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Shoulders in the very asymmetric mass distribution for reactor-neutron-induced fission of ^{238}U

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New experimental data have been obtained on the cumulative yields of some short-lived very asymmetric products in the reactor-neutron-induced fission of ^{238}U. Fission yields of ^{73}Ga($t_{1/2}$ = 4.8 h), ^{167}Ho($t_{1/2}$ = 3.1 h), ^{171}Er($t_{1/2}$ = 7.5 h), ^{173}Tm($t_{1/2}$ = 8.2 h), and ^{179}Lu($t_{1/2}$ = 4.6 h) are reported. These yields together with those reported earlier by us for other relatively longer lived products (half-lives of the order of a few days to a week) for the mass chains 66, 67, 72, 77, 161, 172, 175, 177, 183, and 199 have been used to obtain the mass yield curve of ^{238}U which gives rise to "shoulders" in the highly asymmetric region. This is the first experimental observation of shoulders in this region of mass distribution. The data have been discussed in terms of the recent fragmentation potential calculations available in the literature. The presence of these new shoulders in the highly asymmetric mass region has been ascribed to the occurrence of a new potential energy valley in the far asymmetric mass region due to the influence of the 28-proton shell. The effect of deformed shells on mass distribution has also been discussed.

[NUCLEAR REACTIONS Fission ^{238}U, reactor neutrons, fragmentation potential, 28-proton shell, deformed shells, highly asymmetric mass distribution.]

INTRODUCTION

The mass distribution in the reactor-neutron-induced fission of uranium and other heavy elements is much less defined in the highly asymmetric region, i.e., $70 \geq A \geq 160$ as compared to the asymmetric and symmetric regions. The non-availability of data in this mass region is mainly because of the extremely low yields of the products ($10^{-6}\%$– $10^{-8}\%$) and the necessity of using stringent radiochemical separation procedures for the isolation of extremely low activities from highly radioactive irradiated targets. The only data in the above mass region for reactor neutron induced fission of ^{238}U are those reported earlier from this laboratory for some very asymmetric products.[1] Additional data have now been obtained for the mass chains 73, 167, 171, 173, and 179. These results, together with our earlier results have been used for the construction of the mass yield curve in the highly asymmetric region. Some interesting new features observed in this region of mass distribution are discussed in this paper.

EXPERIMENTAL

Details of the experimental procedure have been reported elsewhere[1] and are briefly summarized below.

A. Target irradiations and radiochemical separation

Nuclear pure uranyl nitrate was exhaustively purified by repeated anion exchange steps followed by TBP extraction. Spectrographic as well as activation analysis of the impurity fractions resulting from the purification of the target and reagent blank were carried out to ascertain the absence of impurities in the target material which might give rise to the products of interest by activation. For example, the formation of ^{171}Er by the reaction ^{170}Er(n, γ) ^{171}Er and ^{179}Lu by the reaction ^{179}Hf(n, p) ^{179}Lu are ruled out as is evident from target impurity analysis (where Hafnium and heavier rare earths were found to be absent) which has been described in detail in our earlier paper.[1] ^{167}Ho and ^{173}Tm can result from ^{167}Er(n, p) ^{167}Ho and ^{173}Yb(n, p) ^{173}Tm, respectively. The nuclide ^{73}Ga can result from successive neutron capture by ^{71}Ga and also from ^{73}Ge(n, p) ^{73}Ga reaction. Such possibilities for the formation of these products from these reactions have been found to be negligible under the experimental conditions. The ^{235}U content of the target was found to be 0.217% by fission track method.[2] About 1 g of the sample wrapped in 1 mm thick Cd foil was sealed in PVC bags and irradiated for periods ranging from 8–10 h in the core position of the APSARA swimming pool reactor at Trombay, at a fast (> 1 MeV) flux of ~10^{11} n/cm^2 sec. The nature of neutron flux in the position of irradiation is the same as described earlier.[1] The products selected for radiochemical isolation were ^{73}Ga, ^{167}Ho, ^{171}Er, ^{173}Tm, and ^{179}Lu which have short half lives varying from 3–8 h. Hence sufficient amounts of product activities could be isolated and identified unambiguously with reasonable counting statistics. About 20–30 mg carriers of each of these elements along with 20 mg of Mo carrier were added to the target solution. Bulk rare earths of interest were separated by an ion exchange method using α-hydroxy isobutyric acid. The detailed separation procedure is being published elsewhere.[3] The rare earths were finally precipitated as oxalates and mounted for counting.

