Charge Distribution in the Fission of Np^{237} and Pu^{239} with Intermediate-Energy Helium Ions*

N. A. WOGMAN,† J. A. POWERS,‡ AND J. W. COBBLE
Department of Chemistry, Purdue University, Lafayette, Indiana
(Received 25 May 1966)

The independent yields of ten fission products from the fission of Np^{237} induced by 40.5-MeV helium ions and from the fission of Pu^{239} induced by 32.0-MeV helium ions have been measured radio chemically. The data can be interpreted in terms of the equal-charge-displacement (E.C.D.) hypothesis, and less rigorously, by the constant-charge-ratio proposal. The most applicable mass equation for the determination of the E.C.D. parameters was found to be a continuous non-shell-corrected function. Information on neutron yield as a function of fission-product mass has been inferred from the charge division. There is no evidence from this research for a 50-proton shell effect on yields or for an isomeric state of Nb^{96}.

INTRODUCTION

PROPOSALS[1–10] regarding the distribution of charge in fission have been available since 1947. At the present time there are three recipes in general use: (a) equal charge displacement (E.C.D.); (b) unchanged charge distribution or constant charge ratio (C.C.R.); and (c) minimum nuclear potential energy (M.N.P.E.). In spite of the fact that some of these proposals are now almost 20 years old, there does not seem to be any agreement as to just which hypothesis is generally appreciable to the fission process. Almost all investigators appear to agree that the independent- and cumulative-yield data from the thermal neutron fission of U^{235} [2,3] are best correlated by some type of E.C.D. treatment. Recently, Wahl and his co-workers[11] have been able to fit an impressive amount of such data to a "universal" Gaussian curve of fractional isobaric yield as a function of the most probable charge Z_p. Other low-energy fission investigations of various elements[12–19] also favor an E.C.D. treatment. Simple logic argues that in very high energy fission (where there is insufficient time available for the fragments to be in energy equilibrium) the ratio of neutrons to protons in the *primary* fission fragment will be similar to the parent nuclide.[5] How these primary fragments will be related to the *secondary* fission fragments (after neutron evaporation) is a further problem about which little is known.

In the intermediate energy range ($\sim$10–100 MeV), the evidence and interpretations do not always agree. Pate, Foster, and Yaffe[20] preferred E.C.D. in the proton fission of Th^{232} (8–90 MeV); other investigators[21–28] found that C.C.R. or M.N.P.E. postulates correlated their medium- and high-energy fission measurements on various elements. The helium-ion-induced fission studies of uranium isotopes carried out in these laboratories resulted in admittedly limited independent-yield data which could be correlated to the C.C.R. rule.[22,27,28] Data[29,30] on the helium-ion-induced fission of Th^{232} were correlated by both M.N.P.E. and C.C.R. rules.

Part of the reason for the present lack of agreement appears to lie in the difficulties associated with the radiochemical determinations. It is not that the individual fission-product cross sections are so difficult to fix, but rather that the total isobaric fission cross sections can rarely be determined directly over the whole

* This work was supported by the U. S. Atomic Energy Commission.
† From the Ph.D. thesis of N. A. Wogman, Purdue University, January, 1966. Present address: Battelle Northwest Laboratory, Richland, Washington.
‡ From the Ph.D. thesis of J. A. Powers, Purdue University, January, 1962. Present address: U. S. Atomic Energy Commission, Washington, D. C.
1 K. Way and E. P. Wigner, Phys. Rev. **73**, 1318 (1948).
2 L. E. Glendenin, C. D. Coryell, and R. R. Edwards, in *The Fission Products*, edited by C. D. Coryell and N. Sugarman, National Nuclear Energy Series (McGraw-Hill Publishers, Inc., New York, 1951), Div. 4, 9, Paper 52.
3 A. C. Pappas, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy*, 1955 (United Nations, New York, 1956), Vol. 7, p. 19.
4 A. C. Wahl, J. Inorg. Nucl. Chem. **6**, 263 (1958).
5 R. H. Goeckerman and I. Perlman, Phys. Rev. **76**, 628 (1949).
6 V. F. Apalin *et al.*, Atomnaya Energ. **8**, 15 (1960).
7 D. C. Brunton, Phys. Rev. **76**, 1798 (1949).
8 P. Fong, Phys. Rev. **102**, 434 (1956).
9 R. D. Present, Phys. Rev. **72**, 7 (1947).
10 W. Swiatecki, results cited by Blann in University of California Radiation Laboratory Report No. UCRL-9190, 1960 (unpublished).
11 A. C. Wahl *et al.*, Phys. Rev. **126**, 1112 (1962).
12 Herbert V. Weiss, Phys. Rev. **139**, B304 (1965).
13 B. L. Cohen, and C. B. Fulmer, Nucl. Phys. **6**, 547 (1958).
14 Kurt Wolfsberg, Phys. Rev. **137**, B929 (1965).
15 D. A. Marsden and L. Yaffe, Can. J. Chem., **43**, 249 (1965).
16 L. E. Glendenin, J. P. Unik, and H. C. Griffin, U. S. Atomic Energy Commission Report No. SM-60 1/31 (unpublished).
17 J. G. Cunninghame *et al.*, Nucl. Phys. **44**, 588 (1963).
18 A. Apallonova *et al.*, Radiochemistry (USSR) 4, 631 (1962).
19 K. M. Broom, Phys. Rev. **133**, B874 (1964).
20 B. D. Pate *et al.*, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 15, p. 149.
21 James A. McHugh, University of California Radiation Laboratory Report No. UCRL-10673 (unpublished).
22 L. J. Colby, Jr., and J. W. Cobble, Phys. Rev. **121**, 1410 (1961).
23 H. Marshall Blann, Phys. Rev. **123**, 1356 (1961).
24 Gösta Rudstam and A. C. Pappas, Nucl. Phys. **22**, 468 (1961).
25 G. Friedlander *et al.*, Phys. Rev. **129**, 1809 (1963).
26 A. C. Wahl, Phys. Rev. **99**, 730 (1955).
27 R. Gunnink and J. W. Cobble, Phys. Rev. **115**, 1247 (1959).
28 L. J. Colby, Jr., Mary LaSalle Shoaf, and J. W. Cobble, Phys. Rev. **121**, 1415 (1961).
29 M. E. Davis, Ph.D. thesis, Purdue University, 1963 (unpublished).
30 J. A. Powers, Ph.D. thesis, Purdue University, 1962 (unpublished).

