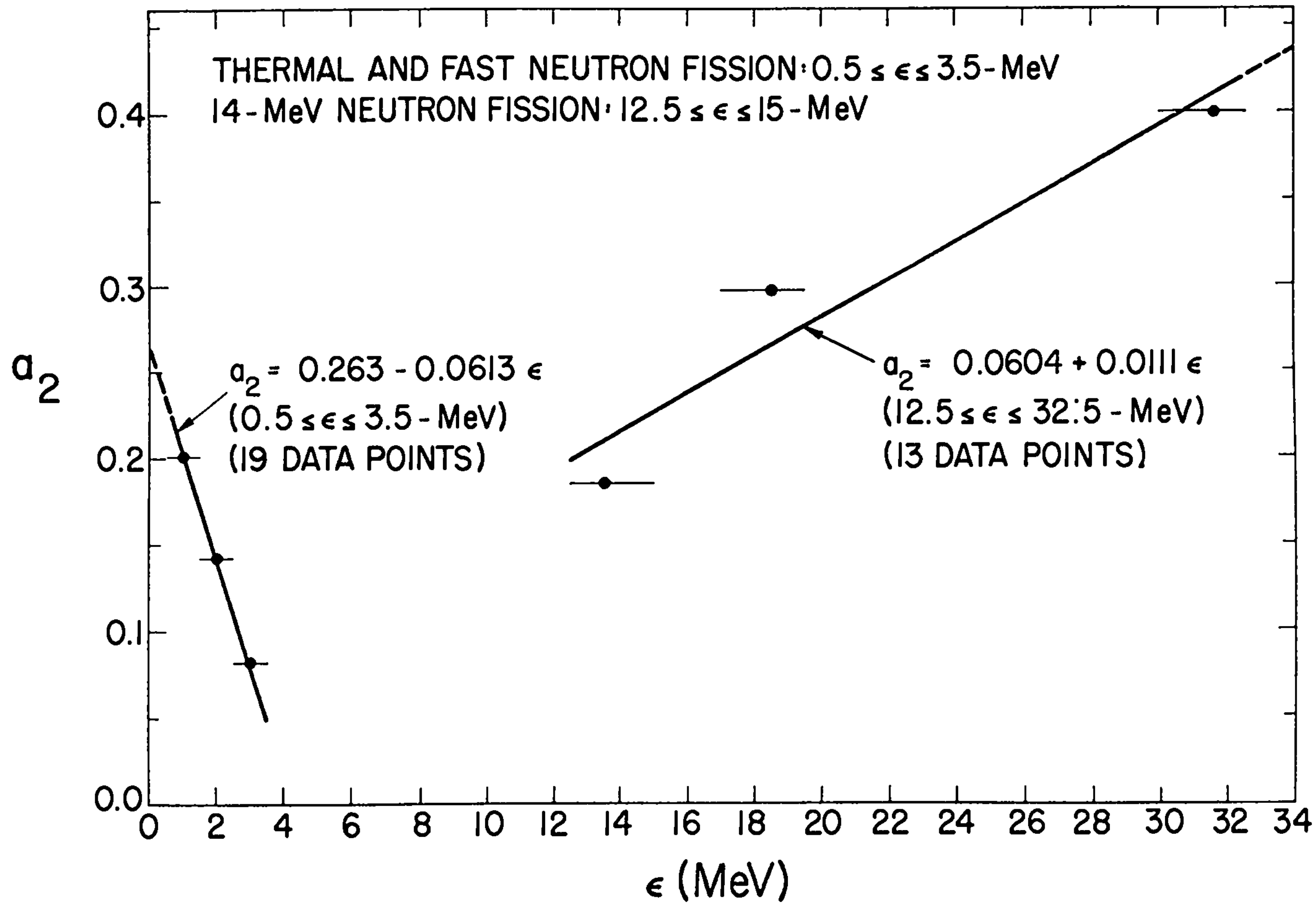

Fig. 9.

The parameter a_2 as a function of ε for two broad ranges in ε together with indicated least squares linear fits. The number of data points used for each range of ε is given. Dashed lines represent extrapolations into regions devoid of experimental measurements.