In the case of ^{73}Ga, separate irradiations were performed. A solvent extraction method[4] using di-isopropyl ether and thenoyl triflouroacetone was employed for the separation and purification of the product. Gallium was finally mounted in the form of oxinate for counting. Since ^{99}Mo was employed as an internal standard, it was also isolated by a standard radiochemical procedure and counted.

B. Counting and evaluation of fission yields

The samples were counted in a low background β proportional counter with anticoincidence shielding (background count rate 0.9 to 1.0 cpm). The radiochemical purity was established by following the decay of the sample for nearly three half lives. Figures 1(a) and 1(b) show typical decay curves of ^{171}Er and ^{73}Ga. In the case of ^{167}Ho and ^{73}Ga, the radiochemical purity was also established by γ ray spectra obtained using a Ge (Li)-multichannel analyzer system [see Figs. 2(a) and 2(b)].

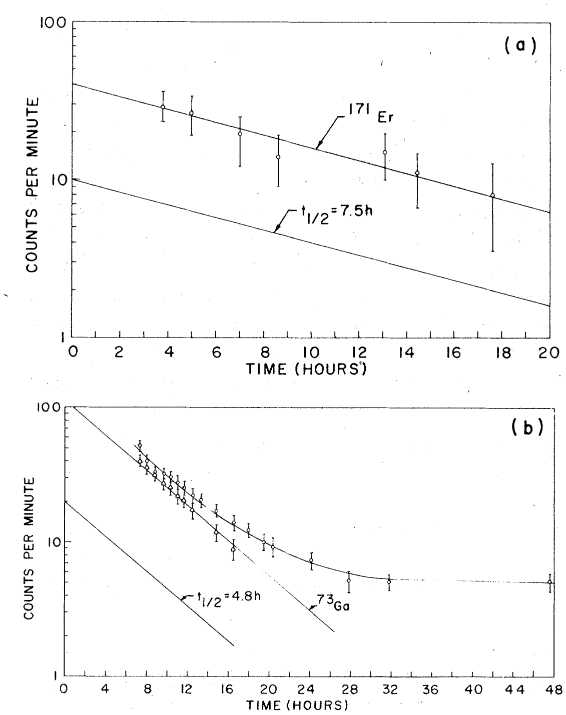

FIG. 1. β decay curves: 1(a) and 1(b) represent the decay curves of ^{171}Er and ^{73}Ga, respectively.

