Reasonable estimates for the other parameters indicate that the effect of the admixture of excited $s$ electron configurations could account for a major fraction of the discrepancy observed by Senitzky and Rabi. Formally, configurations in which a $4s$ electron is excited to any higher $s$ orbit contribute in the same way. Although it might be expected that the series would converge rapidly for high excitation, a systematic investigation of excited states would be desirable.

Somewhat analogous terms arise from the admixture of configurations $4snd5p$, and for $4s4d5p$ the excitation energy is not much larger than that for $4s5s5p$. The effect of these terms is small, however, for two reasons: the matrix elements of the electrostatic interaction are reduced because of angular interference, and the nondiagonal term in $H_z$ is small.

The relative importance of perturbations due to $s \rightarrow s'$ excitation, even when the excitation energy is large, suggests that the quasi-empirical quantitative calculations of Schwartz[6] for gallium may be extended to the $p$ states of the alkalies. It is possible, however, that $p$-shell excitation plays a perceptible role in the hyperfine structure of alkali spectra.

[6] C. Schwartz, Phys. Rev. **99**, 1035 (1955).

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# Mass Spectrometrically Determined Independent Yields of $I^{128}$, $I^{130}$, $Br^{80}$, and $Br^{82}$ for $U^{233}$, $U^{235}$, and $Pu^{239}$ Fission

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The primary yields of the shielded nuclei $I^{128}$ and $I^{130}$ have been accurately determined for the thermal neutron fission of $U^{233}$, $U^{235}$, and $Pu^{239}$. This has been done by the measurement of their stable daughters $Xe^{128}$ and $Xe^{130}$ using sensitive mass spectrometric techniques. The values of these primary yields show marked deviations from those predicted by the present theories of charge distribution. This discrepancy can be explained if the effect of the 50 proton shell is taken into account in the evaluation of the most probable initial nuclear charge $Z_p$ for mass numbers 128 and 130. This can be done by postulating a most probable charge which will yield the greatest energy release in the fission process.

Also the primary yields of $Br^{80}$ and $Br^{82}$ were determined by means of their daughters $Kr^{80}$ and $Kr^{82}$ for the thermal fission of $U^{233}$ and $U^{235}$ and for the fast fission of $Pu^{239}$. The yields of these nuclei for fast neutron fission were ~100 times those for thermal fission, indicating a shift in the charge distribution curve with neutron energy.

## INTRODUCTION

FOR a complete understanding of the fission process it is necessary to know both the charge and mass distribution of the fission fragments. Fission yield studies to date have been concerned mainly with mass distribution, the total or cumulative yield for a given mass being determined by radiochemical or mass spectrometric techniques. In this work the mass spectrometer measurements are less prone to contamination errors and are therefore the most reliable. The question of the distribution of charge for a given mass involves primary or independent yields of the fission fragments. In view of the short half-lives of most of these fragments, primary yield measurements are very difficult. Further, since all the primary yield data obtained to date[1–3] have been obtained radiochemically, the contamination errors are in most cases quite large. A number of accurate primary yield values have now been obtained using very sensitive mass spectrometric techniques. The results of these measurements are reported in this paper.

The fission process can be represented by the equation

$$M(A,Z)+M(1,0) \rightarrow M^*(A+1,Z) \rightarrow M(A_1,Z_1) + M(A_2,Z_2)+\nu M(1,0)+\gamma+Q,$$

where $M(A,Z)$ is the mass of the fissioning nucleus, $M(A_1Z_1)$ and $M(A_2Z_2)$ are the masses of the primary fission fragments, and $\nu$ the total number of prompt neutrons released. $\gamma$ is the electromagnetic energy released at the instant of fission and $Q$ is the kinetic energy of the fragments and the neutrons released. Since the neutron to proton ratio of the fissioning nucleus is considerably higher than that corresponding to stability in the fission product region, the primary fission products are unstable and achieve nuclear stability through a series of $\beta^-$ disintegrations. Therefore two types of fission yields can be defined:

(a) The total or cumulative yield $Y$ of a given mass chain, defined as the percentage of fission acts giving the mass number in question.

* Holder of a National Research Council scholarship 1955–1956.

[1] Glendenin, Coryell, and Edwards, *Radiochemical Studies: The Fission Products*, edited by C. D. Coryell and N. Sugarman, (McGraw-Hill Book Company, Inc., New York, 1951), Paper 52, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.