fission-product mass range. In order to construct a completely unambiguous mass-yield curve representing all of the isobaric chains, only those isotopes which represent >90% of the isobaric yield by *any* hypothesis are of any use. Since the slope of the mass-yield curve, $d\sigma/dA$, is changing rapidly on the outside of the "wings," these regions are particularly difficult to define. Even the instrumental methods of mass determination, such as time-of-flight or solid-state measurements, are of limited use in fixing the mass-distribution curve since such methods have typical dispersions of from 2.5[31] to 4.5 mass units.[32] However, it is at the mass regions (A=65–85; 145–165) where the three proposed charge distributions clearly give different predictions.

It is entirely possible that the complexity of the fission process precludes the existence of any single simple charge-distribution description applicable to all fission-product mass regions at all excitation energies. However, more data are still required to test this point.

The general situation for the fission of lower Z elements is still uncertain. The most recent and accurate results for gold suggest that a C.C.R. treatment is satisfactory, but the data are quite limited.[33]

The object of this communication is to report more extensive data covering a wide range of fission-product mass regions for the heavy-element compound nuclei Am^{241*} and Cm^{243*}. By varying the different parameters in the E.C.D. and M.N.P.E. treatments, over fifteen different modifications have been tested in the intermediate energy range (25–35 MeV of excitation energy).

EXPERIMENTAL PROCEDURE

Thin actinide oxide targets (about 0.2 mg/cm^2) were prepared by electrodeposition[34] on high-purity, radiochemically analyzed aluminum foil. The thickness of neptunium-237 and plutonium-239 oxides was determined by alpha assay of the known target area using a 2π windowless proportional flow counter and a low geometry scintillation counter. The uniformity of the deposits was $\geq$98% as determined by a comparison of the alpha activity from various sectors of the target. Radiochemical analysis of the aluminum foils indicated negligible ($\leq$0.6 parts per million) quantities of As and Cs and approximately 20 parts per billion of fissionable heavy elements (presumably U or Th).

Stacked foil-type target assemblies were irradiated at the external beam facilities of the Argonne National Laboratory 60-in. cyclotron. Range-energy relationships based on the work of Bichsel *et al.*,[35] were used to determine the energies of the incident helium ions. The actinide oxide backing foils of aluminum served to collect the recoil fission fragments and were dissolved along with the heavy element oxides.

Standard radiochemical procedures[29,30,34] common to heavy element fission were used in this investigation. In general as many isotopes as possible were removed from each of the irradiated targets, although a number of replicate runs were also made. The identity and purity of each fission product were established by measurements of half-life and specific activity, in some cases by parent-daughter isotope "milking" procedures, and, when necessary, by analysis of the gamma spectra.

Measurements of radioactivity were carried out using conventional thin window proportional or Geiger counters and low-background (about 0.15 counts/min) anticoincidence shielded Geiger counters.[36] Many of the isotopes involved were standardized by 4π beta techniques[37] similar to those used in our previous heavy-element studies. From such data, accurate corrections for backscattering, self-absorption, forescattering, and effective geometry were generated for a few others (Cs^{136}, Pr^{142}, and La^{140}). Observed counting rates of complex decay curves were analyzed by a modified,[38] least squares, 7094, [PAKAG],[39] computer program. The resulting counting rates were converted to isotopic cross sections by applying the appropriate factors for half-life, counting efficiency, chemical yield, decay during bombardment, and the total helium-ion beam current.

EXPERIMENTAL DATA

The experimental data are summarized in Table I. Errors indicated include either the standard deviations for replicate bombardments, or reasonable estimates from the nature of the isotope involved (indicated parenthetically). The total isobaric cross sections σ_F were those reported in the following paper.[40] They were either directly measured or interpolated from mass-yield curves constructed using isotopic cross sections that had only small corrections due to charge distribution (see Discussion).

DISCUSSION

The three main charge-distribution postulates, M.N.P.E., C.C.R., and E.C.D. predict different values of Z_p in many of the mass regions under investigation. Briefly, the assumptions and requirements of all of these postulates are: (1) The distribution of charge is symmetric about a most probable charge, Z_p; (2) the dis-

[31] J. S. Frazer, J. C. D., Milton, H. R. Bowman, and S. G. Thompson, Can. J. Phys. **41**, 2080 (1963).
[32] J. P. Unik and J. R. Huizenga, Phys. Rev. **134**, B90 (1964).
[33] F. L. Lisman, H. W. Brandhorst, Jr., and J. W. Cobble, Phys. Rev., **140**, B863 (1965).
[34] N. A. Wogman, Ph.D. thesis, Purdue University, 1966 (unpublished).
[35] H. Bichsel, R. F. Mosley and W. A. Aron, Phys. Rev. **105** 1788 (1957).
[36] H. A. André, Ph.D. thesis, Purdue University, 1964 (unpublished).
[37] R. Gunnink, L. J. Colby, Jr., and J. W. Cobble, Anal. Chem. **31**, 796 (1959).
[38] C. Menninga, Ph.D. thesis, Purdue University, 1966 (unpublished).
[39] Los Alamos PAKAG Program, Los Alamos Scientific Laboratory Reports Nos. LA-2367, LA-2367 Addenda, 1963 (unpublished).
[40] J. A. Powers, N. A. Wogman, and J. W. Cobble, following paper, Phys. Rev., **152**, 1096 (1966).

TABLE I. Cross sections for helium-ion-induced fission of Np^{237} and Pu^{239}.

