Pramāṇa – J. Phys., Vol. 26, No. 3, March 1986, pp. 179–189. © Printed in India.

# Correlation between fractional independent yields and neutron-to-proton ratio of fission products in low energy fission

A GOSWAMI, B K SRIVASTAVA, ALOK SRIVASTAVA, S B MANOHAR, SATYA PRAKASH and M V RAMANIAH
Radiochemistry Division, Bhabha Atomic Research Centre, Trombay, Bombay 400085, India

MS received 13 August 1985

**Abstract.** Fractional independent yields of fission products in the thermal neutron-induced fission of $^{233}U$, $^{235}U$, $^{239}Pu$, $^{241}Pu$ and in the spontaneous fission of $^{252}Cf$ have been correlated with the neutron-to-proton ratio of the fission products. The yields of the products from a fissioning system, when plotted as a function of neutron-to-proton $(N/Z)$ ratio of fission products, fall on two Gaussian distribution corresponding to light and heavy fission products. The centroids of the distribution or the most probable value of neutron-to-proton ratio is found to be very close to the $N/Z$ of the fissioning nucleus. From the most probable value of $N/Z$ the various parameters of charge distribution e.g. most probable mass $A_p$, most probable charge $Z_p$, the mass dispersion $\sigma_A$ and the charge dispersion $\sigma_Z$ have been obtained and are in good agreement with the experimental values of $A_p$ and $Z_p$.

**Keywords.** Fractional independent yields; neutron-to-proton ratio; fission products; low energy fission.

**PACS No.** 25·85

## 1. Introduction

The charge distribution studies are generally carried out through investigation of the independent yields of the members of isotopic or isobaric chain. The yield profile thus obtained is of Gaussian nature and is characterized by the most probable charge $Z_p$ and mass $A_p$ and the width of distribution $\sigma_Z$ and $\sigma_A$ respectively. Attempts have been made to arrive at the systematics of charge distribution and several hypothesis exist regarding the mode of charge division, namely unchanged charge distribution (UCD), equal charge displacement (ECD) and minimum potential energy (MPE) (Glendenin 1949; Way and Wigner 1948). It is well known that the systematics of nuclear fission process is largely governed by liquid drop model energetics over which nuclear shell and pairing effects cause local perturbation which is reflected in the observed systematics of the asymmetric nature of mass distribution, and saw-tooth nature of neutron emission probabilities, higher formation probability of elements having even charge compared to those having odd charge etc. The importance of liquid drop potential has been pointed out by Wilkins *et al.* (1976) who demonstrated that the nuclear structural effects play an important role only when in phase with liquid drop minima as demanded by the asymmetry term. With this in view a large number of experimentally obtained independent yield data available in literature have been analyzed to search for an overall correlation between $N/Z$ of the products and their independent yield over the high mass yield region. An attempt was made earlier by Mukherjee (1969) to correlate independent yield of fission fragment with their $N/Z$ ratio. But in view of the limited data then available the conclusions were different from those arrived in the present work.

## 2. Calculation

The fractional independent yield FIY($Z$) for isobaric distribution and FIY($A$), for isotopic distribution is given by

$$\mathrm{FIY}(Z) = \frac{1}{\sqrt{2\pi\sigma_Z^2}} \int_{Z-0\cdot5}^{Z+0\cdot5} \exp[-(Z-Z_p)^2/2\sigma_Z^2]\, \mathrm{d}Z, \tag{1}$$

$$\mathrm{FIY}(A) = \frac{1}{\sqrt{2\pi\sigma_A^2}} \int_{A-0\cdot5}^{A+0\cdot5} \exp -[(A-A_p)^2/2\sigma_A^2]^2\, \mathrm{d}A, \tag{2}$$

where $Z$ and $A$ are the charge and mass of the fission products, $Z_p$ and $A_p$ are the most probable charge and most probable mass and $\sigma_Z$ and $\sigma_A$ are the charge and mass dispersion parameters respectively. In the present work a plot of experimentally determined FIY($A$) as a function of ($N/Z$) of light and heavy fission products from the thermal neutron-induced fission of $^{233}$U, $^{235}$U, $^{239}$Pu and $^{241}$Pu and in the spontaneous fission of $^{252}$Cf has been made. Figure 1 shows such a plot of FIY($A$) as a function of $N/Z$ for light and heavy fission products formed in the thermal neutron-induced fission of $^{235}$U. The yields are represented by two Gaussians corresponding to the light and heavy fission products respectively and FIY($A$) can be given by:

