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Systematics of Neutron-induced fission yields

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ABSTRACT

The main characteristics of the mass and charge distributions for thermal neutron induced fission of actinides are reviewed. We show that these distributions can be reasonably reproduced with only  $\approx 24$  data as input. We use a representation where the element yields together with the most probable mass  $A_p(Z)$  play the dominant role. The ability of this model to calculate mass yields for the fission of not yet measured actinides is also shown.

The influence of the excitation energy of the fissile system on charge and mass distribution is also discussed.

## INTRODUCTION

The distributions of charge and mass are probably the most important characteristic of the fission-process. Although a large amount of experimental data /1,2/ on these distributions in fission has been available, no suitable theory yet exists, which explains all the observation. So some empirical models have been developed and largely used up to now.

The description of these two models : The  $Z_p$  model, and the  $A_p$  model will be done and compared to recent and almost complete experimental results.

The first and most important part of the paper will deal with thermal neutron induced fission. The influence of the increasing neutron energy will be discussed in the second part.

## MASS YIELDS

It is known now since a long time, that the mean mass of the heavy group remain almost constant as function of the masses of the fissioning nucleus. The mean mass of light group increases continuously with the mass of the fissioning nucleus as can be seen in figure 1.

These observations are well explained by the strong influence of the shell effects in the nascent fragments /13/.

This shell effect plays also an important role on the evaporated neutrons, in particular it explains the saw-tooth function of  $\bar{V}_L$  and  $\bar{V}_H$ .

## INDEPENDENT YIELDS

Necessary to complete the task of determining chain mass(A) distributions are the charge distributions for all mass chains. The great number of nuclear species involved makes the experimental task difficult almost impossible. However for problems like decay power or reactivity studies, the complete information on independent yields is required. In consequence after a review of the existing experimental data, the approaches used to obtain a complete set of independent yields

will be discussed.

### EXPERIMENTAL VALUES

The case of the thermal neutron induced fission of  $^{235}\text{U}$  illustrates the difficulties encountered in determining independent yields : although more yield data are available for this isotope than for other fissile systems, the data are not complete, specially in the heavy group of fission products. In the following, we will restrict ourselves to the cases of the three best known charge distributions :  $^{235}\text{U}(\text{nth},\text{f})$ ,  $^{233}\text{U}(\text{nth},\text{f})$  and  $^{239}\text{Pu}(\text{nth},\text{f})$ .

Data concerning these three isotopes are gathered in recent compilations /1,2/ with their respective references. Data from the early days come essentially from radiochemical measurements and for some elements like alcali or rare gases /3,4/, from isotopic separation techniques. The use of high resolution fission fragment spectrometers has added a new powerful technique for the measurement of independent yields. Spectrometers like HIAWATHA/5/ or LOHENGRIN/6/ separate the fission products according to their mass using strong electric and magnetic fields (Lohengrin) or by time of flight measurement of the velocity (Hiawatha). The separation according to the nuclear charge is obtained by measuring the fission product energy loss in a homogeneous plastic or gaseous absorber.

Recently the Lohengrin spectrometer has been equipped to work with transuranium actinides. The charge distribution in the light group of fission products has been measured for  $^{233}\text{U}$ /7/ and for  $^{239}\text{Pu}$ /8/. We present in figures 2 and 3 these two charge distributions. For  $^{233}\text{U}$  the Z numbers range from  $Z=33$  to  $Z=42$  and the mass number A between  $A=80$  and  $A=106$ . For  $^{239}\text{Pu}$  the corresponding values are  $35 < Z < 43$  and  $85 < A < 109$ . The meaning of the continuous curves which are drawn through the data points will be discussed later. These data correspond to yields summed over the ionic charge q and over the kinetic energy  $E_k$ . Therefore they are directly comparable to the values obtained by radiochemical ways.

### CHARACTERISTICS OF THE CHARGE DISTRIBUTIONS

In practice it is generally observed that the charge distribution within a particular mass chain follow a gaussian distribution characterised by its most probable charge  $Z_p(A)$  and a width parameter  $c_z(A)/9/$ . Using this dependence the independent yield of a given nuclear species can be represented

by the expression :

$$YI(A, Z) = (\sigma_{ZVZ\eta})^{-1} \cdot EOF(Z, N) \cdot YT(A) \cdot \exp\left[-(Z - Z_p(A))^2 / 2\sigma_z^2\right] \quad (1)$$

$YT(A)$  being the chain yield for the mass number  $A$  and  $EOF(Z, N)$  a factor which takes into account the parity of  $Z$  and  $N$  numbers.

#### AVERAGE OR MOST PROBABLE CHARGE

The resulting dependence of the most probable charge  $Z_p$  on mass number  $A$  is usually presented in such a way as to exhibit the displacement of  $Z_p$  from the charge calculated assuming an equal charge to mass ratio for both the fragments and the fissioning nucleus (U C D hypothesis).

$$Z_p^{UCD} = (ZF/AF) \cdot (A + \Delta(A)) \quad (2)$$

For all the fissioning systems, the deviation from this simple law is roughly half a unit favouring the lighter fragment.

#### WIDTH OF THE CHARGE DISTRIBUTION

As for the average nuclear charge, the mean value of the width parameter  $\sigma_z$  seems to be more or less independent of the fissioning system : ( $\sigma_z = 0.6$ ). This fact can be understood if we consider that the variance of the charge distribution is only due to zero point oscillation /6,10/. Figure 4 shows the comparison of the rms widths  $c_z$  for the fission of  $^{235}\text{U}$  and  $^{239}\text{Pu}$ . The  $c_z$  values are displayed as a function of the average nuclear charge  $Z$  of the corresponding isobar. The modulation which appears on the curves is a consequence of the proton odd-even effect /11/ which will be discussed in the next paragraph. We can also observe that the amplitude of the

oscillations is less for Plutonium than for Uranium, a fact which is easily explained by the decrease of the odd-even effect.

#### PROTON EVEN-ODD EFFECT (EOZ)

The amplitude of the proton even-odd effect has been measured for a limited set of thermally fissioning nuclei. As can be seen from Figure 5 the amplitude of the EOZ decreases considerably with increasing Z of the fissioning nucleus. However EOZ factors seem to be the same for the different isotopes of a given element. The EOZ factor for  $^{229}\text{Th}$ ,  $^{232}\text{U}$ , and  $^{252}\text{Cf(sf)}$  have been obtained by DE-E techniques /12/, /13/. On the other hand the values of EOZ which are plotted on figure 5, are averaged over the whole charge distribution for a given fissile isotope and it should be noted that the EOZ factor depends on the nuclear charge number Z of the fragments. This fact can be explained by the interplay between shell effects and pairing effects which are known to be anticorrelated.

#### NEUTRON EVEN ODD EFFECT (EON)

Unlike protons, neutrons number is not preserved during the fission process. Data concerning EON are summarized in table I. The great similarity between the different fissioning systems and specially the comparison between thermal and proton induced fission seems to indicate that the EON effect is mainly due to the evaporation process. Moreover W.Lang /10/ has shown using a Monte Carlo simulation that a possible primary EON effect is completely changed by the evaporation. As a consequence, the best choice for EON factor would be a value which is independent of the fissioning systems and equal to the mean value for the three cases mentioned above

$$\text{EON} = 6 + 2 \%$$

#### SETS OF ADJUSTED INDEPENDENT YIELDS

Conventional methods :

Systematical descriptions of independent yields are generally obtained by combining the experimental data with

values calculated using a Gaussian charge distribution function /9,16/ : The  $Z_p$  model.

This method is used in the two most recent compilations /1,2/ and is described in details in many papers. In consequence we will briefly recall the main characteristics of the method. According to equation 1 the evaluation of independent yields depends on 3 parameters :

- The width parameter :  $\sigma_z(A)$
- The most probable charge :  $Z_p(A)$
- The even-odd factor :  $EOF(Z,N)$

This method works reasonably well for isotopes like 235U where the existing data are sufficiently abundant to allow a good determination of the free parameters. However for other fissile systems, the validity of the predictions is limited by some problems, inherent to the method. In particular the major weakness of this method is the need to rely on accurate  $Z_p$  values : due to the small width of the charge distribution the  $Z_p$  values must be calculated with a precision better than 0.1 charge unit.