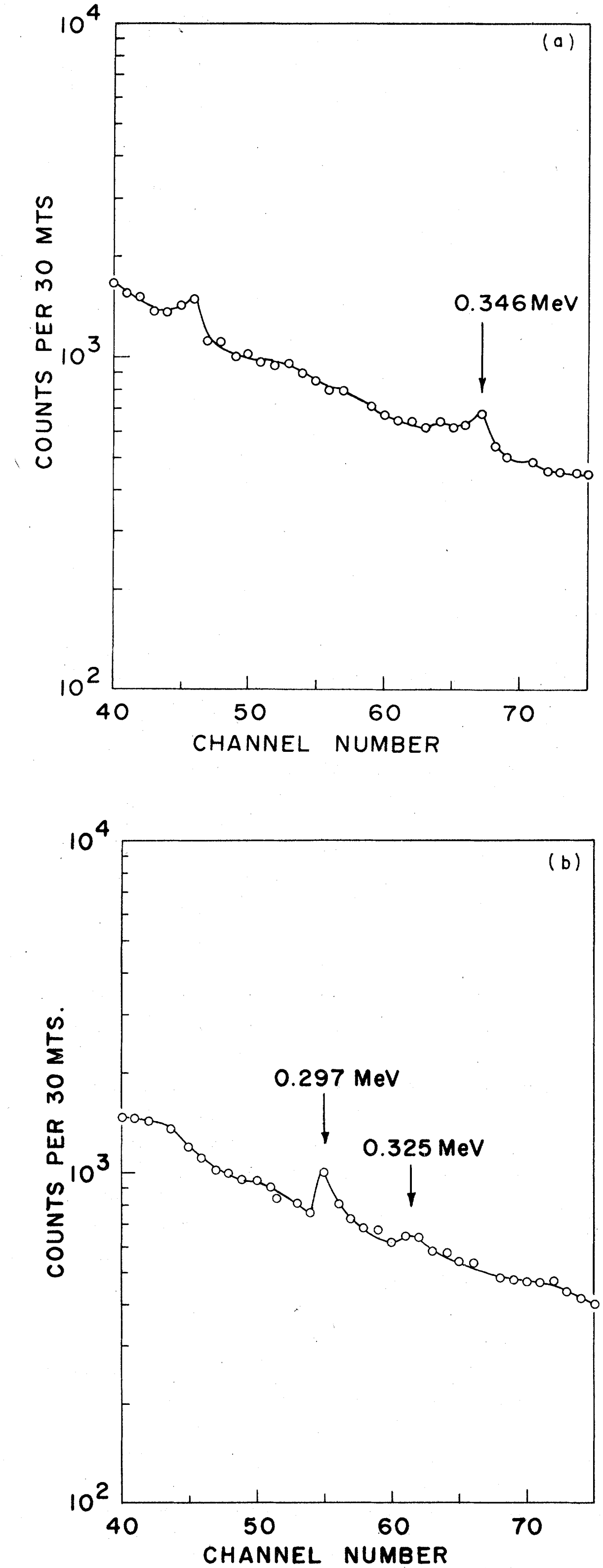

FIG. 2. γ ray spectra: 2(a) and 2(b) represent the γ ray spectra of ^{167}Ho and ^{73}Ga, respectively.

The activities were corrected for chemical yields, decay, and efficiency of the counting setup. Fission yields were calculated relative to the yield of ^{99}Mo using the equation

$$Y_i(\%) = Y_{^{99}\mathrm{Mo}}(\%)\,\frac{N_i}{N_{^{99}\mathrm{Mo}}}, \quad (1)$$

where Y_i is the fission yield of the particular nuclide i produced in the irradiation and N_i is its number of atoms formed. $Y_{^{99}\mathrm{Mo}}$ and $N_{^{99}\mathrm{Mo}}$ are the corresponding yield and number of atoms of ^{99}Mo formed. The number of atoms N_i were calculated using the relation

$$N_i = \frac{A_i \cdot t}{1 - e^{-\lambda_i t}}, \quad (2)$$

where A_i is the activity [in disintegrations per minute (dpm)] associated with the nuclide i at the end of the bombardment, and t is the duration of irradiation in minutes. The yield for ^{99}Mo was assumed to be 6.2%. The observed isotopic yields were corrected for independent yields to obtain the total isobaric yields using Wahl's[5] charge distribution correction. Contributions from ^{235}U fast fission were estimated to be negligible.

RESULTS AND DISCUSSION

The fission yields of 15 mass chains in the highly asymmetric region have been determined and listed in Table I. These include the yields reported earlier[1] and those for the mass chains 73, 167, 171, 173 and 179 from the present study. The number of nuclides chosen in the heavier wing are more because they were amenable to lengthy radiochemical separations on account of their convenient half lives and chemical behavior. These data were used to construct the mass yield curve in the highly asymmetric region.

In view of the very low activities involved, an uncertainty of ±40% is assumed for the measured yields of all the products except ^{73}Ga, ^{77}As, ^{161}Tb, and ^{167}Ho. An uncertainty of ±20% is estimated for these products as sufficiently large quantities of activities (a few hundred dpm) could be isolated in these cases. These uncertainties include the experimental errors associated with radiochemical methods such as the errors due to (i) chemical yield determinations, (ii) uncertainties in counting efficiencies, (iii) decay systematics of the nuclide involved, and finally (iv) due to counting statistics.