$$\mathrm{FIY}(A) = \frac{1}{(2\pi\sigma_{N/Z}^2)^{1/2}} \exp -[\{(N/Z)-(N/Z)_p^2\}/(2\sigma_{N/Z}^2)], \tag{3}$$

where $(N/Z)$ is for the product and $(N/Z)_p$ is the most probable value and $\sigma_{N/Z}$ is the width parameter assumed to be constant for all Z. The $(N/Z)_p$ value thus obtained is related to $A_p$, $Z_p$, $\sigma_A$ and $\sigma_Z$ as follows. Equation (3) can be rearranged to give

$$\mathrm{FIY}(A) = \frac{1}{(2\pi\sigma_{N/Z}^2)^{1/2}} \exp -[\{(1+N/Z)-(1+N/Z)_p\}^2/2\sigma_{N/Z}^2], \tag{4}$$

$$\mathrm{FIY}(A) = \frac{1}{(2\pi\sigma_{N/Z}^2)^{1/2}} \exp -[(A-A_p)^2/(2Z^2\sigma_{N/Z}^2)]. \tag{5}$$

A comparison of (2), (4) and (5) leads to the following relations:

$$A_p = \{1+(N/Z)_p\} \cdot Z, \tag{6}$$

$$\sigma_A = \sigma_{N/Z} Z. \tag{7}$$

Assuming a linear correlation between $A_p$ and $Z_p$ the value of the most probable charge $Z_p$ and charge distribution parameter $\sigma_Z$ is obtained as

$$Z_p = A/\{1+(N/Z)_p\}, \tag{8}$$

$$\sigma_Z = \sigma_{N/Z} \cdot A/\{1+(N/Z)_p\}^2. \tag{9}$$

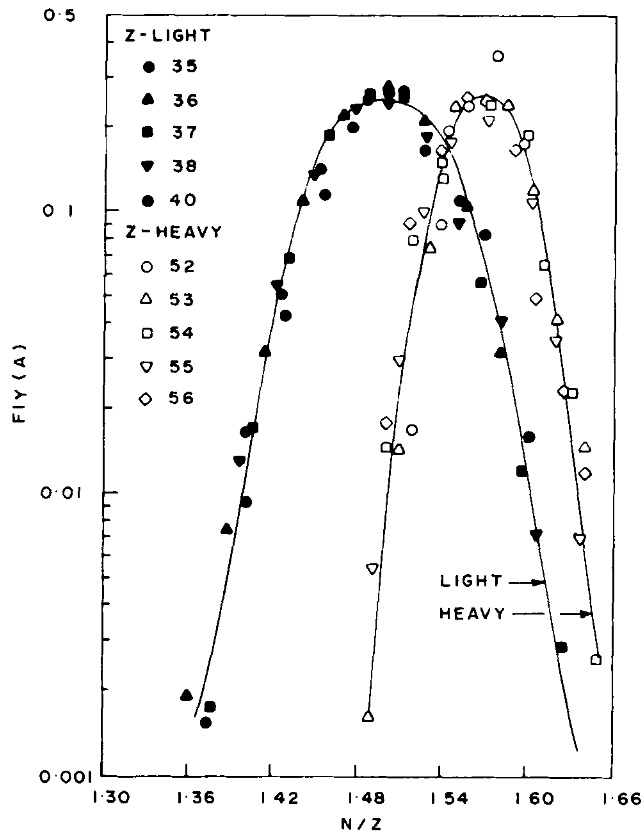


**Figure 1.** FIY($A$) vs ($N/Z$) of fission products from $^{236}U$ fission.

Thus $A_p$, $Z_p$, $\sigma_A$ and $\sigma_Z$ are correlated with the most probable value $(N/Z)_p$ and $\sigma_{N/Z}$ of the products in the light and heavy wing. In general the experimentally reported values are in the form of fractional independent yield FIY($Z$). These are converted to isotopic yield value by using the conventional relation

$$\mathrm{IY} = \mathrm{FIY}(Z)\cdot \mathrm{CY}, \tag{10}$$

$$\mathrm{FIY}(A) = \mathrm{IY}/Y_Z, \quad \mathrm{FCY}(A) = \sum_{i=1}^{n} \mathrm{FIY}(A)_i, \tag{11}$$

