Nuclear Charge Distribution in Fission: Fractional Yields of Krypton and Xenon Isotopes from Thermal Neutron Fission of U^{233} and Pu^{239} and from 14-MeV Neutron Fission of U^{235} and U^{238}*

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Measurements of fractional cumulative yields of Kr^{89} through Kr^{93}, Kr^{95}, and Xe^{137} through Xe^{144} from thermal neutron fission of U^{233} and Pu^{239} and of Kr^{91} through Kr^{93} and Xe^{138} through Xe^{141} from 14-MeV neutron fission of U^{235} and U^{238} have been made. These data and other published data show that isobaric charge dispersion for fission products with mass number 140 from thermal neutron fission of U^{233} and mass numbers 138 and 139 from 14-MeV neutron fission of U^{238} is consistent with the Gaussian curve determined for thermal neutron fission of U^{235}. This curve is used to estimate empirical values of Z_P, the "most probable charge" for a beta-decay chain, from the experimental data. From the variation of Z_P with fragment mass, it is indicated that there is a general deviation from unchanged charge distribution of ≈ 0.6 charge unit in favor of the light fragment for the four fission processes considered as well as for thermal neutron fission of U^{235}.

INTRODUCTION

ALTHOUGH charge distribution in thermal neutron fission of U^{235} has been fairly well characterized by a large number of measurements of fractional yields of nuclides in high-yield fission product chains,[1,2] knowledge about charge distribution for other types of fission has been largely limited to a few measurements of the independent yields of nuclides near stability.

This paper reports the results of measurements of the fractional cumulative yields of inert gas nuclides from fission of U^{233} and Pu^{239} with thermal neutrons and from fission of U^{235} and U^{238} with 14-MeV neutrons. The inert-gas fission products were separated from gross fission-product mixtures by the emanation technique of Wahl.[3] In this technique, the fissionable material is incorporated either as a stearate salt or as a deposit covered with a stearate salt in an evacuated container lined with filter paper. Those fission products that recoil into the stearate, and which are formed as inert gases or become inert gases as a result of beta decay, emanate from the stearate, and their descendants deposit on the filter paper liner; those fission products which do not have gas precursors remain in the stearate.

EXPERIMENTAL

Irradiations

Irradiations were performed in emanation containers similar to those used by Wahl.[3] For the determination of inert-gas yields from thermal neutron fission of U^{233} and Pu^{239}, thin oxide films of these isotopes were covered with barium or praseodymium stearate and placed in the containers. For determination of inert gas yields from 14-MeV neutron fission of U^{235} and U^{238}, preparations of uranyl stearate were used. The composition of the four fissionable materials is given in Table I.

TABLE I. Composition of fissionable materials expressed in terms of atom percent.

Pu^{239}:	94.3% Pu^{239}, 5.4% Pu^{240}, 0.3% Pu^{241}
U^{233} :	>98% U^{233}
U^{235} :	1.07% U^{234}, 93.13% U^{235}, 0.71% U^{236}, 5.09% U^{238}
U^{238} :	0.70% U^{235}, 99.30% U^{238}

Thermal neutron irradiations were made in the 4W port of the Los Alamos Water Boiler reactor in thermal neutron fluxes of $(1 \text{ to } 6)\times 10^{12}$ cm^{-2} sec^{-1}. The duration of irradiation varied from 5 min to 3 h depending on the yields and half-lives of the fission products which were determined; however, the number of fissions were limited to 2×10^{12} to avoid excessive radiation decomposition of the stearates.