#### COMPLEMENTARY DESCRIPTION OF MASS AND CHARGE DISTRIBUTIONS

Following an idea first proposed by Wahl /14/ we have developed /12/ an empirical model which is able to reproduce the data available for fissile isotopes ranging from Thorium to Californium. This model is based on the observation that the isotopic distribution of a given element can be described by a Gaussian function :

$$YI(A_i, Z_j) = EON \cdot \left( \frac{\sigma}{A} \right)^{-1} (2\pi)^{-1/2} \cdot YE(Z_i) \exp \left[ - \frac{(A_i - A(Z_j))^2}{2 \sigma^2} \right]$$

EON corresponds to the residual neutron even-odd effect (6%)

$\sigma_A$  is the rms width of the isotopic distribution

$YE(Z_i)$  is the element yield corresponding to the charge division ( $Z_i, Z_F - Z_i$ )

$A(Z_j)$  is the mean value of the mass of fragments with charge  $Z_j$ .

The quantities  $\bar{A}(Z_j)$  and  $YE(Z_j)$  are summarized in table II for the three fissile isotopes mentioned above and for the light group of fission products. The rms width  $\sigma_A$  is simply given by the relation

$$A = \bar{Z}. (AF/ZF) = 1.5 \pm 0.1 \text{ A.M.U}$$

The excellent reproduction of the experimental data can be seen on Figures 2,3, where the continuous lines correspond to the calculated yields without neutron even-odd effect. However some significant exceptions to this general agreement are found in the mass region  $99 < A < 103$  for both the uranium and plutonium isotopes. This "anomalous" behaviour can be explained /13/ by the presence of a neutron shell in the deformed fragment which enhanced the yields of isotones with  $N=62-64$ .

The numerous advantages of such a representation are the following.

1 - The element yields  $YE(Z)$  are independent of neutron evaporation (conservation of nuclear charge). In consequence the element yields of the light fragment and the corresponding heavy partner are strictly equal :  $YE(Z_L) = YE(Z_H)$

2 - The proton odd-even effect and his dependence on the nuclear charge are automatically included in the element yields

3 - The mean mass value  $\bar{A}(Z)$  is not affected by the proton odd even effect. Therefore the  $\bar{A}(Z)$  data exhibit a smooth behaviour contrary to the  $Z_p(A)$  values. In consequence extrapolation of missing values or even to other fissioning

systems is greatly facilitated

4 - Due to the fact that the total number of evaporated neutrons  $\bar{V}(A)$  does not depend very much on mass number A, the evaluation of  $\bar{A}(ZH)$  values from  $\bar{A}(ZL)$  is simply given by :

$$\bar{A}(ZL) + \bar{A}(ZH) = AF - \bar{V}T$$

$\bar{V}T$  is the average number of evaporated neutrons.

5 - This representation can be used to calculate mass yields as well as independent yields, provided that element yields are known. As an example we have calculated the set of independent yields for the thermal fission of  $^{229}\text{Th}$ . The  $\bar{A}(ZL)$  values are deduced from figure 6 and the  $YE(Z)$  values are taken from reference /12/ and /15/. Figure 7 shows the result of the calculation and the comparison between our deduced mass yields and the recent measurements of Dickens /11/. The agreement between the data and the values obtained by summing the calculated independent yields is generally good except for the region around  $A=100$ .

#### THE A'P MODEL OF A.C. WAHL

We have shown in the preceding sections that the knowledge of the elemental yields  $YE(Z)$  and the evaluation of the mean mass parameters  $\bar{A}(ZL)$  were sufficient to describe the various distributions of mass and charge of the light actinides as far as high yields values ( $>0.1\%$ ) are concerned. However an application of this simple method to the symmetric fission yields or to heavier actinides is not possible for two reasons.

- 1) The elemental yields are not measured.
- 2) In these two cases the neutron evaporation increases considerably. As the consequence the  $\bar{A}(ZL)$  values cannot be extrapolated.

In a recent conference /16/ A.C. Wahl has developed roughly the same model but in a more refined version. In particular the most probable mass of a given element  $A'p(Z)$  is evaluated before neutron evaporation using the UCD hypothesis :

$$A'p(Z) = A' \text{UCD} + \Delta A'(Z)$$

$\Delta A'(Z)$  the deviation from the UCD ratio, is fitted to the experimental data. The knowledge of the elemental yields  $YE(Z)$  results from a least square procedure where independent and mass yields are simultaneously taken into account. As an illustration of the method, we give on Figure 8 the reproduction of the mass yield curve for the spontaneous fission of  $^{259}\text{Md}$ . As for the other fissile isotopes the great influence of the charge  $50(\text{Sn})$  is clearly visible.

#### CHARGE DISTRIBUTION AT HIGHER NEUTRON ENERGY

The increase of excitation energy brought in by the incoming neutron produces change in the mass to charge ratio of the fission fragments. Coryell et al/18/ has proposed a method to evaluate this effect. The method consists of using the known  $Z_p$  function for thermal neutron fission of  $^{235}\text{U}$ , as a reference and calculating the change in  $Z_p$  from changes in the compound nucleus, and excitation energy.

$$Z_p(X) = Z_p(U5) + \Delta Z_p(X)$$

To account for these changes in  $Z_p$ , Nethaway /19/ has obtained from a least-square analysis of the available independent and cumulative yields data, values for the a,b,c, parameters of the following equation :

$$Z_p = a(Zc-92) + b(Ac-236) + c(E^*-6.52) \quad (4)$$

$E^*$  is the excitation energy of the compound nucleus. The odd-even effect has not been included in this equation.

Figure 9 shows the independent yields for  $^{52}\text{Te}$ ,  $^{53}\text{I}$ ,  $^{54}\text{Xe}$ ,  $^{55}\text{Cs}$ , fission products from 3 Mev neutron induced fission of  $^{235}\text{U}$  /12,20/. The dotted line are calculated from the  $Z_p$  model. The  $Z_p$  is derived from the equation 4. The experimental values are almost in agreement with the calculated ones. However yields for the even  $Z$  are higher than the calculated ones, and lower for the odd  $Z$ .

All the element yields measured by Hamelin et Al /20/ on 3 Mev neutron induced fission of  $^{235}\text{U}$  are shown on the figure 10. Here also the dotted lines are obtained using the  $Z_p$  model without EOF. The experimental technic used by the authors doesn't allow to measure all the isotopes for one element (The extrapolated part is always small,  $< 10\%$ ).

On the same figure, the element yields for the thermal neutron induced fission of  $^{235}\text{U}$  are also given. It is evident from the figure that the enhancement of the even  $Z$  has considerably decreased with the increase of the neutron energy.

To represent the odd-even effect, we can define a function

$$D\% = 100 (YE \text{ exp} - YE \text{ calc}) / YE \text{ calc.}$$

The YE calc is derived from a normal Gaussian distribution with  $Z_p$  calculated by the equation.(4)

The results are : =  $22 + 7\%$   $^{235}\text{U}$  (nth,f)  
                   =  $5 + 3\%$   $^{235}\text{U}$  (n3Mev,f)

Amiel /21/ had also derived from the analysis of 1.9 Mev neutron induced fission  $a = 8 + 4\%$ .

It seems clear now that the decreasing magnitude of the pairing effect is strongly correlated with increasing excitation energy of the compound system.

D.G. Madland et Al /22/ have assumed the existence of a correlation between saddle point excitation energy  $E_a$ , and the pairing effect.

$$E_a = (E^* + E_n) - E_{i,o}$$

$E^*$  = excitation energy of the compound system due to absorption of a zero energy neutron

$E_n$  = Incident neutron energy

$E_{i,o}$  = Inner, outer fission barrier heights in the double humped barrier model.

Using the values of pairing effect in thermal and 1.9 Mev neutron fission, they have derived formalism and parameters to

calculate the pairing effect in many systems.

#### MASS DISTRIBUTION FOR HIGHER ENERGY NEUTRON

The Argonne group have published results for  $^{232}\text{Th}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{238}\text{Pu}$  /23,24,25,26/. They used the Argonne fast neutron generator facility. Neutrons with energies below 5 Mev are produced by the  $^7\text{Li}(p,n)$  reaction and neutrons of higher energy by the  $^2\text{H}(d,n)$  reaction. So they are able to induce reactions with relatively monoenergetic neutrons. Some works for different neutron energy have also been published. Better systematics are obtained when the data are coming from the same source, in particular when the variation are very weak. So we'll restrict to these data to derive systematics in mass distribution. About 50 fission product yields have been determined for the fission of  $^{232}\text{Th}$ ,  $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{239}\text{Pu}$  with essentially monoenergetic neutrons of 0.17 to 3 Mev.

The main features apparent from the mass distribution shown in Figure 11 are the strong dependence of fission yields in the valley on  $E_n$  (increased probability of near symmetric fission with increasing excitation energy) and the weak dependence of peak yields on  $E_n$ . The mean mass for the light and the heavy group are plotted in figure 13 for the four nuclides.