where IY and CY are the independent and total chain yield respectively. The charge yield $Y_Z$ is obtained by adding all the independent yields of a given charge. The values of centroid $(N/Z)_p$ and the width of distribution $\sigma_{N/Z}$ for the light and heavy fission products were obtained from the probability plot of the isotopic fractional cumulative yield FCY($A$) as a function of ($N/Z$) as shown in figure 2. The value of $(N/Z)_p$ and $\sigma_{N/Z}$ for both the light and heavy fission products for different fissioning nucleus are given in table 1. Substituting the values of $(N/Z)_p$ and $\sigma_{N/Z}$ in (6) and (7) respectively the values of $A_p$ and $\sigma_A$ were obtained for the light and heavy fission products. Using (6) to (9) a

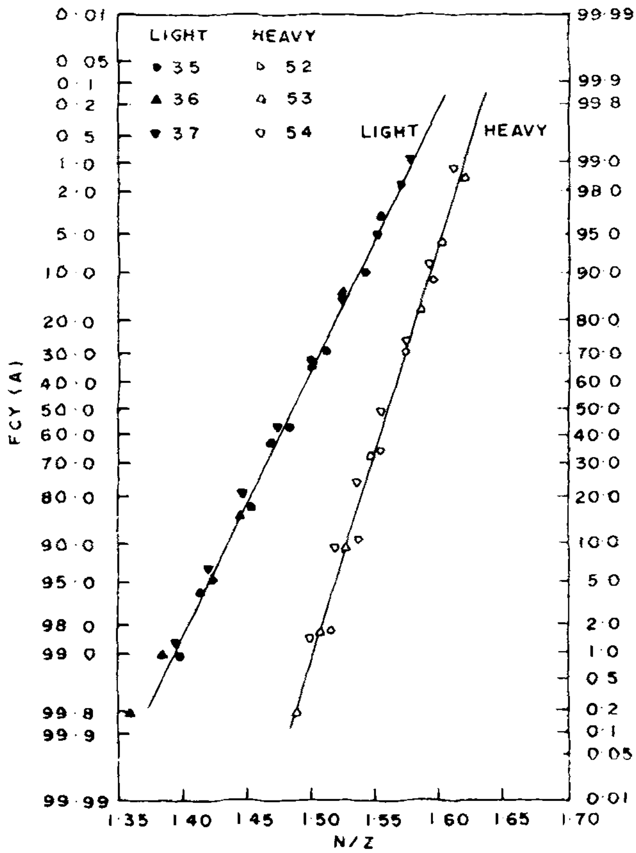


**Figure 2.** Probability plot of FCY(*A*) vs (*N*/*Z*) of fission products from $^{236}U$ fission.

**Table 1.** $(N/Z)p$ and $\sigma_{N/Z}$ for various fissioning nuclei.

| Fission products | $(N/Z)_p$ | $\sigma_{N/Z}$ | $(N/Z)_f$ | $\Delta_{N/Z}$ |
|---|---|---|---|---|
| $^{235}U$: | | | 1·538 | |
| *Light* | 1·487 | 0·040 | | −0·051 |
| Heavy | 1·560 | 0·025 | | +0·022 |
| $^{233}U$: | | | 1·516 | |
| Light | 1·465 | 0·040 | | −0·051 |
| Heavy | 1·540 | 0·020 | | +0·024 |
| $^{239}Pu$: | | | 1·523 | |
| Light | 1·475 | 0·042 | | −0·048 |
| Heavy | 1·542 | 0·020 | | +0·019 |
| $^{241}Pu$: | | | 1·543 | |
| Light | 1·495 | 0·040 | | −0·048 |
| Heavy | 1·560 | 0·025 | | +0·017 |
| $^{252}Cf$: | | | 1·533 | |
| Light | 1·480 | 0·040 | | −0·052 |
| Heavy | 1·550 | 0·025 | | +0·018 |

systematic analysis of charge distribution parameters was carried out for the following systems:

2.1 $^{235}U(n_{th}, f)$

Fractional independent yields of both light and heavy fission products in the thermal neutron-induced fission of $^{235}$U have been reported by a number of workers using physical (Nifenecker *et al* 1980; Schmid *et al* 1981) as well as radiochemical (Wahl *et al* 1969) methods. In the present work the data have been taken from the recent compilation of Wahl (1983), which incorporates the data obtained by both physical and radiochemical methods. The chain yields have also been taken from the same compilation. A plot of FIY($A$) as a function of ($N/Z$) for a number of fission products is shown in figure 1. Two distinct Gaussians appear for the light and heavy fission products with their centroids giving the most probable value of ($N/Z$) for the light and