Irradiation with nominal 14-MeV neutrons were made at the Los Alamos Cockcroft-Walton accelerator where fast neutrons were produced through the $T(d,n)He^4$ reaction by bombarding zirconium-tritium targets with 350-keV deuterons. The target materials were located about 2 cm from the neutron source in a flux of about 4×10^8 cm^{-2} sec^{-1}. The target was so oriented that the energy of the neutrons impinging on it was (14.6 ± 0.2) MeV.[4] Irradiation times varied from 30 min to 3 h; $(0.4 \text{ to } 3)\times 10^9$ fissions were produced. Under these experimental conditions, the ratio of total yields of Cd^{115} and Mo^{99} obtained from irradiation of stearate containing enriched uranium was the same as that obtained by Ford and Gilmore[5] from irradiation of enriched uranium metal with 14.7-MeV neutrons. This indicated that there was very little degradation of energy of the fission-producing neutrons as a result of

* Work performed under the auspices of the U. S. Atomic Energy Commission.
[1] A. C. Wahl, R. L. Ferguson, D. R. Nethaway, D. E. Troutner, and K. Wolfsberg, Phys. Rev. **126**, 1112 (1962).
[2] A. C. Wahl, A. E. Norris, and R. L. Ferguson, 1964 (private communication), and A. E. Norris, Ph.D. thesis, Washington University, 1963 (unpublished).
[3] A. C. Wahl, J. Inorg. Nucl. Chem. **6**, 263 (1958).
[4] J. D. Seagrave, Los Alamos Scientific Laboratory Report LAMS-2162, 1958 (unpublished).
[5] G. P. Ford and J. S. Gilmore, Los Alamos Scientific Laboratory Report LA-1997, 1956 (unpublished).

TABLE II. Fractional cumulative yields from thermal neutron fission of U^{233} and Pu^{239}.

Fission product	Half-life[a]	Emanating power[b]	Descendant determined	$U^{233}(n_{th}, F)$ Average $G\times100$[c]	$U^{233}(n_{th}, F)$ % Cumulative yield[d]	$Pu^{239}(n_{th}, F)$ Average $G\times100$[c]	$Pu^{239}(n_{th}, F)$ % Cumulative yield[d]
Kr^{89}	3.1 min	1.00	Sr^{89}	561 ±5	86.1 ±5	543 ±5	85.6 ±0.5
Kr^{90}	32 sec	1.00	Sr^{90}	188 ±8	66.5 ±1.2	166 ±5	64 ±1
Kr^{91}	8.4 sec	1.00	Sr^{91}	47.7 ±0.9	33 ±1	43.4 ±0.8	31 ±1
Kr^{92}	1.9 sec	>0.98	Sr^{92}	14.1 ±0.2	12.7 ±0.5	12.3 ±0.2	11.3 ±0.5
Kr^{93}	1.2 sec	>0.97	Y^{93}	2.36 ±0.03	2.3 ±0.1	2.04 ±0.03	2.05 ±0.10
Kr^{95}	~1 sec	>0.95	Zr^{95}	≤0.01	≤0.01	0.023±0.005	0.024±0.006
Xe^{137}	3.9 min	1.00	Cs^{137}	767 ±20	89.6 ±0.5	957 ±15	91.7 ±0.5
Xe^{138}	16±2 min	1.00	Cs^{138}	...	82.7 ±1.2	...	85.1 ±1.0
Xe^{139}	40 sec	1.00	Ba^{139}	87.1 ±0.4	47.6 ±0.8	115.2 ±0.5	54.6 ±0.8
Xe^{140}	13 sec	1.00	Ba^{140}	28.1 ±0.3	22.5 ±0.6	41.6 ±0.4	30.1 ±0.8
Xe^{141}	1.7 sec	>0.98	La^{141} and Ce^{141}	5.16 ±0.05	5.1 ±0.3	8.35 ±0.10	7.9 ±0.4
Xe^{142}	1.2 sec	>0.97	La^{142}	0.96 ±0.04	$0.98_{-0.05}^{+0.08}$	1.63 ±0.05	$1.65_{-0.07}^{+0.12}$
Xe^{143}	1 sec	>0.96	Ce^{143}	0.070±0.001	$0.072_{-0.003}^{+0.006}$	0.248±0.004	$0.26_{-0.01}^{+0.02}$
Xe^{144}	~1 sec	>0.96	Ce^{144}	≤0.03	≤0.03	0.028±0.005	0.029±0.006