The relative change in mean mass is greater for the heavy group than for the light group for the four nuclides. The Argonne group has calculated from conservation of mass, values of  $\bar{\nu}$ , the average number of neutrons emitted for fission. Their values are in general in good agreement with those evaluated from experimental measurements by fission coincident neutron counting.

The Figure 12 shows the ratio of  $^{115}\text{Cd}$  to  $^{140}\text{Ba}$  yields as a function of excitation for the fission of  $^{233}\text{Th}$ ,  $^{236}\text{U}$ ,  $^{239}\text{U}$ , and  $^{240}\text{Pu}$  in the region where only first chance fission occurs. The slope of this yield ratio is very similar for the two U, and smaller for the Pu. The authors have tried to derive dissipation energy, i.e. the energy transformed from collective energy into particules excitations as the fissioning nucleus moves from its saddle to its scission configuration.

This dissipation seems also correlated to the odd-even effect.  
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## Figure captions

Figure 1 : Average masses of the light and heavy fission product groups as functions of the masses of the fissioning nucleus

Figure 2 : Charge distribution for  $^{233}\text{U}$  (nth,f)

Figure 3 : Charge distribution for  $^{239}\text{Pu}$  (nth,f)

Figure 4 : Comparison of the RMS widths for the thermal neutron induced fission of  $^{235}\text{U}/^{239}\text{Pu}$

Figure 5 : Variation of EOZ as a function of the charge of the fissile system

Figure 6 : Variation of  $A(ZL)$  values as a function of  $AF/ZF$

Figure 7 : Comparison between experimental and calculated mass distribution of  $^{229}\text{Th}$

Figure 8 : Mass- and independent-yield curves for fragments from spontaneous fission of  $^{259}\text{Md}$ , is for even  $Z$ ,... is for odd  $Z$ , and .x.x.x. are for  $Z = 50$  and  $51$ , respectively.

Figure 9 : Independent yields for Te,I,Xe,Cs in the 3Mev neutron induced fission

Figure 10 : Element yields distribution in 3 Mev and thermal fission of  $^{235}\text{U}$

Figure 11 :  $^{239}\text{Pu}(n,f)$  mass distributions. The solid curves represent the results of present measurements. The 14 Mev (dashed curve) data were taken from Ref. 1

Figure 12 : The ratio of  $^{115}\text{Cd}$  to  $^{140}\text{Ba}$  yields as a function of excitation energy for the fission of  $^{233}\text{Th}$ ,  $^{236}\text{U}$ ,  $^{239}\text{U}$ , and  $^{240}\text{Pu}$  in the region where only first-chance fission occurs. The curves are merely to guide the eye.

Figure 13 : Average masses of the light and heavy fission groups as function of the neutron energy.

TABLE 1

Neutron even-odd effect as a function of the fissioning system

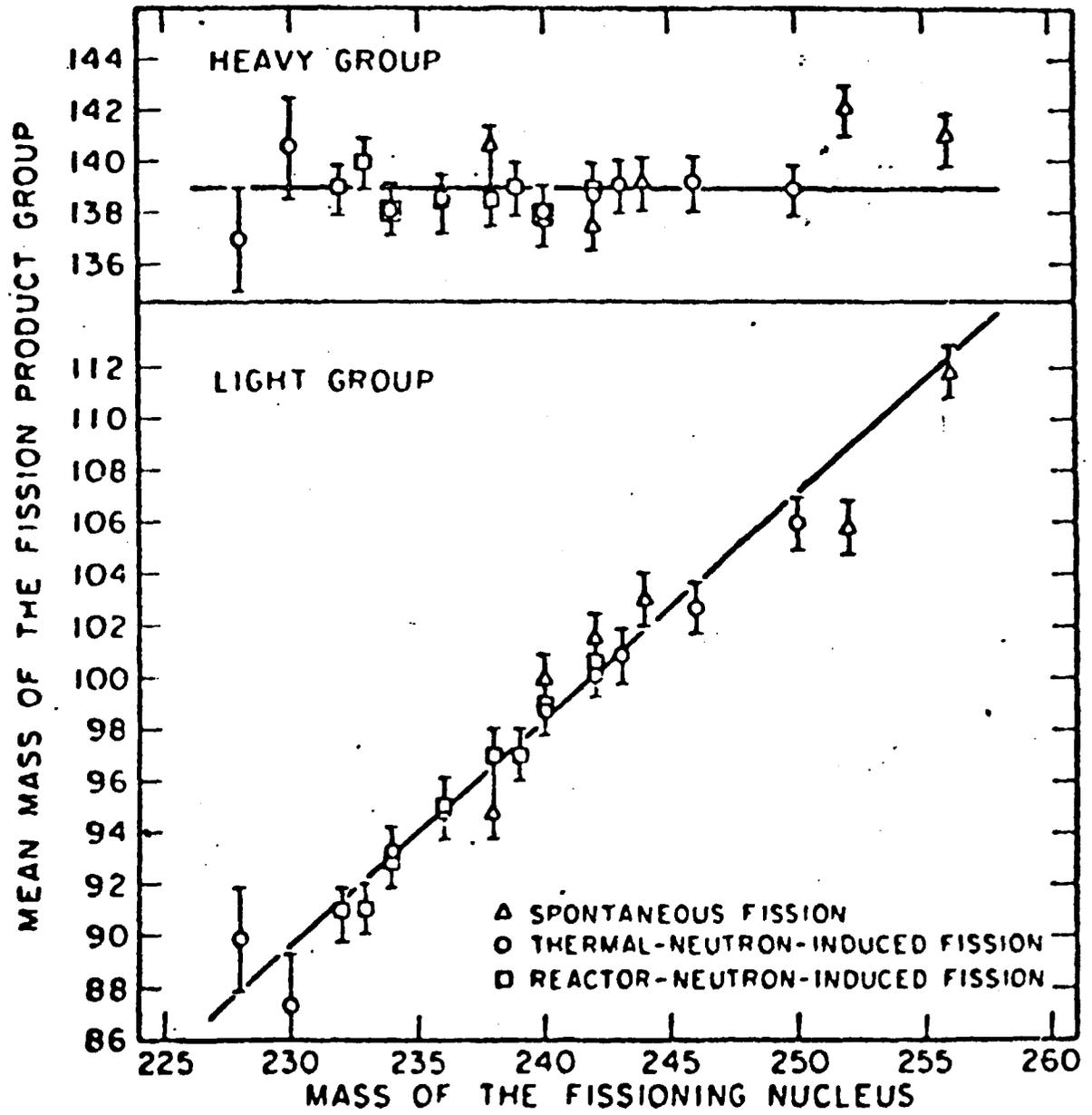
	$^{233}\text{U}+\text{nth}$	$^{235}\text{U}+\text{nth}$	$^{239}\text{Pu}+\text{nth}$	TH, U+50Mev P
E.O.N. %	5.4+1.7	5.4+0.7	6.5+0.6	5 + 2
Reference	7	10	8	14