Figure 3 represents the mass yield curve for the reactor neutron induced fission of ^{238}U constructed using the available data in the literature[6] for the mass range $A \geq 70$ and $A \leq 160$. The curve has been extrapolated to the masses $A = 60$ on the lighter side and $A = 180$ on the heavier side. This has been accomplished using the Gaussian distribution of the type

$$Y = \frac{F}{\sqrt{2\pi}\,\sigma}\exp\left[-\frac{1}{2}\left(\frac{A-\bar{A}}{\sigma}\right)^2\right], \quad (3)$$

TABLE I. Experimental data on the yields of some highly asymmetric products in the reactor neutron induced fission of ^{238}U.

Mass Number	Nuclide	Fission Yield (%)
66	^{66}Ni	$(4.01 \pm 1.6) \times 10^{-6}$ [a]
67	^{67}Cu	$(2.32 \pm 0.93) \times 10^{-5}$ [a]
72	^{72}Zn	$(6.54 \pm 2.50) \times 10^{-5}$ [a]
73	^{73}Ga	$(2.19 \pm 0.44) \times 10^{-4}$
77	^{77}As	$(1.51 \pm 0.30) \times 10^{-3}$ [b]
161	^{161}Tb	$(6.45 \pm 1.29) \times 10^{-4}$ [b]
167	^{167}Ho	$(4.08 \pm 0.80) \times 10^{-5}$
171	^{171}Er	$(5.04 \pm 2.0) \times 10^{-6}$
172	^{172}Er	$(9.40 \pm 3.76) \times 10^{-6}$ [a]
173	^{173}Tm	$(6.17 \pm 2.50) \times 10^{-6}$
175	^{175}Yb	$(8.00 \pm 3.20) \times 10^{-6}$ [a]
177	^{177}Lu	$(6.60 \pm 2.64) \times 10^{-6}$ [a]
179	^{179}Lu	$\leq 2.41 \times 10^{-7}$
183	^{183}Ta	$\leq 1.13 \times 10^{-8}$ [a]
199	^{199}Au	$\leq 4.66 \times 10^{-9}$ [a]

[a] Data taken from our earlier work (Ref. 1).
[b] The uncertainties in these yields have been reevaluated and revised to ±20%.

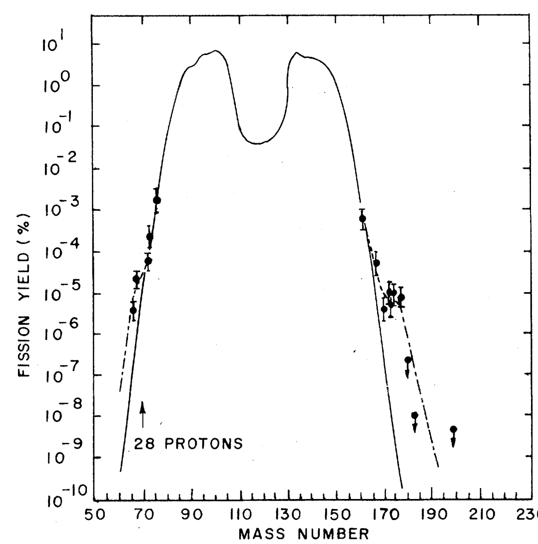

FIG. 3. Mass distribution in the fission of ^{238}U by reactor neutrons. The curve has been extrapolated using the available data (Ref. 6) to the mass numbers $A = 60$ on the the lighter side and $A = 180$ on the heavier side by means of Eq. (3) referred to in the text. Dotted portion is drawn using the present experimental data in Table I.