Isotope	Mass	Independent yield cross section σ_i (mb)	Total isobaric cross section[40] σ_F (mb)	Fractional yield, f_i
		Energy, 40.5 MeV; Np^{237}		
As^{76}	76	0.14 ±0.08	1.80	0.080
Br^{82}	82	0.285±0.015	6.60	0.043
Rb^{86}	86	0.76 ±0.10	13.0	0.059
Nb^{96}	96	1.57 ±0.16	37.5	0.042
Zr^{97a}	97	36.6 ±1.8	39.0	0.94
Ag^{112}	112	7.6 ±2.5	43.0	0.18
Pd^{112a}	112	40.5 (±5)	43.0	0.94
I^{130}	130	15.6 ±2.6	47.5	0.33
I^{131a}	131	33.9 ±1.7	48.0	0.71
I^{133a}	133	20.4 ±1.0	47.0	0.44
Cs^{136}	136	20.4 ±1.6	43.0	0.474
Ba^{140a}	140	16.0 ±0.8	32.5	0.50
La^{140}	140	9.33 ±0.50	32.5	0.287
Pr^{142}	142	1.17 ±0.42	27.5	0.043
Tb^{160}	160	0.370±0.077	1.26	0.294
		Energy, 32.0 MeV; Pu^{239}		
As^{76}	76	0.021±0.018	0.84	0.025
Br^{82}	82	0.46 ±0.18	4.00	0.114
Rb^{86}	86	0.58 ±0.37	7.60	0.076
Nb^{96}	96	1.55 ±0.25	23.8	0.065
Zr^{97a}	97	25.4 ±1.0	36.0	0.71
Ag^{112}	112	3.26 ±0.82	25.4	0.13
Pd^{112a}	112	23 ±4	25.4	0.91
I^{130}	130	11.9 ±1.1	35.0	0.322
I^{131a}	131	26 ±8	37.0	0.70
I^{133a}	133	19.2 (±0.8)	42.0	0.46
Cs^{136}	136	20 ±2	40.0	0.50
Ba^{140a}	140	16.3 ±1.4	27.8	0.58
La^{140}	140	9.99 ±2.9	27.8	0.36
Pr^{142}	142	0.33 ±0.10	23.3	0.014
Tb^{160}	160	0.47 ±0.18	1.40	0.33

[a] Cumulative yields (Ref. 40).

tribution of charge is independent of the number of protons or neutrons of the fissioning nucleus; (3) the relative charge distribution (Z_p versus fractional chain yield) has the same shape for all mass numbers; (4) the average number of neutrons emitted per fission event must be known; (5) the number of prefission neutrons must be known; (6) a neutron function describing the post-fission neutron distribution as a function of fragment mass must be available; and (7) a method is required to fix Z_A for use with the E.C.D. postulate.

The E.C.D. hypothesis proposes that the fission fragment and its complementary product be an equal number of isobaric units away from the line of nuclear stability.

$$Z_{A(L)}-Z_{p(L)}=Z_{A(H)}-Z_{p(H)}\,, \tag{1}$$

where $Z_{A(L)}$ and $Z_{A(H)}$ are the most stable charges of the complementary fission product chains, and $Z_{p(L)}$ and $Z_{p(H)}$ are the most probable charges for the light and heavy fission fragments, respectively. The sum of the charges of the fragments must equal that of the compound nucleus:

$$Z_{p(L)}+Z_{p(H)}=Z_C\,. \tag{2}$$

Combination of Eqs. (1) and (2) yields an expression for the most probable charge of a fission product of mass $A_{(H)}$ at scission:

$$Z_{p(H)}=[Z_{A(H)}-Z_{A(L)}+Z_C]/2.0\,. \tag{3}$$

The second hypothesis, C.C.R., proposes that the compound nucleus fissions so rapidly that the fragments would have both the same neutron-to-proton ratio.

$$Z_{p(L)}/N_{(L)}=Z_{p(H)}/N_{(H)}\,, \tag{4}$$

where $N_{(L)}$ and $N_{(H)}$ are the number of neutrons in the light and heavy fragments, respectively.

$$N_{(L)}=A_{(L)}-Z_{p(L)} \tag{5}$$

$$N_{(H)}=A_{(H)}-Z_{p(H)}\,. \tag{6}$$

Combining Eqs. (2), (4), (5), and (6) yields the expression:

$$Z_{p(H)}=Z_C(A_{(H)})/A_C \tag{7}$$

where A_C is the mass of the compound nucleus at scission.

The third major postulate proposes a distribution of nuclear charge such that a minimum is obtained for the sum of nuclear potential energy and Coulombic repulsion energy.

The potential energy of spherical pair of fragment masses $M_{(H)}$, $M_{(L)}$, with charges $Z_{(H)}$, $Z_{(L)}$, separated by a distance D can be written as[9,10]:

$$\text{P.E.}=M_{(L)}+M_{(H)}+Z_{(H)}Z_{(L)}Q/D\,, \tag{8}$$

where Q is the value of electronic charge, e^2, and D is the effective separation distance of the two fragment centers at the instant of scission. By minimizing the potential energy with respect to the charge of the light fragment the following equation is obtained.

$$\frac{\partial \text{P.E.}}{\partial Z_{(L)}}=0=\frac{\partial M_{(L)}}{\partial Z_{(L)}}+\frac{\partial M_{(H)}}{\partial Z_{(L)}}+\frac{\partial}{\partial Z_{(L)}}\left[\frac{Z_{(H)}Z_{(L)}Q}{D}\right]. \tag{9}$$

Using charge conservation [Eq. (2)] and some form of mass equation for $M_{(L)}$ and $M_{(H)}$, a relationship for the most probable charge of a fragment can be derived. If, for example, a mass equation such as that given by Green[41] is used, an equation of the following type is obtained (before neutron boil-off from fragments):

$$Z_{p(L)}=\frac{Z_c(a_1/A_{(H)}+a_2/A_{(H)}{}^{1/3}-0.5Q/D)}{a_2[1/(A_{(c)}-A_{(H)})^{1/3}+1/A_{(H)}{}^{1/3}]+a_1[1/A_{(H)}+1/(A_{(c)}-A_{(H)})-Q/D]} \tag{10}$$

where a_1 and a_2 are mass-equation constants.