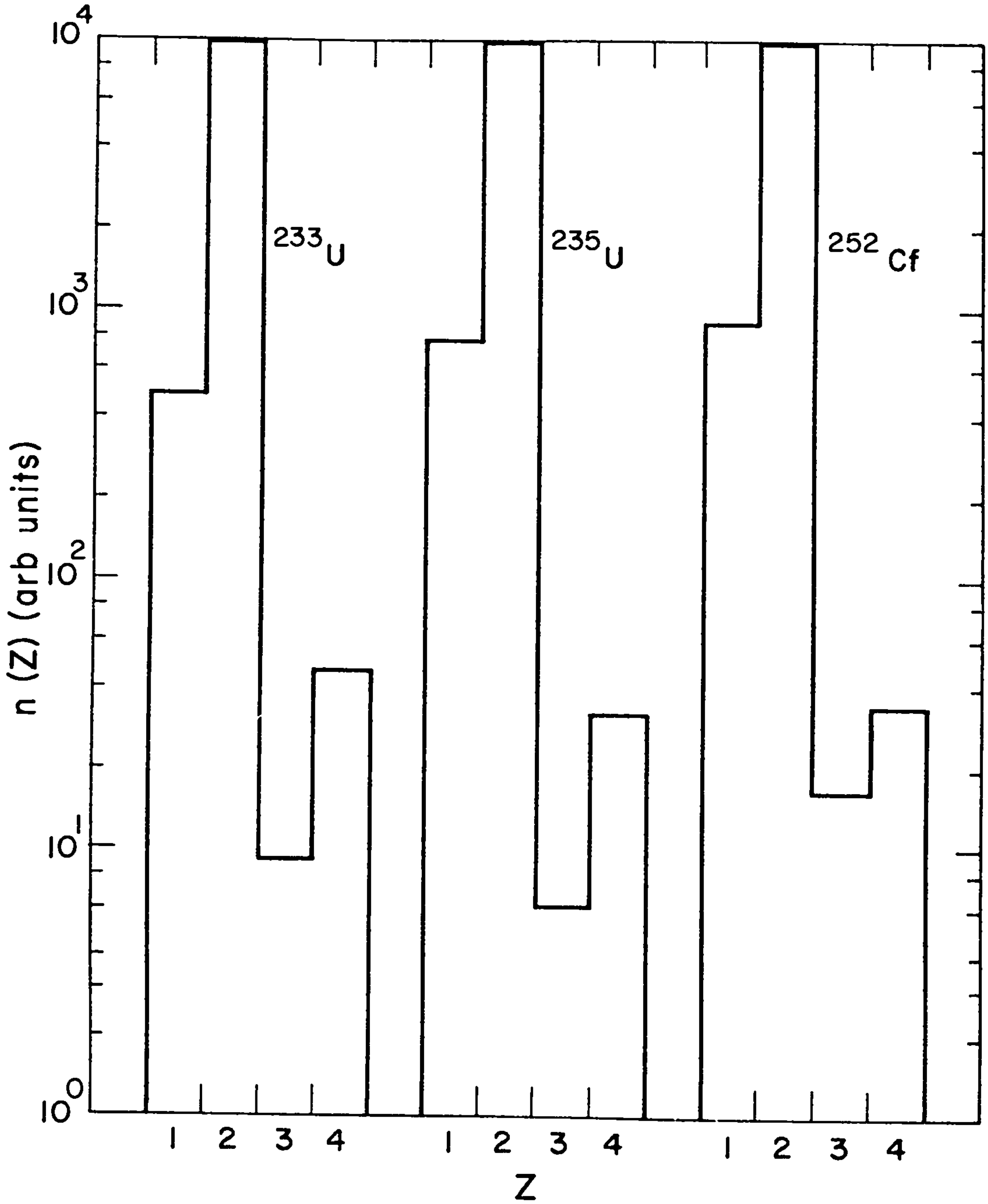

Fig. 10.

Charge distributions, n(Z), obtained from the data of Table III. Each distribution is arbitrarily normalized to 10^4 products of $Z = 2$.

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