where Y is the computed yield, σ is the width of the Gaussian (7.3), F is a normalizing factor, and $\bar{A} = 118.18$ assuming a prompt neutron emission of 2.64. The dotted portion in Fig. 3 is drawn using the experimental data given in Table I. It can be seen that the yields for mass chains 66–67 on the lighter side and 172–177 on the heavier side are about three orders of magnitude higher than the computed values. This large increase in the mass yields as compared to computed values is beyond experimental uncertainties. The mass distribution in this mass region has been depicted separately in Figs. 4(a) and 4(b) for the lighter and heavier wing portions, respectively. It is seen from these figures that the tendency for the yields to increase in this region, particularly in the heavier wing where the data are more extensive, does not continue much beond $A = 177$ as is evident from the upper limits for the yields of ^{179}Lu, ^{183}Ta, and ^{199}Au. Thus an examination of the wing portions of the mass yield curve shows that the yields first decrease smoothly, then show a slight increase around $A \sim 66$–67 and $A \sim 172$–177 and again show a fall. This observation leads us to believe that there are "shoulders" or "bumps" in the low yield wing portions of the mass yield curve. A preliminary observation of this effect was reported by us earlier.[7] The new yields for the short-lived products, i.e., ^{179}Lu, ^{173}Tm, ^{171}Er and ^{167}Ho on the heavier side and ^{73}Ga on the lighter side tend to confirm unambiguously the presence of "shoulders" or "bumps" in this region of very asymmetric mass distribution.

It is well known that shells play an important role in fission mass distribution. For example the familiar asymmetric mass division, fixation of the heavier peak position and the presence of "spikes", etc, are best understood in terms of the influence of the 82-neutron shell in fission. There is a 28-proton shell in the $A \sim 66$–67 region where the yields are found to be higher than expected, resulting in the appearance of a shoulder in this region. The presence of this shoulder can be viewed as being due to the possible influence of the 28-proton core which forces the yields to be higher in this region in much the same way as the 82-neutron core forces the yields to be higher around $A \sim 132$. Since no data are available on neutron evaporation in the highly asymmetric mass division, estimates were made for the fission of ^{238}U induced by reactor neutrons. The method of calculation is similar to the one reported by Mac Murdo and Cobble.[8] The total excitation energy (E_x) available for neutron emission is assumed to be the difference (ΔE) between the total mass energy released in fission (E) and the sum of the total kinetic energy (E_k) and the energy removed