[41] A. E. S. Green, Phys. Rev. **95**, 1006 (1954).

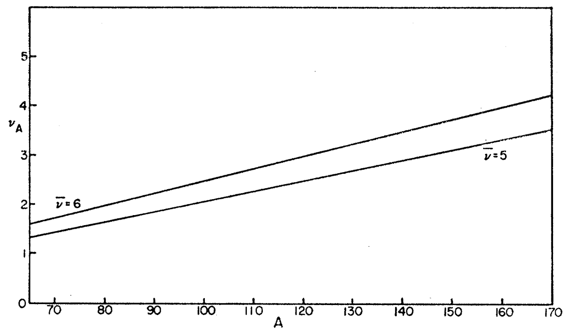

FIG. 1. Continuous neutron function I.

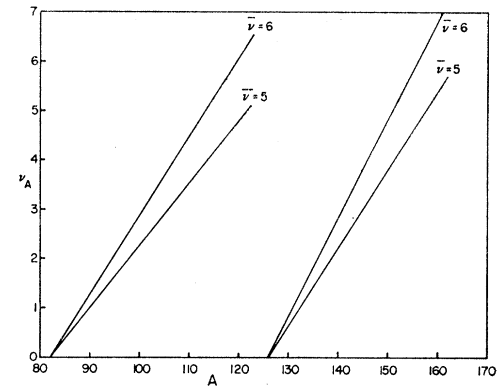

FIG. 2. Discontinuous neutron function II (see Table II).

The early investigators used values of Z_A obtained from simple Bohr-Wheeler theory.[42] In this research, both the non-shell-corrected mass equation of Green[41] and the shell-corrected mass equation of Levy[43] were used to derive Z_A or the mass-energy parameters of the M.N.P.E. treatment.

The sixth requirement, a description of the neutrons emitted by each primary fission fragment, is needed to relate the primary and secondary fission-product yields. Three functions, illustrated in Figs. 1, 2, and 3, were tested. The first (Fig. 1) is a simple linear function which assumes that neutrons are emitted in proportion to the total mass of the fragment

$$\nu_H = \bar{\nu}(m_H/m_C), \qquad (11)$$

where ν_H is the number of neutrons emitted by the heavy fragment, and $\bar{\nu}$ is the average number emitted per fission event; m_H and m_C are the masses of the heavy fragment and compound nucleus, respectively. The second and third functions (Figs. 2 and 3) are variations based on the observation of a discontinuous neutron emission as a function of mass by Terrell.[44] The main difference between these two functions and that used by Terrell is one of slope (Table II) and the provision for the emission of a constant number of neutrons from each fission fragment (see Table II).

TABLE II. Neutron functions.

Average total number of neutrons emitted, $\bar{\nu}$	Function No.[a]	Number of neutrons emitted by the individual fragment[b]
6	II	$\nu_L=0.16(A_L-82)$
6	II	$\nu_H=0.20(A_H-126)$
5	II	$\nu_L=0.126(A_L-82)$
5	II	$\nu_H=0.518(A_H-126)$
6	III	$\nu_L=1.50+0.08(A_L-82)$
6	III	$\nu_H=1.50+0.10(A_H-126)$
5	III	$\nu_L=1.0+0.08(A_L-82)$
5	III	$\nu_H=1.0+0.10(A_H-126)$

[a] II and III refer to the neutron function sin Figs. 2 and 3, respectively.
[b] A_L and A_H are the masses of the light and heavy primary fragments, respectively.

[42] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).
[43] Harris B. Levy, Phys. Rev. **106**, 1265 (1957).
[44] James Terrell, Phys. Rev. **127**, 880 (1962).

The most probable charges of the fission fragments from Am^{241*} and Cm^{243*} were calculated and intercompared (Table III) using all possible combinations of the three charge postulates, two mass equations for the M.N.P.E. and E.C.D. parameters,[45] and these three neutron functions. Comparison was made assuming that the fractional chain yields are described by a Gaussian function:

$$f_i = (c\pi)^{-1/2}e - (Z-Z_p)^2/c, \qquad (12)$$

where the experimental independent yields were converted to fractional chain yields from the isobaric yields of the respective mass curves.[40] Mass curves which were independent of the charge postulates were obtained from cumulative-yield data which had small ($\leq 10\%$) corrections from charge-distribution corrections. This procedure produced an experimental yield fraction that was independent of the charge rule. The fractional yields were fitted to Gaussian equations [Eq. (12)] by a modified[34] least-squares computer program.[39] The results are listed in Table IV, which includes the best single value of the Gaussian constant c. The "best"-fitted Gaussian curves for the Am^{241*} and Cm^{243*} data using E.C.D.–G–I are illustrated in Figs. 4 and 5. Fifteen of

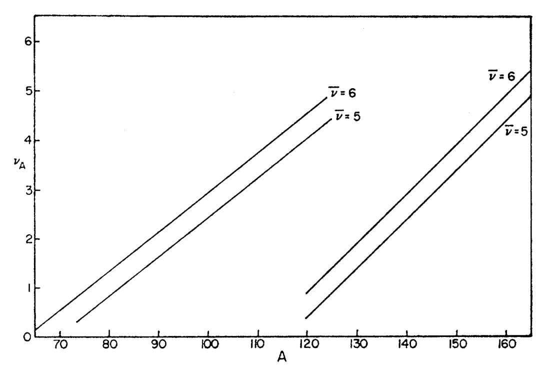

FIG. 3. Discontinuous neutron function III (see Table II).

[45] The values of the scission distance required in the M. N. P. E. treatment were selected from data of H. C. Britt, H. E. Wegner, and Judith C. Gursky, Phys. Rev. **129**, 239 (1963).

TABLE III. A comparison of Z_p calculated by different charge postulates.