[a] Kr^{89-93}, Xe^{137}, and $Xe^{139-143}$: P. Patzelt and G. Herrmann, 1964 (private communication). $Kr^{94,95}$ and Xe^{144}: Refs. 1 and 3. Xe^{138}: selected for consistency with the following divergent values: 18±1 min, D. W. Ockenden and R. H. Tomlinson, Can. J. Chem. **40**, 1594 (1962); 14.0±0.2 min, W. B. Clarke and H. G. Thode, Can. J. Phys. **42**, 213 (1964); and 14.5±0.4 min, P. Patzelt and G. Herrmann, 1964 (private communication).
[b] Estimated from Eq. (7) of Ref. 1. The effective time spent by an inert gas in barium or praseodymium stearate is <0.056 sec. This value is based on emanating power for 4.0-sec Rn^{219} of >0.99 determined for barium stearate (Ref. 17) and for praseodymium stearate (this work).
[c] Defined in text.
[d] Calculated from Eq. 5 or 6 of Ref. 1. The efficiency of collection of the active deposit was 0.98±0.03, and the fraction of the empty space in the irradiation container between the grains of the stearate was 0.011±0.002.

scattering in the stearate or from the walls of the container.

Procedure

In all experiments except those involving the yield of Xe^{138}, the irradiation container was opened after essentially all of the krypton or xenon isotopes of interest had decayed. In those experiments in which the yield of Xe^{138} was measured, the container was opened about 15 min after thermal neutron irradiations of 5 or 10 min and about 5 or 10 min after 14-MeV neutron irradiations of about 30 min. In each experiment the filter paper liner of the container and the stearate were dissolved separately and fission product activities of interest were determined by modifications of standard radiochemical procedures (strontium,[6] yttrium,[7] molybdenum,[8] cadmium,[8] cesium,[9] barium,[10] lanthanum,[7] and cerium[11]).

The ratio of the activity of a late member fission product from the irradiation container liner to the activity of the same fission product in the stearate is defined as G. The methods of calculating the fractional cumulative yields of inert gases from this value are described in Ref. 1.

Since a significant fraction of the fissions with 14-MeV neutrons occurred in both U^{235} and U^{238} in each of the two isotopic compositions of uranium listed in Table I, appropriate corrections were made to obtain the fractional cumulative yields from fission of each isotope. The fission cross sections of U^{235} and U^{238} with 14.6-MeV neutrons (2.21 and 1.26 b[12,13]) and the relative yields of the same mass numbers (extrapolated from the compilation by Katcoff[14]) were used in correcting the experimental data.

RESULTS

The results of measurements of fractional cumulative yields from fission of U^{233} and Pu^{239} with thermal neutrons and of U^{235} and U^{238} with 14-MeV neutrons are given in Tables II and III. The values of G represent the average of two determinations except in the cases of Xe^{141} where they represent the average of two measurements of La^{141} activities and two of Ce^{141} activities. Treatment of experimental data is summarized in the footnotes.

Two of the fractional cumulative yields for 14-MeV neutron fission of U^{238} have been reported by other investigators. For Xe^{138}, Apollonova *et al.*[15] reported a value of 0.917±0.005, in substantial agreement with the 0.94±0.02 value obtained in the present work. For