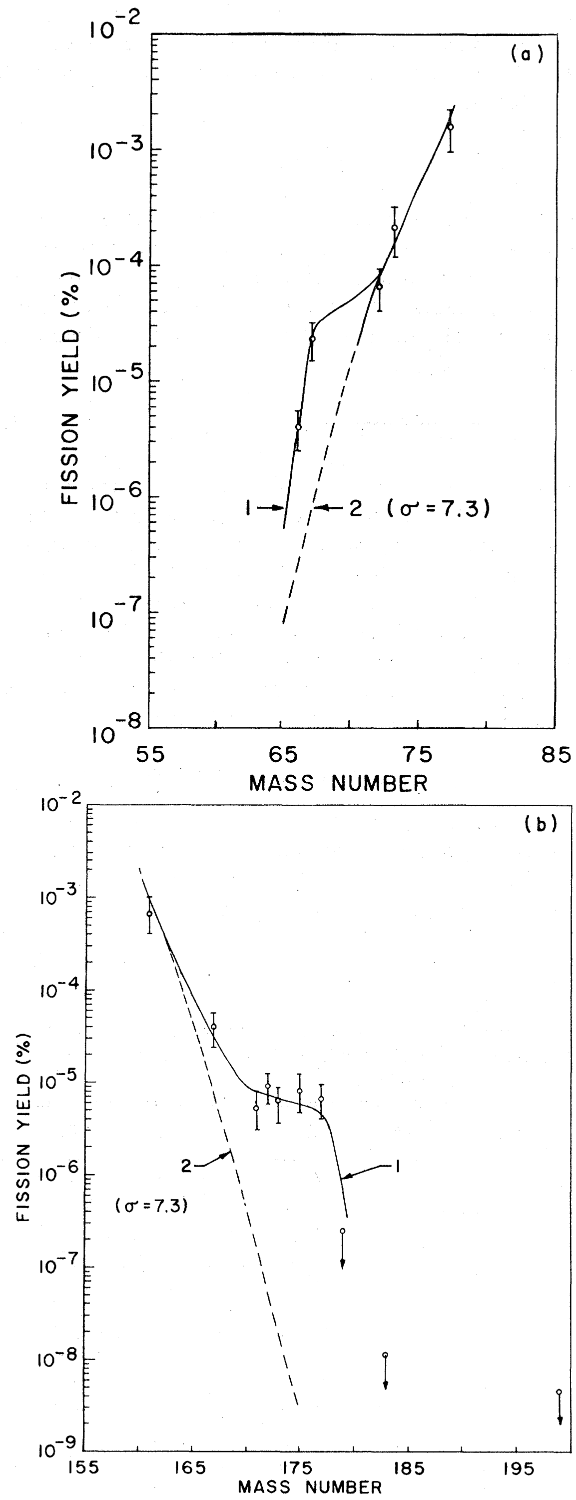

FIG. 4. Mass distribution in the highly asymmetric region in the fission of ^{238}U by reactor neutrons: (a) and (b) represent the lighter and heavier wings, respectively. Solid line 1 is drawn using the present experimental data and dotted line 2 shows the extrapolated curve using Eq. (3) referred to in the text.

by prompt γ rays (~7.5 MeV). An average kinetic energy of 1.5 MeV was assumed to be associated with the reactor neutron inducing the fission and was taken into account while computing the total excitation energy. ΔE is estimated using the mass tables of Hillman.[9] These calculations showed negligible neutron evaporation for mass divisions around $A_L \sim 66-67$ and $A_H \sim 172-177$. In view of this, it is very likely that the shoulder on the heavier side in the mass range 172–177 is the reflection of the shoulder around $A \sim 66-67$ and is complementary in nature. The influence of the 28-proton shell is obviously much weaker than that of the 82-neutron shell.

We have examined the possibility of the 28-proton shell influencing the fission mass distribution in terms of the yield calculations of the Frankfurt group based on the fragmentation theory.[10-13] These calculations are available for ^{226}Ra, ^{236}U, and ^{258}Fm.[10,12,13] Very recently, these calculations are available for ^{238}U and ^{252}No and have been reported by Sandulescu *et al.*[14] It has been shown in these calculations that new mass asymmetry valleys appear in the fragmentation potential $V(l,\eta)$ as a function of length l (elongation of the compound nucleus) and mass asymmetry coordinate $\eta = (A_1 - A_2/A_1 + A_2)$ where A_1 and A_2 represent the mass numbers of the two fragments and $A_1 + A_2$ represents the mass number of the compound nucleus. Calculations were made for a wide range of η values ($0 \leq \eta \leq 0.8$). These valleys were considered to arise due to the correct treatment of shell corrections such that for separated fragments, the shell corrections equal the sum of the shell corrections of individual fragments. This has been done by weighing each level with the probability that its wave function is found in the corresponding fragment and by computing the shell correction for each fragment separately. Sandulescu *et al.*[14] ascribe the new minima in the fragmentation potential to the shell corrections for the smaller fragment with $N = 50$ and $Z = 28$ or maybe $N = 28$ and $Z = 28$. The theoretically calculated yields along with our radiochemical data on the yields of very asymmetric products for ^{238}U fission have been discussed.[14] There is good agreement in the position of shoulders with theoretical prediction. However, the intensities of the peaks differ by many orders of magnitude. This may be due to the dependence of the general behavior of the peaks on the parameters l and θ_0 (temperature of the compound nucleus) used in their calculation.

In another calculation[15] for ^{236}U fission, such additional peaks were predicted where the dynamical effects of collective friction while running down the potential barrier were studied. It has been shown that for no friction as well as for very strong friction the calculated yields change very little whereas for intermediate values, the final distribution shows fine structure. This is apparently due to the kinetic energy term in the Hamiltonian. Thus in addition to the 28-proton shell effect, the apparent small asymmetry (noncomplementarity) in the low yield shoulders of heavy and light masses (see Fig. 3) could arise due to dynamical effects of the kinetic energy or friction effects. These theoretical calculations lend credence to the experimentally observed shoulders in the highly asymmetric mass distribution. In yet another calculation on the basis of the order-disorder model, Ganguly *et al.*[16,17] computed independent yields of fission products using available experimental data as input. These calculations also showed a small peak around the 28-proton number.