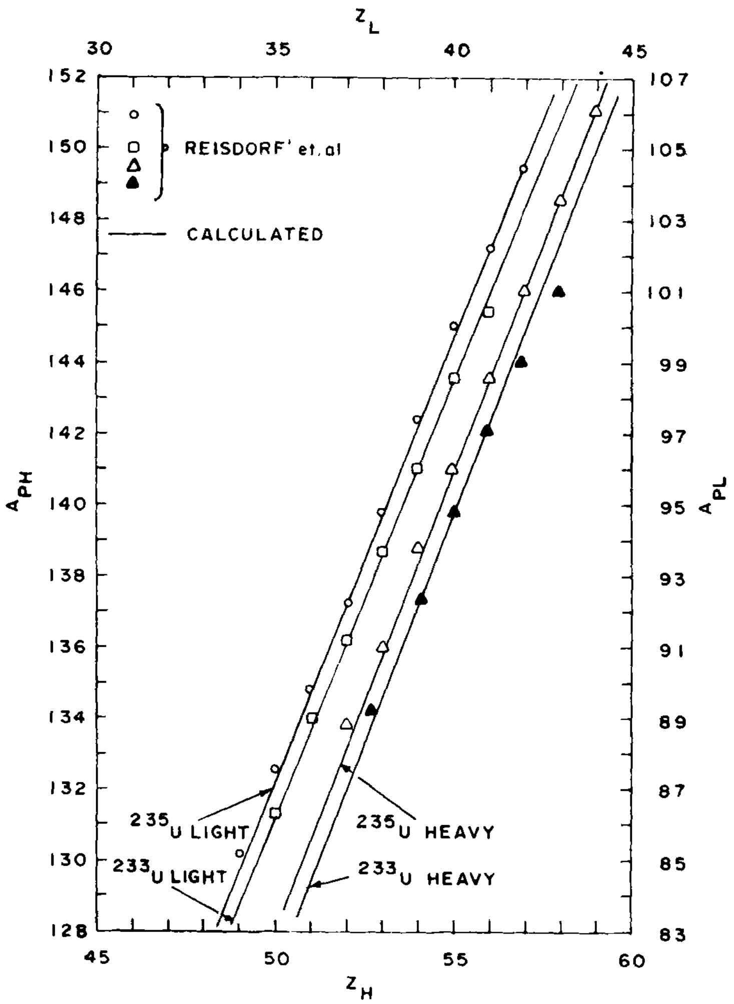


**Figure 3.** Most probable mass for light and heavy fission products.

heavy fission product respectively. The light fission products data are for charges $Z = 36$–$42$, while that of heavy fission products for $Z = 52$–$56$. Figure 3 shows a plot of the calculated values of $A_p$ along with experimental value from Reisdorf *et al* (1971). Fairly good agreement is shown between the calculated and the experimental value. Further the value of $Z_p$ was obtained using (8) for both light and heavy mass chains and is shown in figure 4 indicating a linear correlation between $A_p$ and $Z_p$.

## 2.2 $^{233}U(n_{th}, f)$

The fractional independent yield values of a number of light fission products from $Z = 35$ to $Z = 42$ has been measured using the HIWATHA mass separator (Wehring *et al* 1980). FIY($Z$)'s were converted to FIY($A$) using (10) and (11) and a plot of cumulative isotopic yield FCY($A$) against ($N/Z$) of fission products is shown in figure 5. It is evident that the Gaussian assumption is valid. Similar analysis was carried out for heavier fission products with $Z = 53$, 54 and 55. The values of $(N/Z)_p$ and $\sigma_{N/Z}$ are shown in table 1 for the light and heavy fission products. A comparison of the calculated and experimental values of $A_p$ and $Z_p$ are shown in figures 3 and 4 respectively.