[6] L. E. Glendenin, in *Radiochemical Studies: The Fission Products*, edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, Inc., New York, 1951); National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV, p. 1460.
[7] K. Wolfsberg, Anal. Chem. **34**, 518 (1962).
[8] Los Alamos Scientific Laboratory Report LA-1721, edited by J. Kleinberg, 2nd ed., 1958 (unpublished).
[9] H. B. Evans, in *Radiochemical Studies: The Fission Products*, edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, Inc., New York, 1951), National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV, p. 1646.
[10] L. E. Glendenin, in *Radiochemical Studies: The Fission Products*, edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, Inc., New York, 1951); National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV, p. 1657.
[11] L. E. Glendenin, K. F. Flynn, R. F. Buchanan, and E. P. Steinberg, Anal. Chem. **27**, 59 (1955).
[12] H. L. Smith, R. K. Smith, and R. L. Henkel, Phys. Rev. **125**, 1329 (1962).
[13] R. K. Smith, 1964 (private communication).
[14] S. Katcoff, Nucleonics **18**, 201 (1960).
[15] A. N. Apollonova, I. T. Krisyuk, and V. N. Ushatskii, Radiokhimiya (USSR) **4**, 711 (1962).

TABLE III. Fractional cumulative yields from 14-MeV neutron fission of U^{235} and U^{238}.[a]

Fission product	Emanating power[b]	$U^{235}(n_{14}, F)$ Average $G\times100$[c]	$U^{235}(n_{14}, F)$ % Cumulative yield[d,e]	$U^{238}(n_{14}, F)$ Average $G\times100$[c]	$U^{238}(n_{14}, F)$ % Cumulative yield[d,e]
Kr^{91}	>0.98	55 ±2	36_{-1}^{+2}	175 ±5	65_{-1}^{+2}
Kr^{92}	>0.95	17.8 ±0.4	$15.6_{-0.5}^{+1.3}$	75.6±1.2	44_{-1}^{+3}
Kr^{93}	>0.92	3.9 ±0.2	$3.9_{-0.2}^{+0.5}$	18.9±0.4	$16.4_{-0.5}^{+1.6}$
Xe^{138}	1.00	···	75±2	···	94±2
Xe^{139}	1.00	71.6 ±1.5	42.9±0.9	373 ±5	80.5±0.6
Xe^{140}	>0.99	24.7 ±0.5	$20.4_{-0.6}^{+0.8}$	148 ±2	$61.1_{-0.8}^{+1.4}$
Xe^{141}	>0.94	5.77±0.07	$5.6_{-0.2}^{+0.6}$	39.1±1.5	29_{-1}^{+3}

[a] The footnotes of Table II apply unless otherwise noted. For the yields of Xe^{141} only La^{141} was determined.
[b] The effective time spent by an inert gas in uranyl stearate is <0.14 sec. This value is based on the emanating power of the preparations used for 1.9-sec Kr^{92} of >0.95.
[c] These are the measured values for each of the mixtures of uranium isotopes in Table I.
[d] The fraction of the empty space between the grains of uranyl stearate was 0.016±0.003.
[e] Corrections were made for the fissions of other isotopes in each material.

Xe^{139}, Krisyuk *et al.*[16] reported a value of 0.658, whereas the corresponding value from the present work was 0.805±0.006. In this case, the discrepancy may be due to the fact that Krisyuk *et al.* swept the xenon activity from solution with argon. Since Wahl and Daniels[17] could not quantitatively sweep 56-sec Rn^{220} from solution with air, it is likely that the emanating power of the argon-swept solutions was less than 100% for 40-sec Xe^{139}.

DISCUSSION

Isobaric Charge Dispersion Curves

Z_P is defined as the value of Z for the maximum of the independent yield-versus-Z curve and is often called the "most probable charge." For fission of U^{235} with thermal neutrons it has been shown[1,2] that for a number of isobaric chains the fractional cumulative yield of element with charge Z is given by the Gaussian equation

$$\sum_0^Z (P_n)=\frac{1}{\sigma(2\pi)^{1/2}}\int_{-\infty}^{Z+0.5}\exp[-(n-Z_P)^2/2\sigma^2]dn$$
$$=\tfrac{1}{2}+\tfrac{1}{2}f[(Z-Z_P+\tfrac{1}{2})/\sigma], \quad (1)$$

in which $f(X)$ is the normal probability integral. From measurements of two or more fractional yields of each of ten beta decay chains a value of 0.59±0.06 for σ has been obtained.[2] These ten chains all represent masses with high fission yields.