TABLE 2

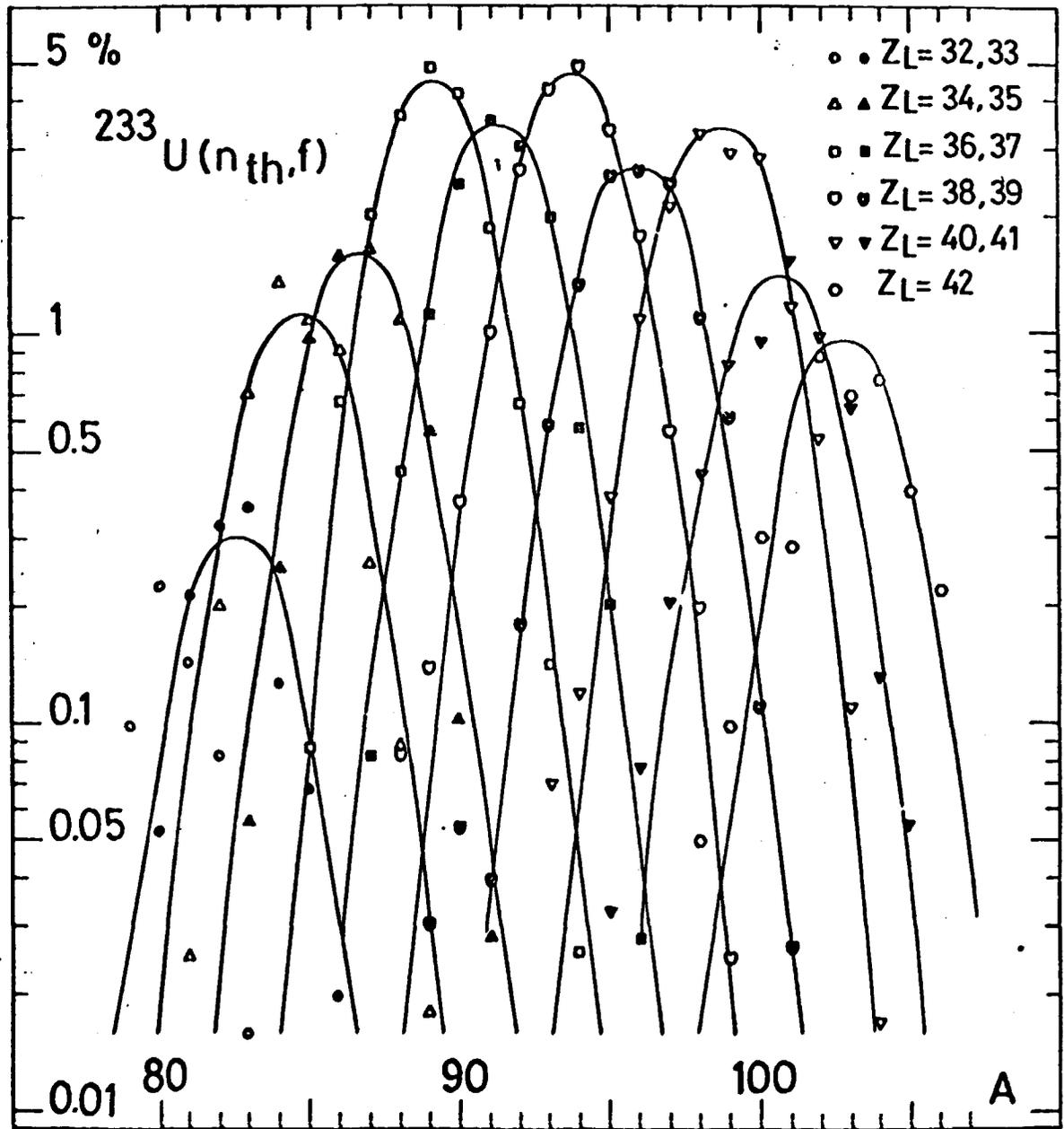
Parameters of the elemental distributions : experimental values

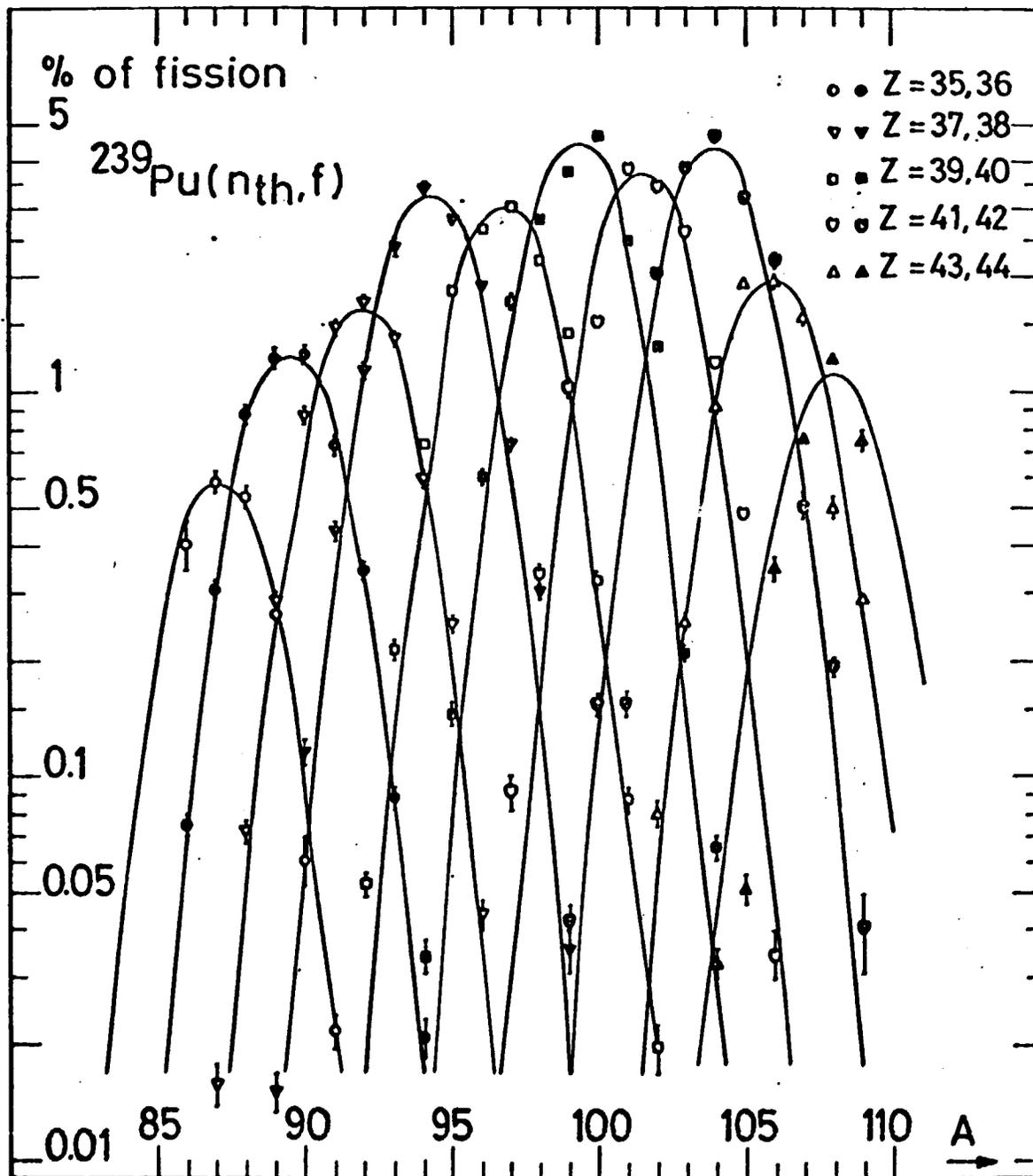
<sup>233</sup> U(nth,f)			<sup>235</sup> U(nth,f)			<sup>239</sup> Pu(nth,f)		
ZL,ZH	AL*	YE(ZL)	ZL,ZH	AL	YE(ZL)	ZL,ZH	AL	YE(ZL)%
32,60	80.4	0.62	32,60			34,60	85.	1.5
33,59	82.5	1.26	33,59	83.16	0.9	35,59	87.25	2.24
34,58	84.6	4.6	34,58	85.48	4.1	36,58	89.55	4.79
35,57	86.7	6.35	35,57	87.60	4.9	37,57	91.9	6.36
36,56	89.03	18.	36,56	89.93	15.1	38,56	94.25	12.51
37,55	91.23	13.3	37,55	92.16	12.0	39,55	96.8	11.96
38,54	93.7	19.3	38,54	94.72	18.8	40,54	99.3	16.92
39,53	95.92	11.7	39,53	96.99	11.9	41,53	101.6	13.80
40,52	98.58	14.85	40,52	99.63	17.8	42,52	104.0	16.20
41,51	100.58	5.85	41,51	101.4	7.1	43,51	106.0	7.83
42,50	102.9	3.67	42,50	103.8	4.4	44,50	108.0	4.2