A	Z	Neutron function[a]	C.C.R.	M.N.P.E.[b] G	M.N.P.E.[b] L	E.C.D.[b] G	E.C.D.[b] L
				$Z-Z_p(\mathrm{Cm}^{243})$			
76	33	I	2.35	1.35	2.20	1.67	2.06
		II	3.30	2.25	2.91	2.60	2.82
		III	2.72	1.68	2.36	2.04	2.19
82	35	I	1.91	1.05	1.71	1.36	1.75
		II	2.60	1.71	2.25	2.00	2.50
		III	2.15	1.26	1.85	1.56	1.86
86	37	I	2.32	1.53	0.59	1.80	1.30
		II	2.80	2.00	2.48	2.26	2.95
		III	2.43	1.64	0.70	1.92	1.45
96	41	I	2.28	1.70	2.28	1.92	1.66
		II	2.28	1.73	2.28	1.91	1.66
		III	2.05	1.58	2.16	1.78	1.56
112	47	I	1.85	1.65	1.84	1.72	1.65
		II	1.08	0.90	1.03	0.96	0.91
		III	1.29	1.13	1.26	1.20	1.15
130	53	I	0.56	0.85	1.01	0.75	1.41
		II	1.33	1.58	1.77	1.48	2.13
		III	0.99	1.27	1.46	1.18	1.82
136	55	I	0.15	0.55	0.65	0.43	0.10
		II	0.52	0.93	1.03	0.78	0.46
		III	0.37	0.78	0.91	0.66	0.33
140	57	I	0.53	0.27	1.16	0.87	0.75
		II	0.66	1.16	1.29	0.97	0.87
		III	0.61	1.13	1.28	0.98	0.83
142	59	I	1.72	2.28	2.05	2.10	2.05
		II	1.73	2.28	2.41	2.05	2.06
		III	1.74	2.31	2.48	2.12	2.08
160	65	I	0.48	1.46	1.03	1.13	0.82
		II	−0.72	0.30	−0.14	−0.04	−0.45
		III	0.76	1.73	0.46	1.38	0.03
				$Z-Z_p(\mathrm{Am}^{241})$			
76	33	I	2.28	1.33	1.93	1.68	1.88
		II	3.48	2.36	3.12	2.79	2.96
		III	2.61	1.55	2.24	1.96	2.14
82	35	I	1.85	1.03	1.56	1.35	1.66
		II	2.68	1.73	2.38	2.09	2.40
		III	2.03	1.14	1.74	1.48	1.81
86	37	I	2.23	1.50	0.62	1.78	1.32
		II	2.80	1.98	2.58[c]	2.29	2.70[c]
		III	2.32	1.54	0.69	1.85	1.40
96	41	I	2.18	1.69	2.19	1.87	1.63
		II	2.09	1.58	2.13	1.78	1.56
		III	2.03	1.50	2.03	1.72	1.65
112	47	I	1.72	1.60	1.73	1.65	1.55
		II	0.60	0.55	0.65	0.58	0.51
		III	1.14	1.07	1.20	1.11	1.68
130	53	I	0.45	0.70	0.82	0.61	1.36
		II	1.39	1.60	0.88	1.50	2.21
		III	0.93	1.18	1.38	1.08	1.82
136	55	I	0.03	0.41	0.58	0.28	−0.08
		II	0.42	0.83	0.95	0.65	0.28
		III	0.30	0.72	0.83	0.55	0.21
140	57	I	0.41	0.87	1.08	0.71	0.40
		II	0.45	0.98	1.11	0.75	0.43
		III	0.55	1.06	1.25	0.88	0.60
142	59	I	1.58	2.12	2.35	1.94	1.65
		II	1.46	2.04	2.21	1.80	1.50
		III	1.66	2.25	2.45	2.03	1.80
160	65	I	0.32	1.25	0.92	0.92	0.95
		II	−1.45	−0.30	−0.78	−0.73	−0.75
		III	−0.22	0.84	0.54	0.43	0.60

[a] I, I, and III refer to the neutron functions in Figs. 1, 2, and 3, respectively.
[b] G and L refer to the use of the Green and Levy mass equations, respectively.
[c] Crosses a closed-shell configuration in postfission neutron emission.

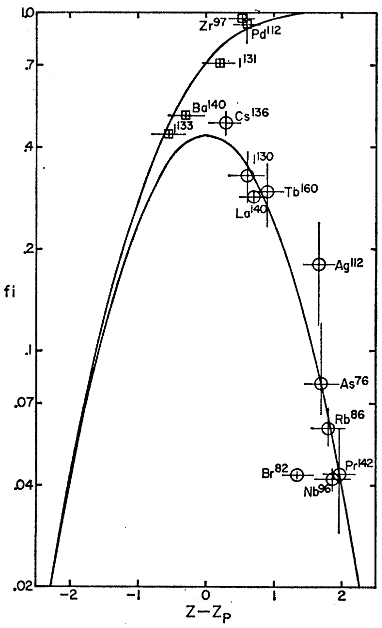

FIG. 4. Gaussian charge distribution for Am^{241*} using E.C.D.–G–I (see text). Square symbols refer to cumulative yields.

the twenty measured independent yields give a satisfactory Gaussian curve when correlated by the E.C.D. Further, the shape of the curve, although wider, is similar to that reported for the thermal fission of U^{235}.[11] The uncertainty in $Z-Z_p$ (± 0.25 charge units) is essentially a summation of the estimated (34) error introduced in the calculation[46] of the most probable charge, Z_p. The data from this research and those of Colby and Cobble[22] on plutonium isotopes can be fitted to a Gaussian distribution function with $c=1.7$. The difference between this Gaussian constant and that of Wahl[12] on U^{236*} (excitation energy = 6.5 MeV; $c=0.95$) can be due either to the increase in mass of the compound nucleus, or to the increase in excitation energy, or to both. Other recent investigations of protons on Th^{232} by Porile and Benjamin[47] indicate that the charge-distribution curve broadens with increased excitation energy (20–90 MeV), whereas the data of McHugh[21] compared with those of Wahl (6.5–39 MeV; U^{236*}) indicate a single charge-distribution curve. The assumption of a universal charge distribution (independent of mass number) may not, of course, be valid.[48]

A comparison of the Am^{241*} and Cm^{243*} fission data at various energies was made using a modification of the proposal of Coryell *et al.*[49] Using the 32.0-MeV Pu^{239} data as a standard, the Coryell expression was revised to include a variation of Z_A with A and to permit different numbers of neutrons to be emitted from the

[46] ± 0.15 units arise from the uncertainty in $\bar{\nu}$ (± 0.5 neutrons); ± 0.05 units from the estimation of preneutron emission; ± 0.02 units from an uncertainty in $\bar{D}$, and ± 0.03 units introduced by the arbitrary choice of constants in the Green mass equation.