There are few data on isobaric charge dispersion for other types of fission. Values of the ratio of the independent yield of xenon to the cumulative yield of iodine have been reported[15,18] for the mass-138 and mass-139 chains from 14-MeV neutron fission of U^{238}. The independent yield of La^{140} from thermal neutron fission of U^{233} has been measured.[19] If a Gaussian curve is assumed to hold, these measured values can be used with the values of the cumulative yields of the respective xenon isotopes to get values of σ and Z_P by the method which is described in Ref. 1. The values obtained are listed in Table IV. The σ's are consistent with the value of 0.59±0.06.

TABLE IV. Charge dispersion data.

Fission product	Fractional cumulative yield	Z_P	σ
$U^{233}(n_{th}, F)$:			
Xe^{140}	0.225 ±0.006[a]		
Ba^{140}	0.9962±0.0001[b]	54.94±0.03	0.59±0.03
$U^{238}(n_{14},F)$:			
I^{138}	0.56 ±0.03[c]		
Xe^{138}	0.94 ±0.02[d]	53.40±0.08	0.75±0.10
I^{139}	0.22 ±0.02[e]		
Xe^{139}	0.805 ±0.006[a]	53.98±0.07	0.63±0.05

[a] From this paper.
[b] From Ref. 19.
[c] Calculated from the ratio of the independent yield of Xe^{138} to the cumulative yield of I^{138} of 0.67±0.05 (Ref. 15) and the cumulative yield of Xe^{138}.
[d] From this paper and Ref. 15.
[e] Calculated from the ratio of the independent yield of Xe^{139} to the cumulative yield of I^{139} of 2.6±0.2 (Ref. 18) and the cumulative yield of Xe^{139}.

Charge dispersion curves used by other workers[20–22] for fission processes at moderate excitation energies induced by charged particles are essentially the same as that used for thermal neutron fission of U^{235}. Accordingly, we shall assume that the shape of the charge dispersion curve is the same for all chains for the fission processes considered, and empirical values of Z_P will be calculated through the use of Eq. 1 with $\sigma=0.59$ ±0.06. These values appear in Table V.

Division of Charge in Fission

Since charge division between fission fragments occurs before the emission of prompt neutrons, it is of interest to consider Z_P's as a function of the fragment mass number, A'. The average fragment mass number is obtained by increasing the product mass number, A, by ν, the average number of neutrons emitted from the fragments. For thermal neutron fission of U^{233} and Pu^{239}, ν may be estimated from the "universal" neutron yield curve of Terrell[23]:

$$\nu_l=0.08(A_l'-82), \quad \nu_h=0.10(A_h'-126), \quad (2)$$

[16] I. T. Krisyuk, G. P. Levnev, and N. P. Platunova, Radiokhimiya (USSR) 2, 743 (1960) [English transl.: Soviet Radiochemistry 2, 231 (1962)].
[17] A. C. Wahl and W. R. Daniels, J. Inorg. Nucl. Chem. 6, 278 (1958).
[18] A. N. Apollonova, I. T. Krisyuk, and V. N. Ushatskii, Radiokhimiya (USSR) 4, 587 (1962) [English transl.: Soviet Radiochemistry 4, 515 (1962)].
[19] W. E. Grummitt and G. M. Milton, J. Inorg. Nucl. Chem. 5, 99 (1957).
[20] H. M. Blann, University of California Report UCRL-9190, 1960 (unpublished).
[21] J. A. McHugh, Jr., University of California Report UCRL-10673, 1963 (unpublished).
[22] B. D. Pate, J. S. Foster, and L. Yaffe, Can. J. Chem. 36, 1691 (1958).
[23] J. Terrell, Phys. Rev. 127, 880 (1962).

where subscripts l and h refer to the light and heavy fragments. Neutron yield curves for 14-MeV neutron fission are estimated in the Appendix from neutron-yield curves obtained for other fission processes at moderate excitation energies. Average mass numbers for the fissioning nuclei are also estimated in the Appendix.