It may be observed that the yields of highly asymmetric fission products are very much energy dependent in the same way as the yields of symmetric products.[1] If we assume that shell effects (28-proton) are responsible for the observed shoulders in the fast fission of ^{238}U, then we can make some interesting speculations on the influence of excitation energy in deciding the presence or absence of shoulders in fission mass distribution. The shell effects are more prominent and preseved in a better way at lower energies, e.g., thermal neutron fission of heavy elements. However, there will be a sharp decrease in the yields of the products with decrease in the excitation energy of the compound nucleus and it would be difficult to make yield measurements and observe the "shoulders", say in the thermal neutron fission of ^{235}U. In fact none of the very asymmetric products that were isolated in the fast fission of ^{238}U could be detected[1] in the thermal neutron fission of ^{235}U. On the other hand, at higher excitation energies, say in the 14 MeV neutron fission, even though the yields are higher and easily measurable, it is possible that the influence of the 28-proton shell (which is much weaker than the 82-neutron shell) is almost completely washed off and therefore the shoulders are not observable.[18,19] These speculations are depicted in Fig. 5 where the yield values shown are only order of magnitude estimates. Thus it can be seen that in the case of fission mass distribution of ^{238}U by reactor neutrons, there are influences of (i) the 82-neutron shell giving rise to the familiar asymmetric distribution and (ii) the 28-proton shell resulting in the occurrence of "shoulders" in the highly asymmetric region. This idea may be extended a little further to speculate on the possibility of an 82-proton shell influencing fission mass distribution which might give rise to additional shoulders around $A = 208$ and the com-

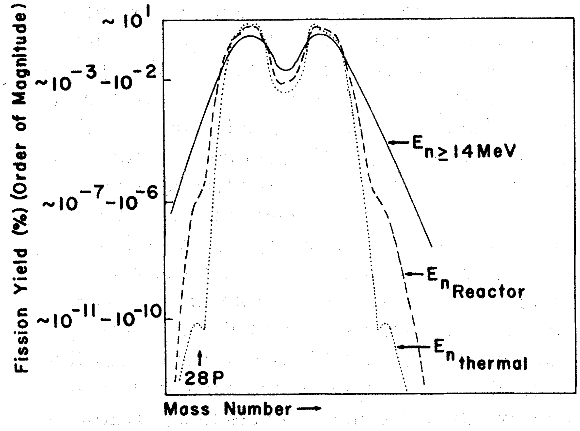

FIG. 5. Effect of neutron energy in fission mass distribution. The dotted, dashed, and solid curves correspond, respectively, to thermal-, reactor-, and 14 MeV neutron-induced fission.

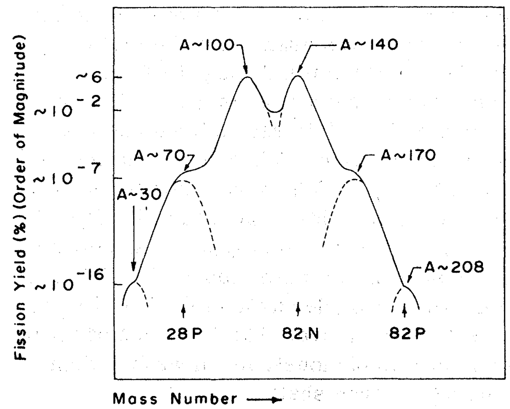

FIG. 6. Possible influence of the nucleon shells (28*P*, 82 *N*, 82 *P*) in fission mass distribution.

plementary region around $A = 30$. From the known trends of the variation of fission yield values with asymmetry of mass division, the yields in these mass regions are, however, expected to be extremely small, making experimental measurements very difficult. Considering the effects of various closed shells, the mass distribution in fission may be wavy in nature as depicted in Fig. 6 where again the yields shown are only order of magnitude estimates.