[47] Phillips P. Benjamin, Ph.D. thesis, McGill University, Montreal, Canada, 1965 (unpublished).

[48] P. O. Strom, D. L. Love, A. E. Greendale, A. A. Delucchi, D. Sam, and N. E. Ballou, Phys. Rev. **144**, 984 (1966).

[49] C. D. Coryell, M. Kaplan, and R. D. Rink, Can. J. Chem. **39**, 646 (1961).

light and heavy fragments. The resulting equation is

$$\Delta Z_{p(\Delta_L)} = Z_{p(A)} - Z_{p(A)\text{reference}} = \tfrac{1}{2}(Z_c - 96)$$
$$-\tfrac{1}{2}(A_c - 243)\frac{dZ_{A(H)}}{dA} + 0.38\left(\frac{A_L}{A_c}\right)(\nu_T - 5.0). \quad (13)$$

In Eq. (13), dZ_A/dA actually varies from ~ 0.36 for the heavy-mass regions to ~ 0.41 for the light-mass regions. $\nu_L = \bar{\nu}(A_L/A_C)$ was used as the neutron function.

Figures 6 and 7 compare the most probable charge calculated from various postulates to the experimental values of Z_p read from the curves in Figs. 4 and 5. The data follow best the equal charge displacement postulate, but not without several serious deviations. The C.C.R. postulate can correlate the data almost as well as the E.C.D. treatment, while the M.N.P.E. results in a poorer fit.

It is possible, however, to eliminate certain sets of charge-distribution postulates and neutron functions from further consideration. For example, with some combinations, the experimental cumulative cross sections for the heavy mass numbers are corrected excessively high (Table V) so as to distort the mass-yield curve.[40] Therefore, the combinations of C.C.R., E.C.D.-Green mass equation, or E.C.D.-Levy mass equation charge rules with the discontinuous neutron function (II) of Fig. 2 were omitted from further study.

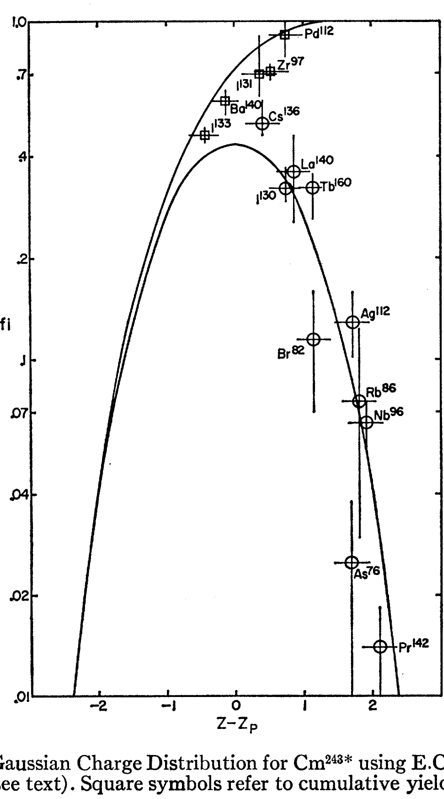

FIG. 5. Gaussian Charge Distribution for Cm^{243*} using E.C.D.–G–I (see text). Square symbols refer to cumulative yields.

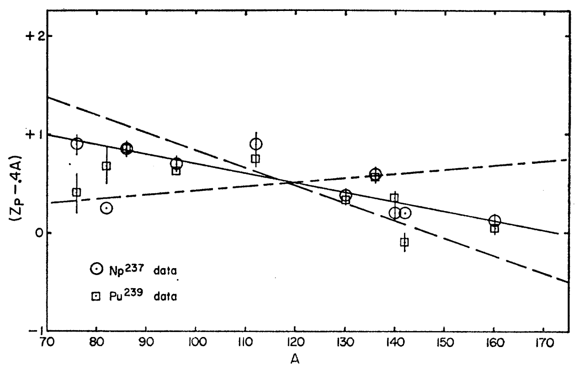

FIG. 6. A comparison of charge postulates as given by Eq. (13) using the mass equation of Green (see text). Symbols are as follows: C.C.R. — — —; E.C.D.(G,I) ——; M.N.P.E.(G,I)— —.

Over-all, two criteria indicate that a shell-correcting mass formula (such as the Levy equation) is not desirable in the present treatment. The first is that the experimental fission-product yield data do not fit a smooth mass-yield curve[34] after correction of the cumulative yields by a shell corrected Z_p function (Fig. 8).

TABLE IV. Values of the Gaussian constants obtained from various charge-distribution functions.