[2] L. E. Glendenin, Technical Report No. 35, Laboratory for Nuclear Science and Engineering, Massachusetts Institute of Technology, 1949 (unpublished).

[3] A. C. Pappas, Technical Report No. 63, Laboratory for Nuclear Science, Massachusetts Institute of Technology, 1953 (unpublished).

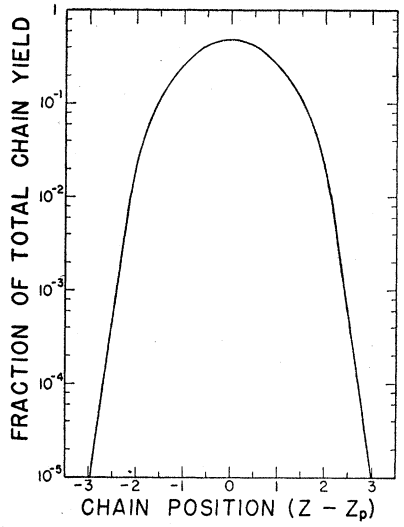


FIG. 1. The empirical charge distribution curve as determined by Glendenin.[2]

(b) The independent or primary yield $Y_i$ of a fission fragment, defined as the percentage of fission acts giving the nuclide in question by direct formation in the ground state or by formation in very short-lived excited states. Therefore the independent yields for a particular mass actually give the charge distribution for that chain.

The measurement of independent yields along a given mass chain is practically impossible with present techniques because of the very short half-lives and the correction necessary for decay and production rates. For the most part primary yield data are confined to a few so-called shielded isotopes, which are isotopes with stable or long-lived precursors. For example, in the mass chain

$$Sn^{130} \xrightarrow{\beta^-} Sb^{130} \xrightarrow{\beta^-} \text{stable } Te^{130}, \quad I^{130} \xrightarrow{\beta^-} \text{stable } Xe^{130},$$

the primary yields of tin, antimony, and tellurium accumulate at stable $Te^{130}$. Any yield of the shielded $I^{130}$ is therefore the independent or primary yield of this isotope. The yields of some of the shielded nuclides having stable or long-lived precursors have been determined radiochemically. However, the detection of a shielded nuclide in the presence of isotopic activities of much greater yield is only possible when its half-life or radiation characteristics, or both, are sufficiently different from those of the higher yield isotopes to permit identification by decay and absorption measurements. A method which avoids these difficulties is to measure the $Xe^{130}$ yield by means of a high-sensitivity mass spectrometer. This paper is concerned with such a method and the evaluation of several independent yields.

The results obtained by Glendenin, Coryell, and Edwards,[1] Glendenin,[2] and later by Pappas[3] have been sufficient to determine a semiempirical charge distribution which is assumed independent of the mass chain, the fissioning nucleus and the neutron energy. The theory of equal charge displacement postulated by Glendenin[1,2] assumes that the most probable charges for both fragments are equally displaced from the stability line. The probability curve is of the Gaussian type, centered around $Z_p$, the most probable charge for the mass chain considered, and is shown in Fig. 1. The value of $Z_p$ is given by

$$Z_p = Z_A - \tfrac{1}{2}\{Z_A + Z_{(A'-A-\nu+1)} - Z'\}, \qquad (1)$$

where $Z_A$ is the most stable charge for mass number $A$ which is found from the Bohr-Wheeler stability line.[4] $A'$ and $Z'$ are the mass number and charge of the fissioning nucleus and $\nu$ is the average number of neutrons emitted per fission. Therefore the most stable charge for each fragment satisfies the equation

$$(Z_p - Z_A)_{\text{light}} = (Z_p - Z_A)_{\text{heavy}}. \qquad (2)$$

Pappas found marked deviations from Glendenin's distribution curve. However, the discrepancy was practically eliminated when he calculated the $Z_A$ values from the lines of maximum stability given by Coryell, Brightsten, and Pappas.[5] These lines have discontinuities at nuclear shell boundaries. He defined $Z_p$ by

$$Z_p = Z_A - \tfrac{1}{2}\{(Z_A - Z_{(A'-A+1)} - Z'\}, \qquad (3)$$

so that $Z_p$ corresponds to a mass chain $(A-1)$ because the prompt-neutron emission is considered to occur from the fragments themselves. In most cases $Z_p$ is nonintegral.