A convenient display of the division of charge in fission is a plot of the deviation from unchanged charge distribution, $Z_P - A'(Z_F/A_F)$, as a function of fragment mass[1] (see Fig. 1). Z_F and A_F are the charge and average mass number of the fissioning nucleus. The abscissa and ordinate are folded to superimpose values for complementary fragments. If there were no deviation from unchanged charge distribution, the charge density of the fragments would be the same as that of the fissioning nucleus, and the value of the ordinate would be zero. Data from thermal neutron fission of U^{235} indicate

TABLE V. Empirical Z_P values.

Fission product	A'	Fractional chain yield[a]	Z_P (empricial)
$U^{233}(n_{th}, F)$:			
Br^{80}	80.0	1×10^{-3} [b]	<32.87
Br^{82}	82.0	$(1.1\pm0.5)\times10^{-3}$	32.69±0.25
Rb^{86}	86.3	7×10^{-5}	34.25±0.23
Kr^{89}	89.6	*0.861±0.005	35.86±0.08
Kr^{90}	90.7	*0.665±0.012	36.25±0.04
Y^{90}	90.7	$<8\times10^{-5}$	<36.50
Kr^{91}	91.8	*0.33 ±0.01	36.76±0.04
Kr^{92}	92.9	*0.127±0.005	37.17±0.08
Kr^{93}	94.0	*0.023±0.001	37.67±0.12
Kr^{95}	96.1	$*\leq1\times10^{-4}$	>38.47
Nb^{96}	97.2	$(1.3\pm0.2)\times10^{-3}$	38.72±0.11
Nb^{97}	98.3	$(1.1\pm0.4)\times10^{-4}$	39.32±0.21
I^{128}	128.2	$(1.0\pm0.1)\times10^{-4}$	50.31±0.21
I^{130}	130.4	$(1.3\pm0.2)\times10^{-3}$	50.72±0.16
Te^{131m}	131.5	0.23	51.08±0.04
Cs^{136}	137.1	0.014±0.004	53.21±0.20
Xe^{137}	138.2	*0.896±0.005	53.76±0.09
Xe^{138}	139.3	*0.827±0.012	53.95±0.08
Xe^{139}	140.4	0.476±0.008	54.54±0.02
$A=140$	141.6	(From Table IV)	54.94±0.03
Xe^{141}	142.7	*0.051±0.003	55.46±0.11
Xe^{142}	143.8	$*(9.8_{-0.5}{}^{+0.8})\times10^{-3}$	$55.88_{-0.25}{}^{+0.15}$
Xe^{143}	144.9	$*(7.2_{-0.3}{}^{+0.6})\times10^{-4}$	56.38±0.18
Xe^{144}	146.0	$\leq3\times10^{-4}$	>56.32
$Pu^{239}(n_{th}, F)$:			
Rb^{86}	86.3	3.1×10^{-5}	34.14±0.24
Kr^{89}	89.6	*0.856±0.005	35.87±0.08
Kr^{90}	90.7	*0.64 ±0.01	36.29±0.05
Kr^{91}	91.8	*0.31 ±0.01	36.79±0.05
Kr^{92}	92.9	*0.113±0.005	37.21±0.08
Kr^{93}	94.0	$*(2.05\pm0.10)\times10^{-2}$	37.70±0.13
Kr^{95}	96.1	$*(2.4\pm0.6)\times10^{-4}$	38.56±0.24
Nb^{96}	97.2	$(7.7\pm1.0)\times10^{-4}$	38.63±0.21
Nb^{97}	98.3	$(1.5\pm0.4)\times10^{-2}$	39.22±0.18
I^{128}	128.2	$(2.4\pm0.2)\times10^{-4}$	50.44±0.22
I^{130}	130.4	$(2.0\pm0.1)\times10^{-3}$	50.80±0.16
Te^{131m}	131.5	0.18	50.97±0.05
Cs^{136}	137.1	$(1.0\pm0.7)\times10^{-2}$	$53.13_{-0.42}{}^{+0.25}$
Xe^{137}	138.2	*0.917±0.005	53.68±0.07
Xe^{138}	139.3	*0.851±0.010	53.89±0.09
Xe^{139}	140.4	*0.546±0.008	54.43±0.02
Xe^{140}	141.5	*0.301±0.008	54.81±0.05
Xe^{141}	142.7	*0.079±0.004	55.33±0.10
Xe^{142}	143.8	$*(1.65_{-0.07}{}^{+0.12})\times10^{-2}$	55.76±0.15
Xe^{143}	144.9	$*(2.6_{-0.1}{}^{+0.2})\times10^{-3}$	56.15±0.17
Xe^{144}	146.0	$*(2.9\pm0.6)\times10^{-4}$	56.53±0.24
$U^{235}(n_{14}, F)$:			
Br^{82}	82.6	$(1.6\pm0.1)\times10^{-3}$ [c]	32.56±0.17
Kr^{91}	92.4	$*0.36_{-0.01}{}^{+0.02}$	36.71±0.04
Kr^{92}	93.4	$*0.156_{-0.005}{}^{+0.013}$	37.10±0.08
Kr^{93}	94.5	$*0.039_{-0.002}{}^{+0.005}$	37.54±0.13
Nb^{96}	97.6	$(5.9\pm0.2)\times10^{-4}$ [c]	38.59±0.20
Te^{131m}	133.2	0.35 ±0.05[d]	51.31±0.09
I^{131}	133.2	<0.08[d]	<51.60
I^{132}	134.2	0.16 ±0.01[d]	51.92±0.08
I^{133}	135.2	<0.40[d]	<52.42
I^{134}	136.2	0.43 ±0.02[d]	52.45±0.06
Cs^{136}	138.2	0.045±0.005[c]	53.50±0.01
Xe^{138}	140.2	*0.75 ±0.02	54.12±0.07
Xe^{139}	141.2	*0.429±0.009	54.61±0.04
Xe^{140}	142.2	$*0.204_{-0.006}{}^{+0.008}$	54.99±0.07
Xe^{141}	143.2	$*0.056_{-0.002}{}^{+0.006}$	55.44±0.12