Postulate examined[a,b,c]	Gaussian constant, c	Sum of the square of the deviations
	Np^{237}	
E.C.D.–G–I	1.67 ±0.27	2.69×10⁻²
C.C.R.–I	2.49 ±0.50	2.88×10⁻²
C.C.R.–III	2.31±0.50	2.94×10⁻²
E.C.D.–G–III	1.89 ±0.37	3.64×10⁻²
E.C.D.–L–I	2.33 ±0.56	5.74×10⁻²
M.N.P.E.–G–III	1.53 ±0.33	6.87×10⁻²
M.N.P.E.–G–I	1.41 ±0.33	7.25×10⁻²
M.N.P.E.–L–II	4.43 ±1.97	7.62×10⁻²
E.C.D.–G–II	3.43 ±1.31	7.90×10⁻²
E.C.D.–L–III	2.63 ±0.81	8.62×10⁻²
M.N.P.E.–G–II	3.29 ±1.29	9.78×10⁻²
C.C.R.–II	5.02 ±2.76	10.0 ×10⁻²
M.N.P.E.–L–I	3.88 ±1.85	10.6 ×10⁻²
M.N.P.E.–L–III	4.19 ±2.24	11.6 ×10⁻²
E.C.D.–L–II	0.048±0.011	30.6 ×10⁻²
	Pu^{239}	
E.C.D.–G–I	1.75 ±0.39	3.43×10⁻²
C.C.R.–III	2.40 ±0.98	6.00×10⁻²
M.N.P.E.–G–I	1.50 ±0.46	7.63×10⁻²
E.C.D.–L–I	2.08 ±0.73	8.03×10⁻²
E.C.D.–G–III	2.95 ±1.37	10.9 ×10⁻²
C.C.R.–II	3.90 ±2.76	12.4 ×10⁻²
E.C.D.–L–III	3.00 ±1.61	14.0 ×10⁻²
E.C.D.–G–II	4.18 ±2.8	14.6 ×10⁻²
M.N.P.E.–L–I	3.40 ±2.35	14.9 ×10⁻²
M.N.P.E.–G–III	3.30 ±2.07	16.3 ×10⁻²
M.N.P.E.–G–II	4.43 ±3.34	17.0 ×10⁻²
E.C.D.–L–II	5.63 ±5.10	18.9 ×10⁻²
M.N.P.E.–L–III	5.43 ±5.56	19.6 ×10⁻²
M.N.P.E.–L–II	6.91 ±7.36	20.6 ×10⁻²
C.C.R.–I	2.31 ±7.85	37.1 ×10⁻²

[a] E.C.D., C.C.R., and M.N.P.E. refer to the use of equal charge displacement, constant charge ratio, and minimum nuclear potential energy postulates, respectively, in the calculation of the most probable charge.
[b] G and L refer to the use of the Green or Levy mass equation which are needed in the Z_p calculation.
[c] I, II, and III refer to the use of the various neutron functions (Figs. 1, 2, and 3, respectively).

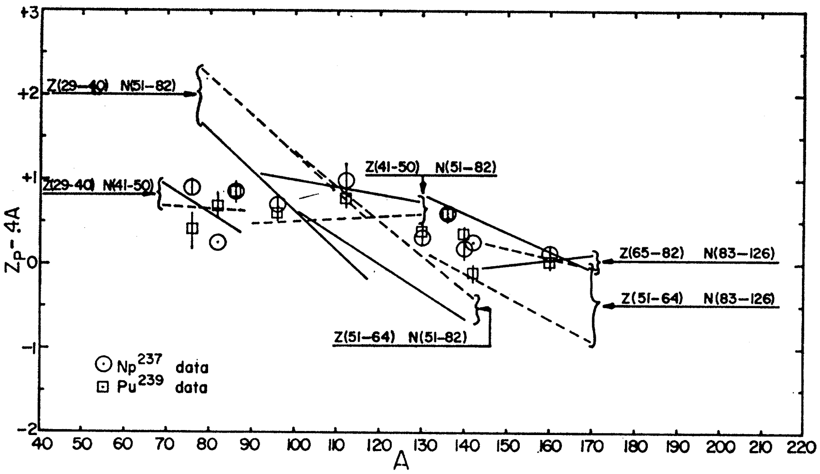

FIG. 7. A comparison of Charge postulates as given by Eq. (13) using the mass equation of Levy (see text). Symbols are as follows: E.C.D.(L,I) ——; M.N.P.E.(L,I) — —.

The second is that the resulting independent-yield data do not fit a Gaussian charge curve (Table IV).

Although the Ag^{112} and As^{76} data do not fit on the charge curve of Cm^{243*} (also Ag^{112} in Am^{241*}), the errors involved are such that these deviations are not thought to be significant. However, the Br^{82} data on both the Am^{241*} and Cm^{243*} curves have only a small total error. Other investigators have also obtained low Br^{82} yields in various medium-energy fission processes.[17,50] The existence of a metastable state of Br could account for the low yield if beta decay occurred form the excited state. However, this explanation is not valid since recent investigations[51,52] indicate a half-life of 6 min by a gamma transition $\geq 98\%$ of the time. Another possibility is chemical loss of Br^{82} produced in the target; however, the chemically similar I^{130} yields appear to be satisfactory. A third possibility of the 50-neutron shell (the primary fragment may be Br^{85}) having an effect seems improbable since both Cs^{136} and Rb^{86} have normal yields. These species are produced in primary fragments at or above the closed neutron shell configurations. It is also possible that the charge distribution varies with the mass chain in a way similar to that observed in the thermal-neutron-induced fission of U^{235}.[48] Therefore, the low Br^{82} yield is believed to be real but as yet unexplained.

The Nb^{96} data, on the other hand, are not low as had been previously found[53–56] in the thermal neutron fission of U^{233}, U^{235}, and Pu^{239}, or photofission[17] of U^{238} and Th^{232}. The previous explanation of either a stable 50-proton shell or an isomer state is not supported by this research or recent published[53,57–59] and un-

TABLE V. Total isobaric fission cross sections (mb).