As an alternative explanation for the occurrence of the "shoulders" in fission mass distribution, we have considered the possibility of deformed shells[20] influencing the mass distribution in the far asymmetric mass split. From a consideration of the deformation stiffness, the heavy fragment resulting from the very asymmetric mass split will be highly deformed while the light fragment may tend to be close to spherical. Wilkins *et al.*[20] have considered the influence of deformed neutron and proton shells in mass distribution and explain several phenomena such as the single, double, and triple humped distribution, etc. The calculations which are based on a static scission point model predict new deformed shells corresponding to $N \sim 66$, 88, 106, etc. and $Z \sim 38$, 44, 66, etc. The deformed proton shell at $Z \sim 66$ (where deformation parameter $\beta = 1.0$) and neutron shell at $N \sim 106$ ($\beta = 0.8$) are of interest to us in the present studies.

With a view to analyzing our yield data based on this model, the nuclei studied by us were converted back to their primary neutron and proton numbers prior to neutron evaporation (2.5 to 3.0 neutrons[21]) and β decay. Based on the assumption that (i) neutron evaporation occurs mainly from the heavy fragment, (ii) N/Z ratio of the two fragments is equal to Z_{UCD} shifted by 0.5 Z units towards the light fragment due to the Coulomb interaction term, the products can then be shown to arise from the following primary fragments as given in Table II. At $\beta = 1.0$, there is a strong proton shell centered at $Z = 66$ and a fairly strong neutron shell at $N = 106$. N/Z (106/66) of these two shells is nearly identical to compound system (147/92). These two shells add coherently to lower the potential energy of the total system, thereby enhancing the yield in the mass region around $A = 172$ and the shoulder in the lighter mass region is considered as a reflection of the one on the heavier side. The strength and position of these deformed shells are not known accurately due to the limitations in these calcula-

TABLE II. Calculated primary fragment *N* and *Z* values for the nuclides studied.

	Primary		Primary Complement	
Nuclide	*Z*	*N*	*Z*	*N*
^{66}Ni	26	40	66	107
^{67}Cu	26	41	66	106
^{72}Zn	28	44	64	103
^{73}Ga	29	44	63	103
^{77}As	30	47	62	100
^{161}Tb	63	101	29	46
^{167}Ho	65	105	27	42
^{171}Er	67	107	25	40
^{172}Er	67	108	25	39
^{173}Tm	67	109	25	38
^{175}Yb	68	110	24	37
^{177}Lu	69	110	23	37

Data in Table II were obtained from Wilkins (Ref. 21).

tions.[21] However, there seems to be good qualitative agreement within the uncertainties associated with the sensitivity of shell corrections.

In conclusion, it may be said that our first experimental observation of shoulders in the far asymmetric mass region in the neutron induced fission of ^{238}U can be considered to arise either from the influence of the relatively strong 28-proton shell or from the presence of nucleonic shells in the deformed heavier fragment. A precise answer to the problem could come from new experimental data such as highly asymmetric mass distribution studies at low excitation energies (where shell effects are best preserved) using monoenergetic (1–4 MeV) high flux neutrons and also from spontaneous fission of ^{252}Cf and other heavier actinides. Studies on $^{232}Th(n,f)$ or $^{244}Pu(n,f)$ in which N/Z ratio is similar to the ratio $N = 106$ and $Z = 66$ shells also can resolve the problem of deformed shell effects influencing the highly asymmetric mass distribution.

SUMMARY

We have measured radiochemically the cumulative yields of the mass chains 73, 167, 171, 173, and 179 in the reactor neutron induced fission of depleted uranium. These data together with the data for the mass chains 66, 67, 72, 77, 172, 175, and 177 have been used to construct the mass yield curve for the neutron fission of ^{238}U. The yields of these highly asymmetric products which are in the range 10^{-5}–10^{-7}% show the presence of bumps or shoulders in this region which is the first experimental observation.

The data have been interpreted in terms of the recently published fragmentation potential energy calculations of the Frankfurt Group for ^{238}U system. The presence of these new asymmetric shoulders has been ascribed to the formation of new valley in the fragmentation potential as a result of the influence of the 28-proton shell. The effect of deformed shells based on the scission point model of nuclear fission could also lead to enhanced yields of the heavier fragment at $A \sim 172$ due to new shells around $N \sim 106$ and $Z \sim 66$ in the heavier fragment. An interesting speculation of a wavy nature of the mass distribution based on various shell effects has been made.

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