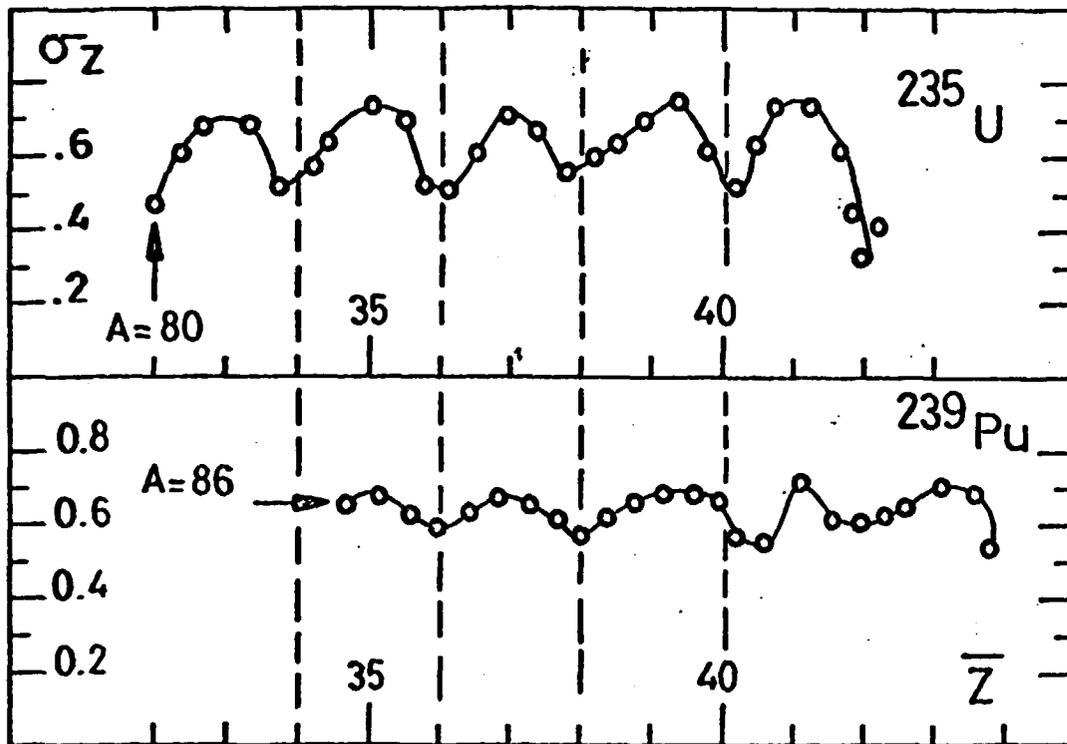
\* The typical uncertainty on AL value is .1 AMU



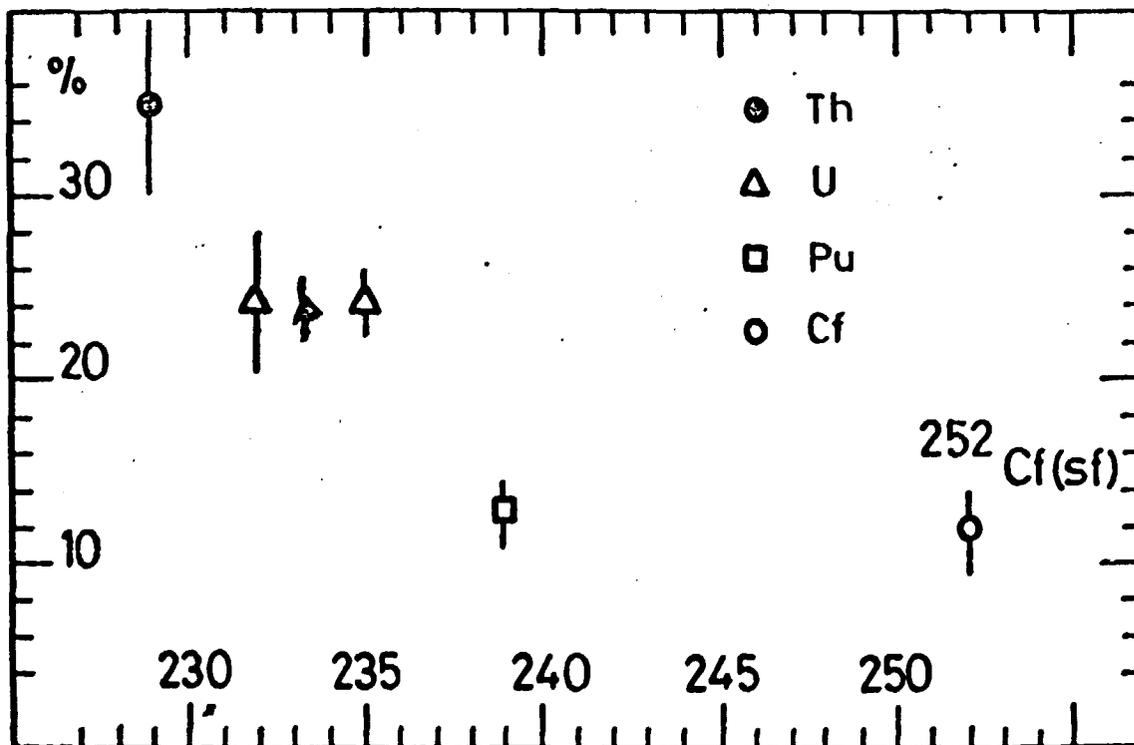
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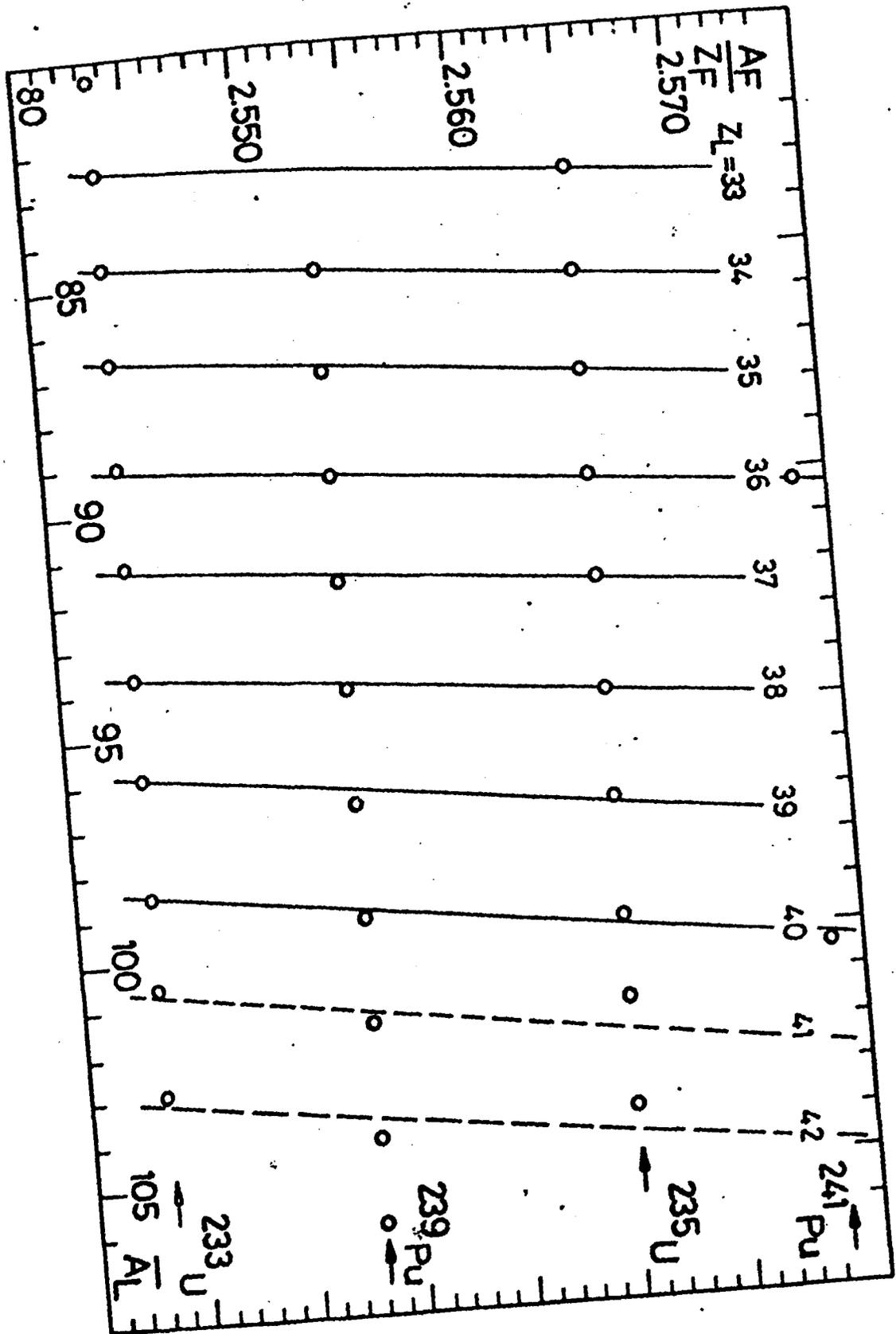