## 2.3 $^{239}Pu(n_{th}, f)$ *and* $^{241}Pu(n_{th}, f)$

The experimental data on the fission of $^{240}$Pu and $^{242}$Pu are very scanty. Brissot *et al* (1977) determined the independent yield of gaseous fission products, bromine, iodine, krypton and xenon using mass separator. In the case of $^{239}$Pu some independent yield data of technetium and antimony are also available. All the data were analyzed in the same way as in the case of $^{236}$U and the most probable value of $(N/Z)_p$ and $\sigma_{N/Z}$ so

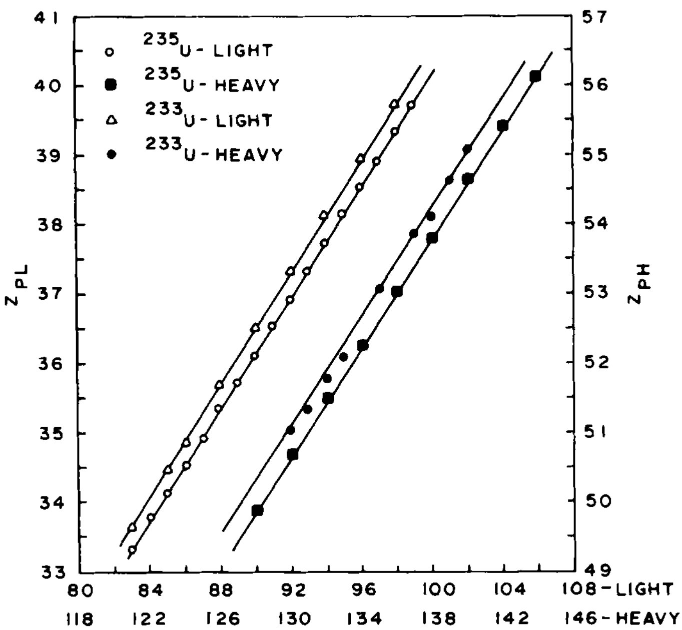


**Figure 4.** Most probable charge for light and heavy fission products.

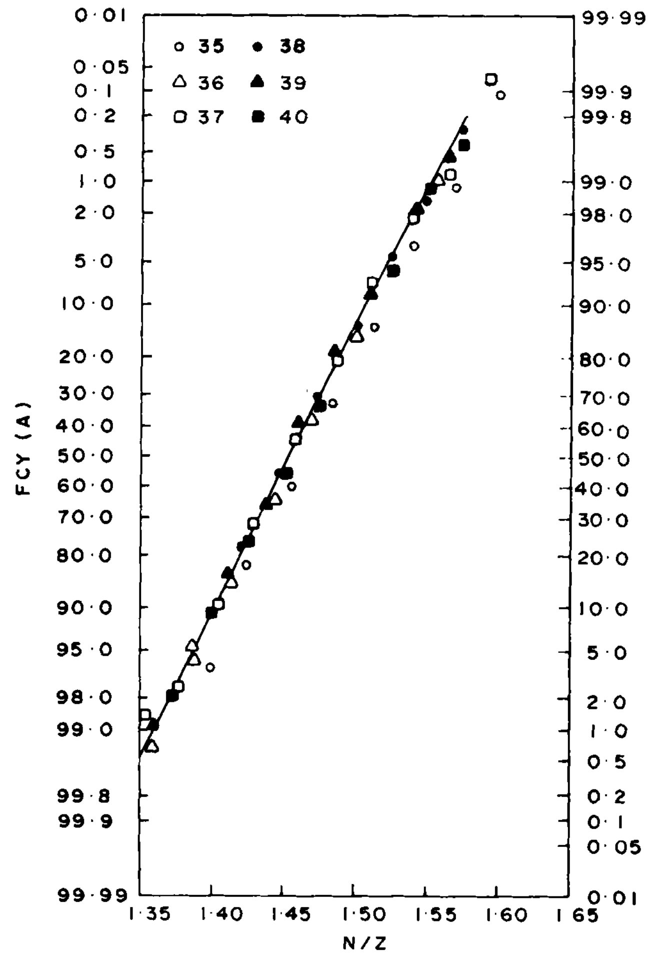


**Figure 5.** FCY($A$) vs ($N/Z$) of fission products from $^{234}$U fission.

obtained are given in table 1. These values were further used for the calculation of $A_p$, $Z_p$, and $\sigma_A$, $\sigma_Z$. Figure 6 shows the calculated values of $A_p$ and $Z_p$ along with the experimental values of Brissot *et al* (1977). The values are in good agreement with the experimental values for the fissioning nuclei $^{240}$Pu and $^{242}$Pu.