## EXPERIMENTAL

Samples of $U^{233}$, $U^{235}$, and $Pu^{239}$ in the form of metals, aluminum alloys, and oxides were irradiated for various lengths of time at differing fluxes in the N.R.X. reactor until about $10^{17}$ fissions had occurred. The rare gases produced in fission were extracted by dissolving the oxide in nitric acid, dissolving the metal in cupric potassium chloride and by melting the aluminum alloys. The wet chemical extraction required the use of a drying train and all methods of extraction included the use of a calcium furnace for the final purification.

The rare gases were analyzed with a 10-in. 90° direction-focusing mass spectrometer using an electron multiplier for the detector. The gain of the multiplier was $3\times10^4$ so that the over-all sensitivity of the instrument was several orders of magnitude greater than if ordinary single ion collection had been employed. The sensitivity of the instrument is sufficient to permit a complete analysis of xenon isotopes present to a volume of $1.5\times10^{-6}$ cu mm at N.T.P. The relative abundances of the isotopes of xenon and krypton in normal samples were measured and found to agree within $\frac{1}{2}\%$ of the accepted values given by Nier.[6] The isotope ratios

[4] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).
[5] Coryell, Brighsten, and Pappas, Phys. Rev. **85**, 732 (1952).
[6] A. O. Nier, Phys. Rev. **79**, 450 (1950).

obtained from fission product xenon and krypton were therefore assumed to be absolute within this percentage. The output from the electron multiplier was integrated and amplified by a vibrating reed electrometer and a permanent record was obtained.

The sensitivity of the instrument is limited only by the residual hydrocarbon background which was reduced but not eliminated by baking the instrument at 350°C. However, the large gain of the multiplier made it possible to increase the resolving power[7] of the instrument until the isotopes of xenon and krypton were resolved from the hydrocarbons at the corresponding masses. A resolution of 650 was found to be sufficient to separate hydrocarbons from pure elements of the same mass number in the mass region 1 to 200. This separation is clearly shown in Fig. 2.

## RESULTS

Figure 2 gives a typical mass spectrogram obtained for a sample of fission product xenon extracted from irradiated plutonium. These spectrograms showed for the first time the presence of $Xe^{128}$ and $Xe^{130}$ as fission products. The $Xe^{131}$, $Xe^{132}$, $Xe^{134}$, and $Xe^{136}$ peaks represent the cumulative yields of their respective mass chains. The $Xe^{128}$ and $Xe^{130}$ represent the primary yields of $I^{128}$ and $I^{130}$, respectively, as well as a small contribution from the primary yields of the two xenon isotopes themselves ($\sim 10^{-3}$ of the peak height). The presence of $Xe^{129}$ (25.6% abundant in nature) is principally normal contamination since the 129 mass chain is blocked at $1.3\times10^{7}$ year $I^{129}$ and since the primary yield of $Xe^{129}$ itself (about $5\times10^{-5}$ of the 129 chain) is very small. The isotope pattern will be very sensitive to normal xenon contamination, particularly at mass 129. Any appreciable 129 is assumed to be normal contamination and a small correction must be made to the

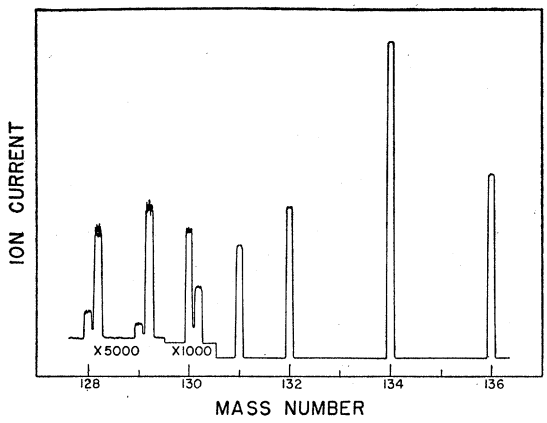


FIG. 2. Mass spectrogram of the xenon pattern for the thermal-neutron fission of $Pu^{229}$. The xenon isotopes fall at the position where the mass is marked while the hydrocarbon having the corresponding mass is displaced towards higher masses.

[7] The resolving power is defined as the distance between the centers of two adjacent peaks times the average mass number divided by the width of the peak at the base.