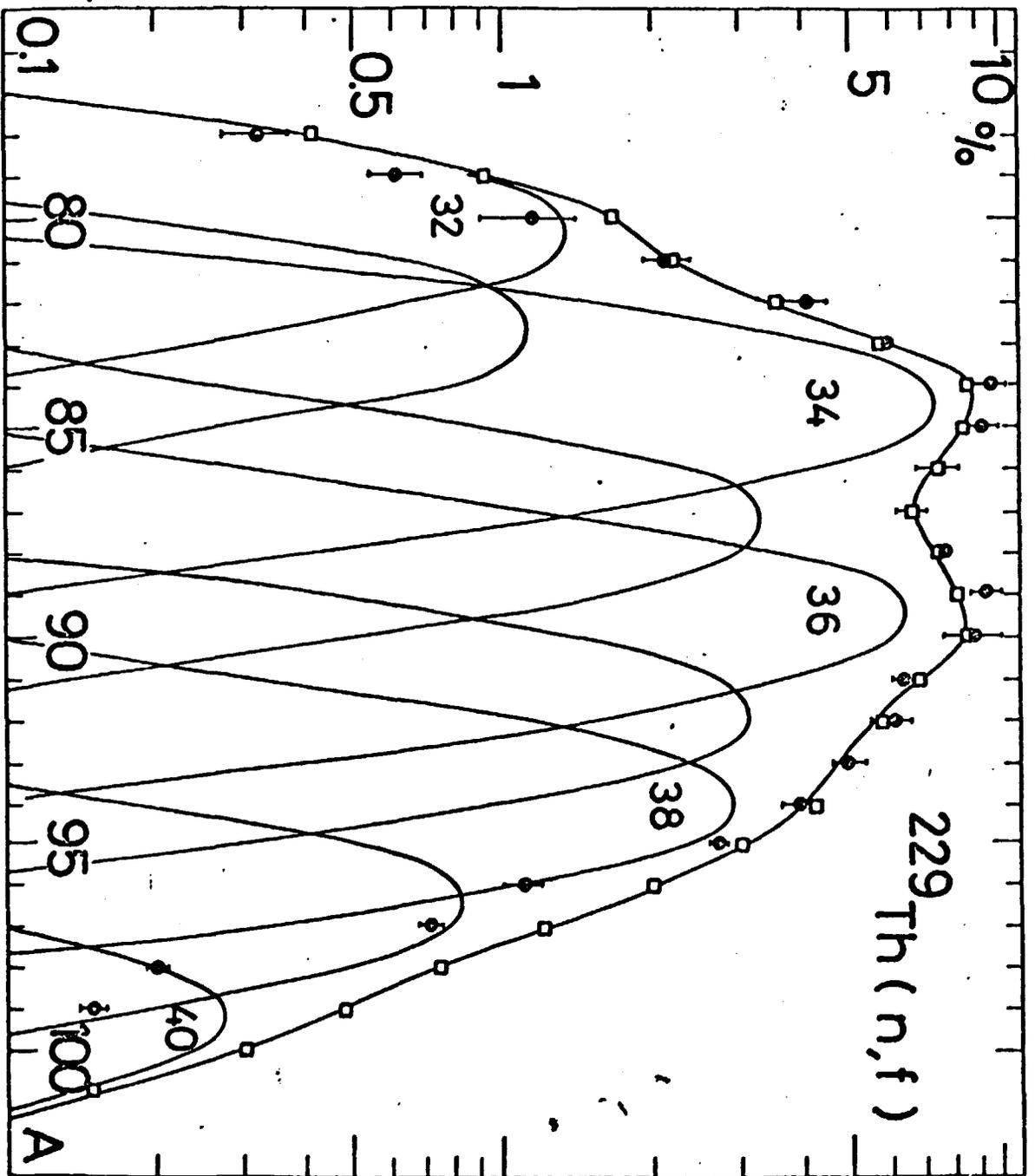


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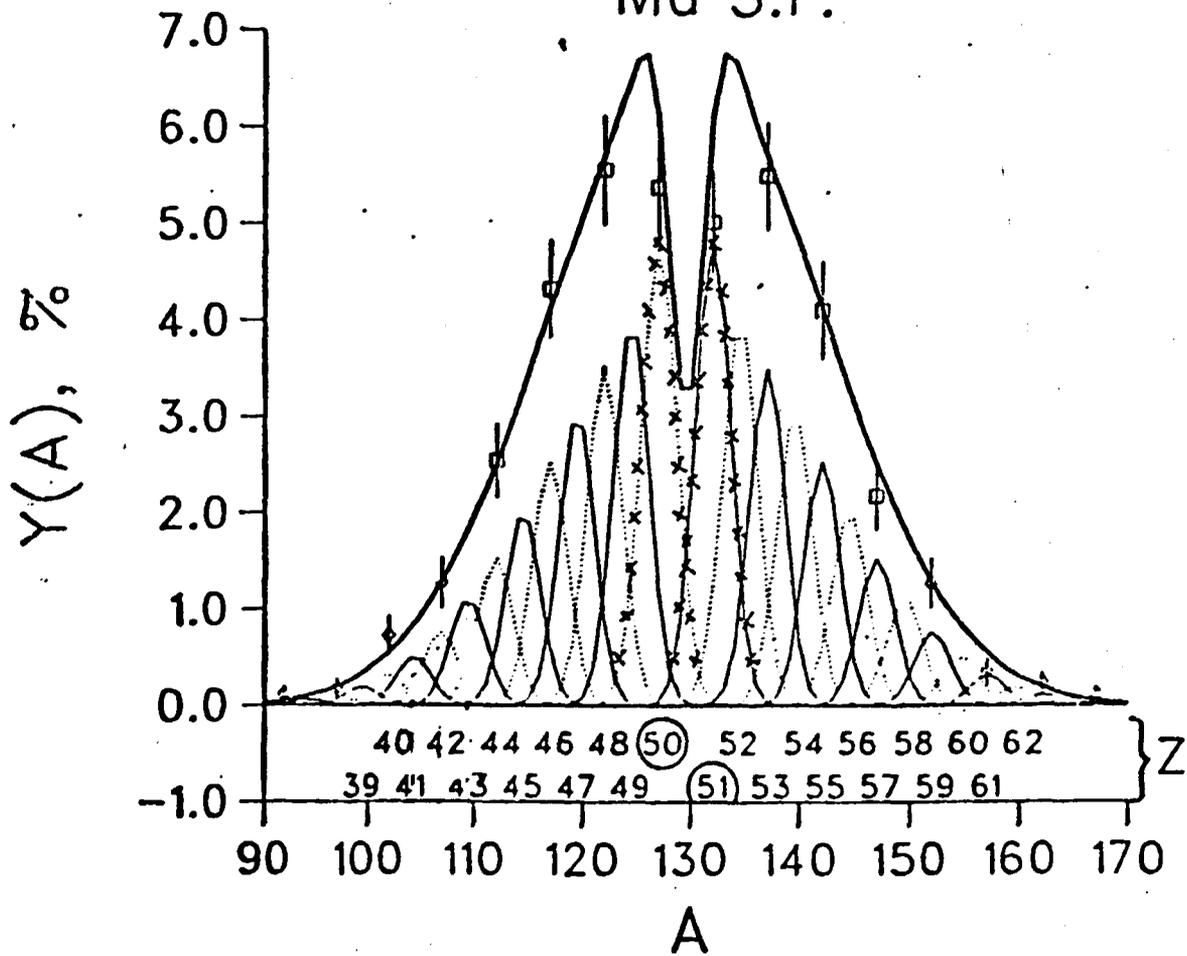
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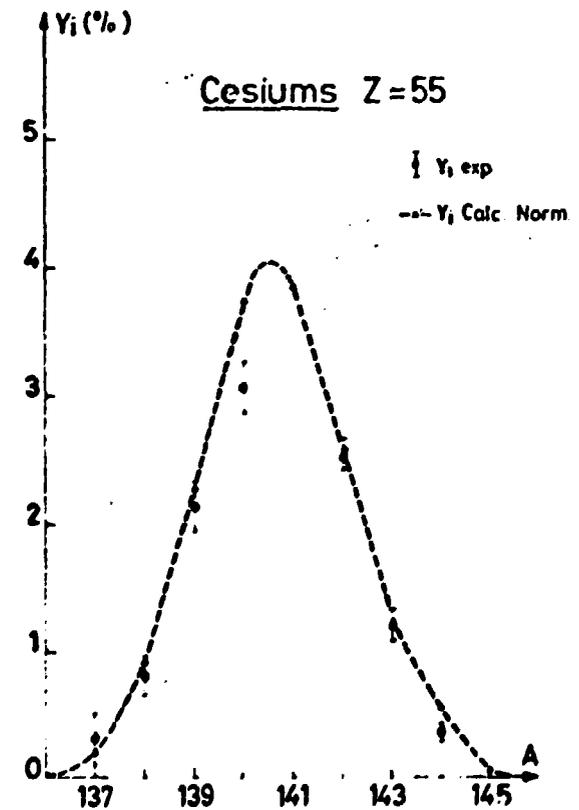
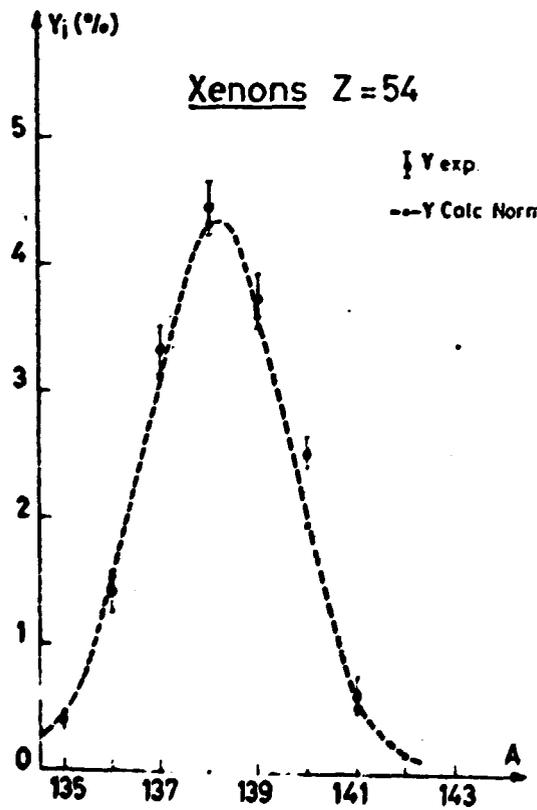
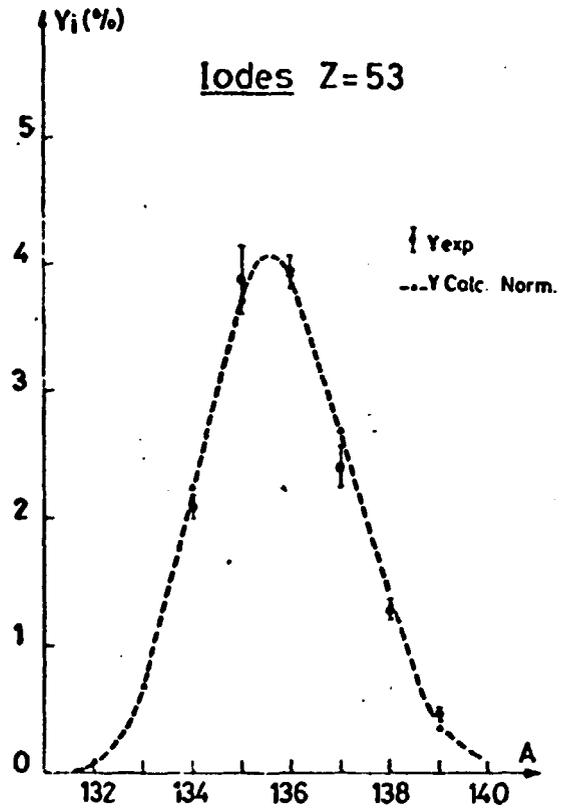
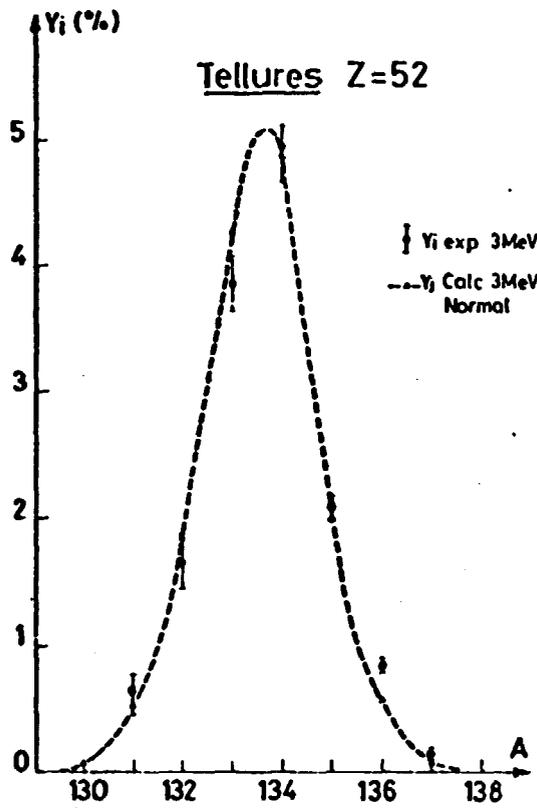