## 2.4 $^{252}Cf\,(SF)$

In the spontaneous fission of $^{252}$Cf the fractional independent yields of technetium isotopes (Srivastava *et al* 1984) and its complementary caesium have been determined recently. With data on only one isotopic chain in both heavy and light fission product it is difficult to assess the valadity of $(N/Z)_p$ and $\sigma_{N/Z}$ for charges other than 43 and 55. Since for $^{252}$Cf(SF) the systematics of charge distribution are similar to that of other fissioning nuclei like $^{234}$U, $^{236}$U, $^{240}$Pu and $^{242}$Pu, and the value of $(N/Z)_p$ and $\sigma_{N/Z}$, based on the technetium and caesium data, are similar to that in other systems the method can be extended for the other charges. With this assumption the values of $A_p$

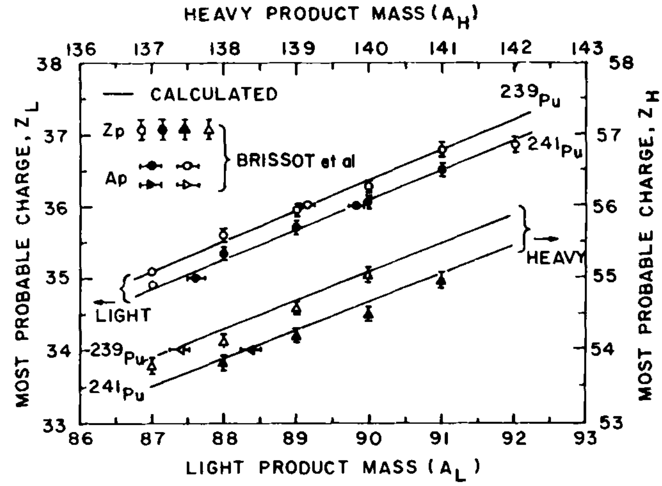


**Figure 6.** Calculated and experimental values of $Z_p$ and $A_p$ for thermal neutron induced fission of $^{239}Pu$ and $^{241}Pu$.

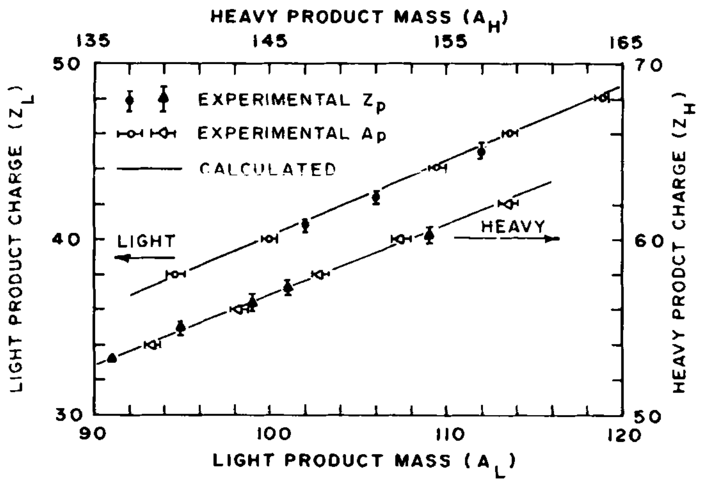


**Figure 7.** Calculated and experimental values of $Z_p$ and $A_p$ for spontaneous fission of $^{252}Cf$.

and $\sigma_A$ were calculated for other charges and are shown in figure 7 and 8 along with the experimental values of Reisdorf *et al* (1971) and Cheifetz *et al* (1971). Figures 7 and 8 also give the values of $Z_p$ and $\sigma_Z$ respectively for both the light and heavy fission products. The calculated values are in good agreement with that of the experimental values. The slight disagreement at some places of $A_p$ value is observed.

## 3. Discussion

To a good approximation over a large number of masses for light and heavy fission products, where the yields are high and the number of evaporated neutrons over the mass region does not change appreciably the centroid remains constant. For symmetric