	Np^{237} Fission (40.5 MeV)				Pu^{239} Fission (32.0 MeV)			
Isotope	σ obs.[a]	C.C.R.[b] II[d]	E.C.D.[b] G[c] II[d]	E.C.D. L[c] II[d]	σ obs.[a]	C.C.R. II	E.C.D. G II	E.C.D. L II
Zn^{72}	0.41	NC[e]	NC	NC	0.14	NC	NC	NC
As^{77}	3.38	NC	NC	NC	0.67	NC	NC	NC
Br^{83}	6.77	NC	NC	NC	5.6	NC	NC	NC
Pr^{145}	19.0	24.3	21.1	22.9	14.6	16.5	14.8	NC
Nd^{147}	14.3	18.3	15.4	16.6	11.5	12.5	11.7	11.6
Sm^{153}	5.01	23.9	8.95	10.4	5.22	9.49	6.37	5.38
Eu^{156}					1.86	6.41	2.86	1.90
Eu^{157}	1.27	127	12.7	17.4	1.56	15.6	4.22	1.98
Gd^{159}	1.09	109	9.4	10.9	1.63	14.2	3.79	1.85
Tb^{161}	0.79	79	6.07	6.07	0.91	7.02	1.82	3.97

[a] These values have been taken from Ref. 40.
[b] C.C.R. and E.C.D. indicate that the constant charge ratio and equal chain displacement postulates were used.
[c] G and L refer to the use of the Green and Levy mass equations.
[d] II refers to the use of the neutron function II (Fig. 2).
[e] NC indicates no significant change.

[50] R. A. Schmitt and N. Sugarman, Phys. Rev. **95**, 1260 (1954).
[51] J. Emery, J. Inorg. Nucl. Chem. **27**, 903 (1965).
[52] O. Anders, Phys. Rev. **138**, B1 (1965).
[53] I. F. Croall, J. Inorg. Nucl. Chem. **16**, 358 (1961).
[54] D. E. Troutner, Ph.D. thesis, Washington University, St. Louis, Missouri (unpublished).
[55] G. B. Cook, Ph.D. thesis, Cambridge University, 1951 (unpublished).
[56] E. Hagebø, J. Inorg. Nucl. Chem., **25**, 1201 (1963).
[57] A. C. Wahl and D. R. Nethaway, Phys. Rev. **131**, 830 (1963).
[58] David Troutner *et al.*, Phys. Rev. **134**, 1027 (1964).
[59] P. O. Strom, G. R. Grant, and A. C. Pappas, Can. J. Chem. **43**, 9, 2493 (1965).

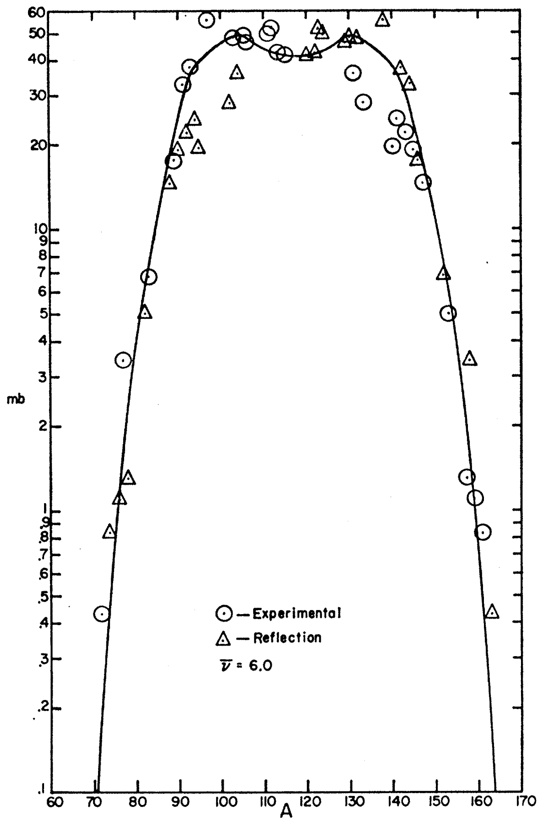

FIG. 8. Apparent mass distribution for the fission of Np^{237} induced by 40.5-MeV helium ions. Primary yields were corrected by M.N.P.E.–L–I (see text). The resulting distortion of the yield curve is evident.

published[60] data. Since this effect must take place at the expense of the complementary fragments, low yield would be expected for light fragments having one proton more or less than $Z=44$ and 46 in $_{95}Am^{241}$, or than $Z=45$ and 47 in $_{96}Cm^{243*}$ fission. The fact that the fission appears normal would seem to exclude any need for a special 50-proton shell effect in medium-energy fission. The normal Nb^{96} yield obtained in this research indicates the absence of any significant isomeric state. A normal yield for Nb^{96} has also been recently observed in $_{81}Tl^{201*}$ fission at 37 MeV.

[60] I. F. Croall, Ref. 16 of J. G. Cunninghame *et al.*, Nucl. Phys. 44, 588 (1963).

The experimental data do not unequivocally endorse any one charge-distribution postulate. The present investigation demonstrates a need for further study of independent yields in the 8- to 45-MeV excitation energy region, which would provide data on dZ_p/dE for various light and heavy fragments. A recent investigation[61] found that the Z_p value of a heavy fragment changed faster with energy than that for a light fragment, which implies that the E.C.D. rule would become increasingly less successful with increase in excitation energy. The E.C.D. rule postulates equal chain lengths which forces dZ_p/dE to be the same for the light and heavy fragments.

SUMMARY

The results of the present study on helium-ion-induced fission of Np^{237} and Pu^{239} at medium energies can be summarized as follows:

(1) The independent- and cumulative-yield data were most consistent, first with the equal charge displacement (E.C.D.) hypothesis, and second with constant charge ratio (C.C.R.). The poorest fit was obtained using the minimum nuclear potential energy (M.N.P.E.) treatment.

(2) A non-shell-corrected, continuous mass equation satisfactorily correlated the parameters of the E.C.D. hypothesis.

(3) The number of neutrons emitted by a fission fragment was consistent with the equation $\nu_H = \nu_T(m_H/m_C)$, where m_H and m_C represent the heavy and compound nuclear masses.

(4) There was no evidence for a 50-proton shell effect in the fission yields.

(5) The independent-yield data of Nb^{96} were normal.

ACKNOWLEDGMENTS

The authors wish to acknowledge the assistance and helpful discussions of C. Menninga and G. Raisbeck. The cooperation of M. Oselka and the staff of the Argonne National Laboratory 60-in. cyclotron is also greatly appreciated.

[61] D. R. Nethaway and H. B. Levy, Phys. Rev. 139, B1505 (1965).