TABLE I. Primary yields of $I^{128}$ and $I^{130}$.

| Fissioning nuclide | Fission yield % $I^{128}$ | $I^{130}$ | $I^{128}/Y^{128}$ [a] | $I^{130}/Y^{130}$ |
|---|---|---|---|---|
| $U^{233}$ | $1.3\times10^{-4}$[b] | $3.9\times10^{-3}$[b] | $(1.1\pm0.1)\times10^{-3}$ | $(3.8\pm0.3)\times10^{-3}$ |
| $U^{235}$ | $3.6\times10^{-5}$[c] | $5.0\times10^{-4}$[c] | $(9.8\pm1.0)\times10^{-5}$ | $(2.8\pm0.2)\times10^{-4}$ |
| $Pu^{239}$ | $2.0\times10^{-4}$[d] | $5.2\times10^{-3}$[d] | $(2.4\pm0.2)\times10^{-4}$ | $(2.0\pm0.1)\times10^{-3}$ |

[a] $I^{128}/Y^{128}$ is the yield of $I^{128}$ relative to the total yield of the mass 128 chain ($Y^{128}$).
[b] Measured relative to $Xe^{131}=3.74\%$.
[c] Measured relative to $Xe^{131}=2.93\%$.
[d] Measured relative to $Xe^{131}=3.6\%$.

128 and 130 yields since both $Xe^{128}$ and $Xe^{130}$ appear in nature.

Consideration of the neutron capture contribution must also be made since the reactions $I^{127}(n,\gamma)I^{128}$ and $I^{129}(n,\gamma)I^{130}$ are both possible. The determined $Xe^{128}/Xe^{131}$ and $Xe^{130}/Xe^{131}$ ratios are used to evaluate the fraction of the 128 and 130 chains appearing as iodine primary fission fragments. From these considerations the following relation can be derived:

(measured yield of $Xe^{130}$) = (primary yield of $I^{130}$) + (fractional capture of 129 chain),

or mathematically

$$\alpha Y^{130}+\left[1-\frac{(1-e^{-\sigma\varphi t})}{\sigma\varphi t}\right]Y^{129}=\frac{Xe^{130}Y^{131}}{Xe^{131}}, \qquad (4)$$

where $\alpha$ is the fraction of the 130 chain formed as $I^{130}$, $\varphi$ the flux, $t$ the irradiation time and $\sigma$ the neutron capture cross section of $I^{129}$. $Y^{A}$ represents the total chain yield for mass number $A$, and was obtained from the smooth radiochemical mass-yield curve.[8] An analogous calculation gives the primary yield of $I^{128}$. The iodine yields so obtained are listed in Table I.

An investigation in the krypton region revealed the presence of $Kr^{80}$ and $Kr^{82}$ indicating for the first time that $Br^{80}$, $Br^{82}$, $Kr^{80}$, and $Kr^{82}$ are all fission products. The analyses of these shielded nuclides are difficult since krypton yields are generally smaller than zenon and normal contamination errors are greater since krypton is 17 times more abundant in nature. The sample has to be essentially void of adsorbed gases since 1 cu mm of air at N.T.P. contains as much $Kr^{80}$ as is produced in $10^{17}$ fissions. Because of these difficulties only preliminary results have been obtained which are given in Table II.

A sample of $Pu^{239}$ was irradiated with fission spectrum neutrons and preliminary measurements were made of the shielded nuclides. The value for the $I^{130}$ yield was of the same order of magnitude as for the thermal neutron fission of $Pu^{239}$. However, the $Br^{80}$ and $Br^{82}$ yields increased several orders of magnitude giving primary yields of about $10^{-1}$ of the total chain.

[8] *Radiochemical Studies: The Fission Products*, edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, Inc., New York, 1951), Appendix B, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.

TABLE II. Primary yields of $Br^{80}$ and $Br^{82}$.

| Fissioning nuclide | $Br^{80}/Y^{80}$ [a] | $Br^{82}/Y^{82}$ |
|---|---|---|
| $U^{233}$ | $<1\times10^{-3}$ | $(2.5\pm0.8)\times10^{-3}$ |
| $U^{235}$ | $\sim1\times10^{-4}$ | $\sim6\times10^{-4}$ |

[a] $Br^{80}/Y^{80}$ is the yield of $Br^{80}$ relative to the total yield of the mass-80 chain $Y^{80}$.