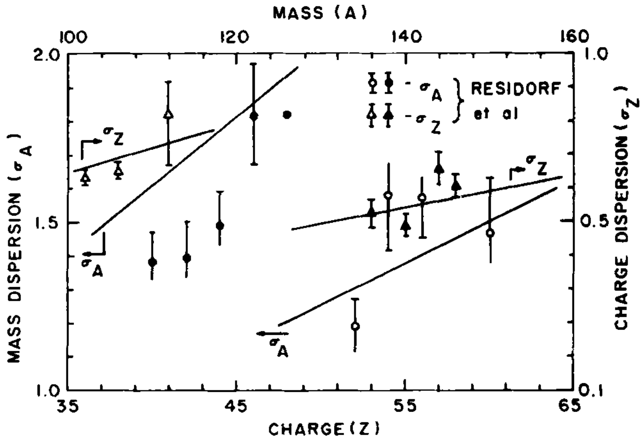


**Figure 8.** Plot of calculated values of $\sigma_A$ and $\sigma_Z$ along with the experimental data for $^{252}$Cf.

and highly asymmetric splits where no accurate independent yield data are available, it is not possible to check the validity of the present analysis. However the deviation in the values of $(N/Z)_p$ and $\sigma_{N/Z}$ may occur due to large neutron evaporation. It would have been better to carry out such an analysis using fragment distribution. But due to the lack of method for neutron correction for each fragment, we have confined the analysis to fission product. Quite a few experimental data are available on the effect of nucleon pairing on the independent yield of elements. The even $Z$ elements have higher yields compared to odd $Z$ elements. In the present analysis though the fine structure is ignored, but as shown in figure 9 $(N/Z)_p$ vs FIY$(Z)$ plot shows the yield value at $(N/Z)_p$ for even $Z$ is 22% higher compared to odd $Z$ in case of $^{236}$U. But the odd-even effects due to neutrons are not reflected in figure 1 as its magnitude is small.

Another feature which comes out from the present analysis is that the most probable $(A/Z)_p$ for light and heavy fission products agrees well with the average value of $(\bar{A}/\bar{Z})$ for light and heavy fission products as shown in table 2 for the fissioning nuclei $^{234}$U, $^{236}$U, $^{240}$Pu and $^{252}$Cf. As mentioned earlier the values of $A_p$, $Z_p$, $\sigma_A$ and $\sigma_Z$ can be obtained using (6)–(9). But a simplified form emerges if $(N/Z)_p$ is correlated with $(N/Z)_f$ of the fissioning system. The factor $\Delta_{N/Z}$ is defined as:

$$\Delta_{N/Z} = \{(N/Z)_p - (N/Z)_f\} \tag{12}$$

where $(N/Z)_f = (A - Z - \bar{\nu})/Z$ is the neutron-to-proton ratio of the fissioning system, $A$ and $Z$ are its mass and charge and $\bar{\nu}$ is the average number of neutrons emitted in the fission. Table 1 gives the values of $\Delta_{N/Z}$ for different fissioning systems. The value of $\Delta_{N/Z}$ for light and heavy fission products remain practically constant. Thus if the average value of $\Delta_{N/Z}$ is taken then

$$(N/Z)_p = \Delta_{N/Z} + (N/Z)_f. \tag{13}$$

Thus $(N/Z)_p$ is related to $(N/Z)_f$ through $\Delta_{N/Z}$ factor and is given by:

$$(N/Z)_{pL} = \{(N/Z)_f - (0{\cdot}05 \pm 0{\cdot}0019)\}, \tag{14}$$

$$(N/Z)_{pH} = \{(N/Z)_f + (0{\cdot}02 \pm 0{\cdot}0029)\}. \tag{15}$$

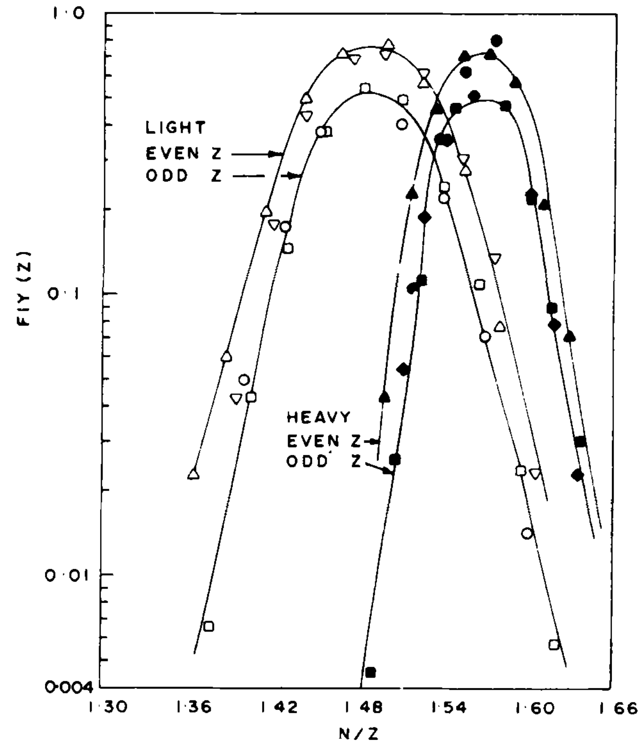


**Figure 9.** Proton odd-even effects in the thermal neutron-induced fission of $^{235}U$.

**Table 2.** Most probable and average value of $\bar{A}/\bar{Z}$ for light and heavy fission products in different fissioning systems.