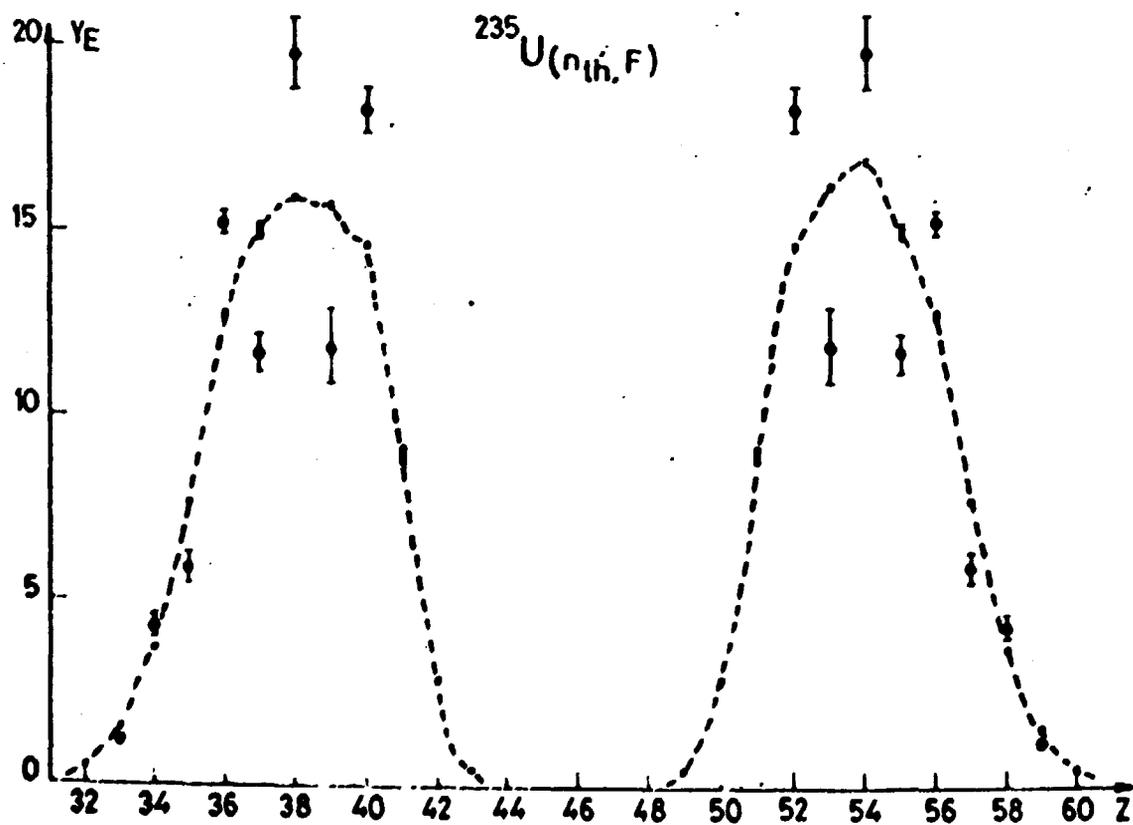
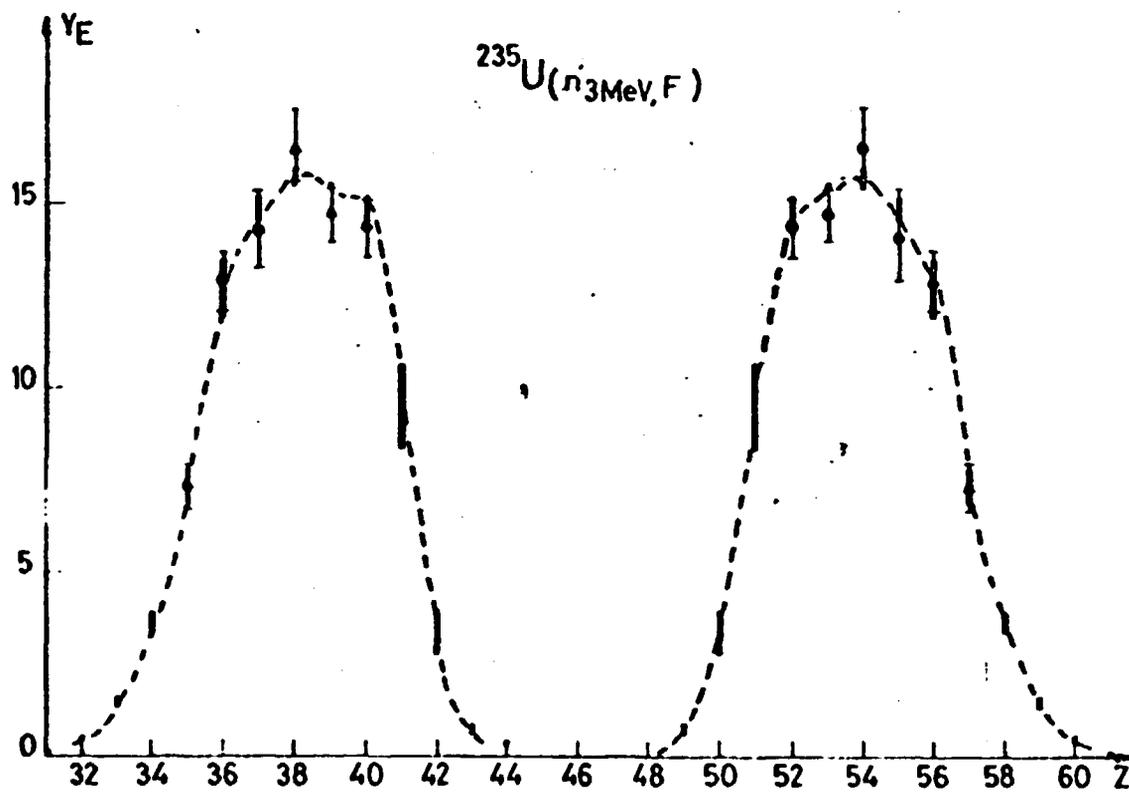