## DISCUSSION OF RESULTS

The accurate primary yields obtained for $I^{128}$ and $I^{130}$ for $U^{233}$, $U^{235}$, and $Pu^{239}$ fission provide a severe test for the theories of charge distribution. Both are formed as the same element and therefore the value of $Z$ is constant. Since the most probable charge, $Z_p$, is in the region of the 50 proton shell, a discrepancy between theory and experiment might be expected here. Table III gives the most probable charge for mass chains 128 and 130 for various fissioning nuclides using both Glendenin's and Pappas' calculations. The value $(Z-Z_p)=\Delta Z$ for both the 128 and 130 chains is $(53-Z_p)$. Figure 3 shows the determined yields relative to the smooth curves predicted by Glendenin and Pappas. It is immediately apparent that the yields do not agree with the predicted curves and also that a smooth curve cannot be drawn through all the points. In all cases the calculated value $\Delta Z$ is too large and there is a displacement according to mass. Pappas' curve agrees more closely with the determined yields than does that of Glendenin, particularly for the 130 case. However, in both curves the 128 yields are displaced considerably. In order to account for this discrepancy, it is necessary to consider the theory of equal chain lengths in more detail.

The assumptions made by Glendenin were:

(a) the curve shown in Fig. 1 is independent of the mass and charge of the fissioning nuclide;

(b) the curve is independent of the mass of the fission fragment;

(c) the value of $Z_p$ is only dependent on $Z_A$ and is a linear function of $Z_A$.

Since the $I^{130}$ yields all lie on the curve, assumption (a) appears valid within the accuracy of the experiment. Radiochemical determinations indicate that (b) is also a good assumption. From this it can be inferred that the displacement of the 128 yields is due principally to the method used to calculate $Z_p$. It has been noted that $Z_A$ has been corrected for shell effects when the mass chain $A$ is 87, 95, 120, or 146. However, no such correction is made in the evaluation of $Z_p$ when it is in the vicinity of a closed shell. It might be expected for example that $Z_p$ would remain close to 50 over several mass numbers in the neighborhood of the 50-proton shell. Such an effect could account for the displacement of the $I^{128}$ yields in Fig. 3. We propose therefore that shell effects be taken into account in the determination of $Z_p$ for a given $A$. This can be done by postulating a $Z_p$ which will yield the greatest energy release in the fission act. For example, in the fission of $U^{235}$ the value of $Z_p$ is then defined in such a way that

$$E=M(236,92)-3M(1,0)-M(A,Z_p)-M(233-A,92-Z_p) \quad (5)$$

is maximum.

For Eq. (5) to be maximized, knowledge of masses far removed from the stability line are required. Since the energetics in that region have not been experimentally investigated, it is necessary to make extrapolations by means of a mass formula which will include shell effects. The mass formula most applicable to date is that of Kumar and Preston[9] in which shell effects and spin terms have been included. Their formula is most useful for isobaric sequences and hence it is readily applicable to the evaluation of Eq. (5). To obtain the maximum energy release, Eq. (5) is solved for various values of $Z$ and a curve is plotted of $Z$ against energy release. The value of $A$ remains constant so that a parabolic relation between $Z$ and the released energy is obtained which shows distortion when $Z$ is in the region of a closed shell. The value of $Z_p$ is determined graphically. Preliminary calculations of $Z_p$ have been

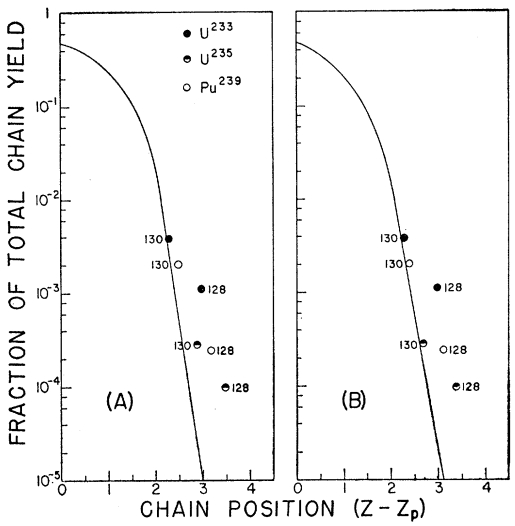


FIG. 3. Comparison is made between the results obtained by the authors and the empirical results of (*A*) Glendenin[2] and (*B*) Pappas.[3]

TABLE III. Calculated most probable charge for masses 128 and 130.[a]

| Nuclide | Glendenin 128 | Glendenin 130 | Pappas 128 | Pappas 130 |
|---|---|---|---|---|
| $U^{233}$ | 50.0 | 50.7 | 50.0 | 50.7 |
| $U^{235}$ | 49.5 | 50.1 | 49.6 | 50.3 |
| $Pu^{239}$ | 49.8 | 50.5 | 49.9 | 50.6 |

[a] The error is ±0.1 charge unit in all cases.