| Nuclei | $\bar{A}_L$ | $\bar{Z}_H$ | $\bar{A}_L/\bar{Z}_H$ | $(A/Z)_p^L$ | $\bar{A}_H$ | $\bar{Z}_H$ | $(\bar{A}/\bar{Z})_H$ | $(A/Z)_p^H$ |
|---|---|---|---|---|---|---|---|---|
| $^{233}U$ | 99·30 | 37·71 | 2·474 | 2·465 | 138·20 | 54·29 | 2·540 | 2·540 |
| $^{235}U$ | 94·90 | 37·95 | 2·500 | 2·487 | 138·60 | 54·05 | 2·564 | 2·560 |
| $^{239}Pu$ | 98·90 | 39·90 | 2·478 | 2·475 | 138·10 | 54·10 | 2·552 | 2·542 |
| $^{252}Cf$ | 106·10 | 42·26 | 2·510 | 2·480 | 142·10 | 55·74 | 2·549 | 2·550 |

Similarly the average value of width of distribution is given by

$$\sigma_{N/Z}^{L} = 0{\cdot}040 \pm 0{\cdot}001, \tag{16}$$

$$\sigma_{N/Z}^{H} = 0{\cdot}023 \pm 0{\cdot}003, \tag{17}$$

where $L$ and $H$ refers to the light and heavy fission product respectively. For any fissioning system a set of values for $A_p$, $Z_p$, $\sigma_A$ and $\sigma_Z$ can thus be generated using the above mentioned prescription.

## 4. Conclusion

The present work based on an analysis of the fractional independent yield data on a number of fissioning systems shows that the liquid drop potential with isospin dependence plays a major role in nuclear charge distribution with shell effects having local perturbations. A simple universal prescription has been obtained which enables calculation of charge distribution parameters $A_p$, $Z_p$, $\sigma_A$ and $\sigma_Z$ for those fissioning nuclei which have not been studied exhaustively as well as for new fissioning nuclei. This simple approach would be useful in calculation of reactor parameters like decay heat and neutronics etc wherever the experimental data are not available.

## Acknowledgement

The authors are grateful to Dr P R Natarajan, Head, Radiochemistry Division for his keen interest in this work. One of the authors (AS) is thankful to the Department of Atomic Energy for a research fellowship.

## References

Brissot R, Crancon J, Ristori Ch, Bocquet J P and Moussa A 1977 *Nucl. Phys.* **A55** 109
Cheifetz E, Wilhelmy J B, Jared R C and Thompson S G 1971 *Phy. Rev.* C4 1913
Glendenin L E 1949 Technical Report no 35, Lab for Nucl. Sci., Mass Inst. Technol.
Mukherjee S 1969 *Nucl. Phys.* **A129** 297
Nifenecker H A, Blachot J, Bocquet J P, Mariolopouls G and Ristori J 1980 *Proc. Phys. and Chem. of Fission*, IAEA, Vol II, 35
Reisdorf W, Unik J P, Griffin H C and Glendenin L E 1971 *Nucl. Phys.* **A177** 337
Schmid M, Nir-El Y, Engler G and Amiel S 1981 *J. Inorg. Nucl. Chem.* **43** 867
Srivastava A, Nair A G C, Srivastava B K, Manohar S B, Prakash Satya and Ramaniah M V 1984 *Radio. Chim. Acta.* **35** 15
Wahl A C 1983 Fission yield compilation, Personal communication.
Wahl A C, Norris A E, Rouse R A and Williams J C 1969 *Proc. Sec. Int. Symp. Phys. and Chem. of Fission*, IAEA, Vienna 813
Way K and Wigner E P 1948 *Phys. Rev.* **73** 1318
Wehering B W, Lee S and Hart G 1980, UILU Eng. 80-5312, HIWATHA Technical Memorandum.
Wilkins B D, Steinberg E P and Chasman R R, 1976 *Phys. Rev.* **C14** 1832