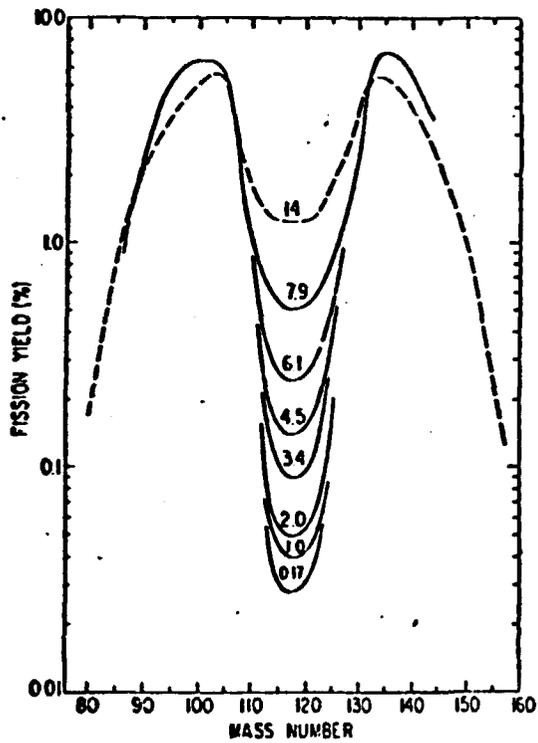
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$^{259}\text{Md}$  S.F.

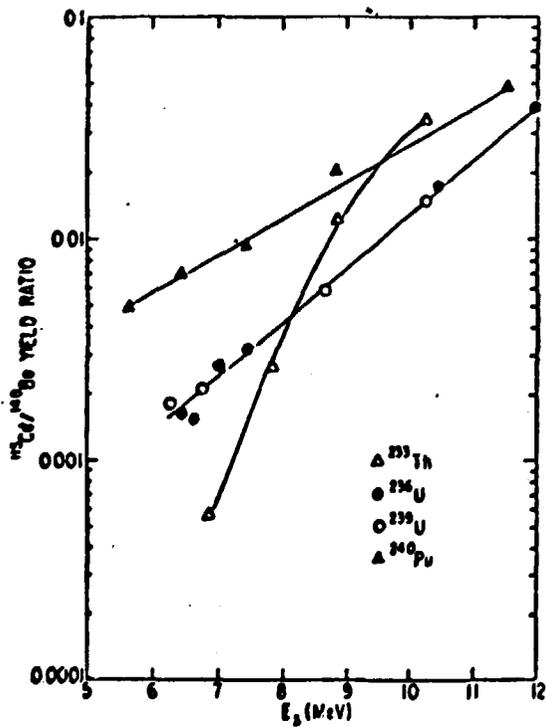




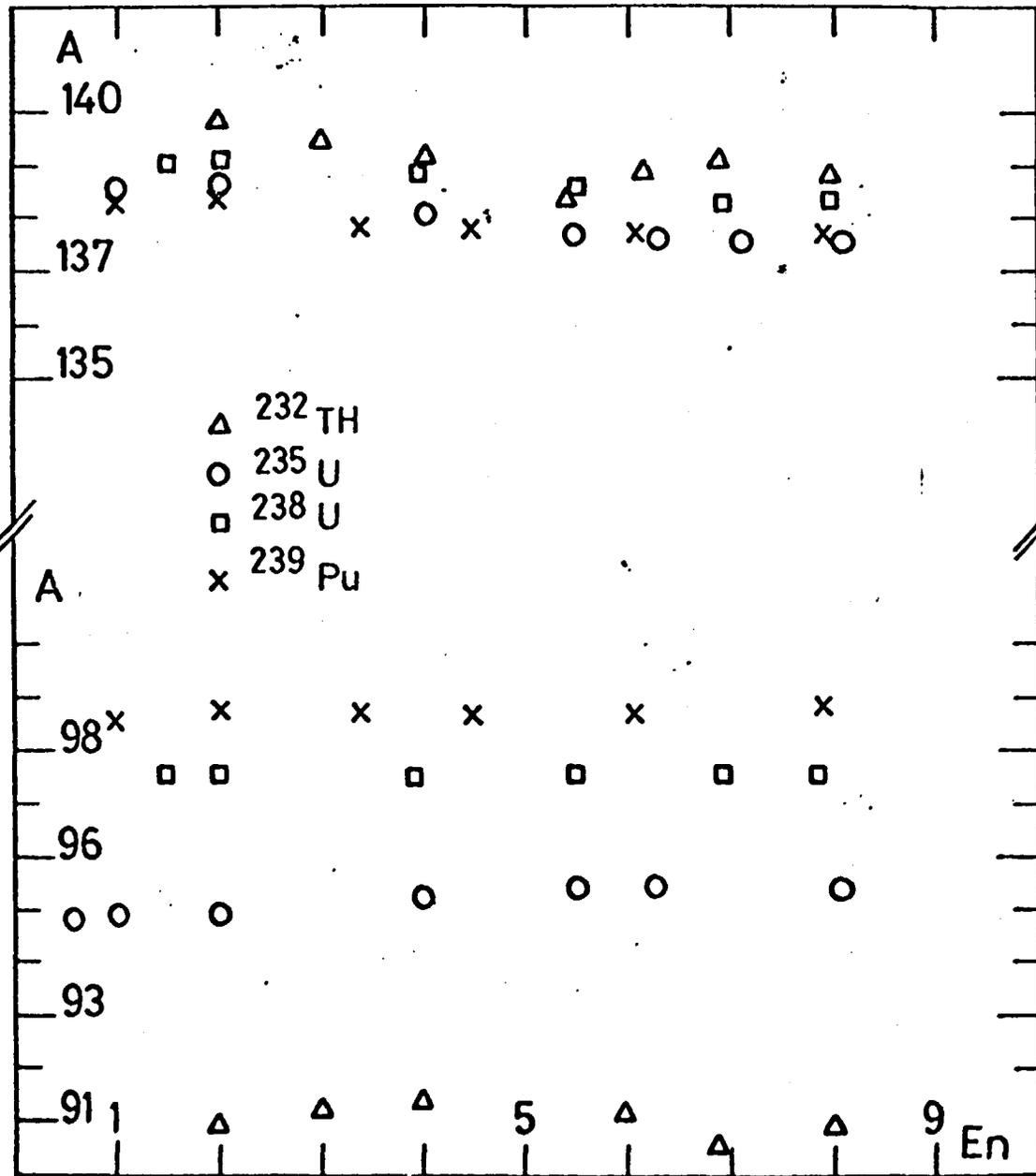




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