[9] K. Kumar and M. A. Preston, Can. J. Phys. **33**, 298 (1955).

made over the mass range $A=126$ to $A=136$ for $U^{233}$, $U^{235}$, and $Pu^{239}$ for the case where the number of prompt neutrons is 3 and the results so calculated are plotted in Fig. 4.

From Fig. 4 the effect of the 50-proton shell and the 82-neutron shell are clearly indicated. It is seen that the value of $Z_p$ remains very nearly 50 from $A=128$ to $A=132$ which is quite different from the behavior of $Z_p$ determined from the theory of equal chain lengths shown as $Z_p'$ in Fig. 4. For the $U^{235}$ case, for example, the value of $Z_p$ calculated from energetic considerations is 0.4 charge unit higher at mass 128 than that calculated from the theory of equal chain lengths. This indicates that the whole charge distribution curve at mass 128 is displaced by 0.4 unit due to the influence of the 50 proton shell. Actually, it is seen in Fig. 3 that the measured primary yields for the 128 chain are displaced by $\sim$0.5 charge unit in the same direction. Therefore, the primary yield results do support the postulate of maximum energy release in the theory of nuclear charge distribution. Also, the values of $Z_p$ calculated by the method of maximum energy release suggests that the yields of the 131 and 132 chains will be low or lie to the left of the predicted curve. This has been confirmed by the results of Wahl[10] who found that the primary yields of $Te^{131}$ and $I^{132}$ were low. In nonshell regions both methods of determining $Z_p$ give essentially the same value.

Also, it might be pointed out that the mass yield curves for $U^{233}$, $U^{235}$, and $Pu^{239}$ show no appreciable fine structure in the mass range 128 to 132 so that cross chain branching due to favored prompt neutron emission seems unlikely in this region. The preliminary calculations of $Z_p$ by the method of maximum energy release indicate that in certain cases the charge distribution depends on $A$. Calculations of $Z_p$ for the complete fission range are being made and the values obtained will be correlated with all the primary yield data available. Although this method of evaluating $Z_p$ may give a good description of charge distribution, a more complete understanding of the fission process we believe, may be obtained from a careful study of Fong's[11,12] statistical model of fission.

[10] A. G. Wahl, Phys. Rev. **99**, 730 (1955).
[11] P. Fong, Phys. Rev. **89**, 332 (1953).
[12] P. Fong, Phys. Rev. **102**, 434 (1955).

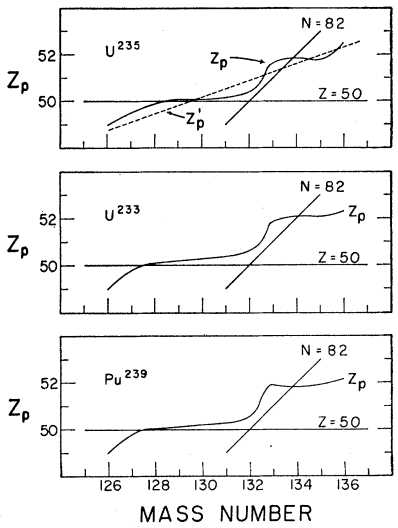


FIG. 4. Curves showing the relation between $Z_p$ and $A$ for various fissioning nuclides when $Z_p$ is calculated as the charge corresponding to the maximum energy release. The dashed curve shown for $U^{235}$ is the relation between $Z_p$ and $A$ when $Z_p$ is calculated by the theory of equal chain lengths. In all cases three prompt neutrons are assumed to be released.

The extremely high primary yields of $Br^{80}$ and $Br^{82}$ obtained for the fast fission of $Pu^{239}$ as compared to fission by thermal neutrons indicates a marked shifting of the charge distribution curve with neutron energy. This shifting of the charge distribution curve was also observed by Wahl[10] in his studies of the fission of $U^{235}$ with 14-Mev neutrons and can be attributed to the emission of extra neutrons at the higher energies. This means that in the high-energy fission of plutonium the yields obtained correspond to the fission of the nuclide $(Pu^{240-x})^*$, where $x$ is the number of extra neutrons emitted at the higher energies.

Irradiations are being carried out to obtain primary yield data for $Br^{80}$ and $Br^{82}$ for both thermal and fission spectrum neutrons and $I^{128}$ and $I^{130}$ yields for fission spectrum neutrons.

## ACKNOWLEDGMENTS

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