TABLE V (*continued*).

Fission product	A'	Fractional chain yield[a]	Z_P (empirical)
$U^{238}(n_{14}, F)$:			
Kr^{91}	92.2	$*0.65_{-0.01}{}^{+0.02}$	36.27±0.06
Kr^{92}	93.3	$*0.44_{-0.01}{}^{+0.03}$	36.59±0.04
Kr^{93}	94.3	$*0.164_{-0.005}{}^{+0.016}$	37.08±0.07
Cs^{136}	138.0	$(5.2\pm0.2)\times10^{-3}$ [c]	52.99±0.16
$A=138$	139.0	(From Table IV)	53.40±0.08
$A=139$	140.0	(From Table IV)	53.98±0.07
Xe^{140}	141.0	$*0.611_{-0.008}{}^{+0.014}$	54.33±0.04
Xe^{141}	142.0	$*0.29_{-0.01}{}^{+0.03}$	54.83±0.09

[a] Yields are taken from this paper or from the compilation in Ref. 1 unless another reference is given. Starred values are for cumulative yield, others for independent yield.
[b] T. G. Kennet and H. G. Thode, Phys. Rev. 103, 323 (1956).
[c] From Ref. 5.
[d] A. C. Wahl, Phys. Rev. 99, 730 (1955).

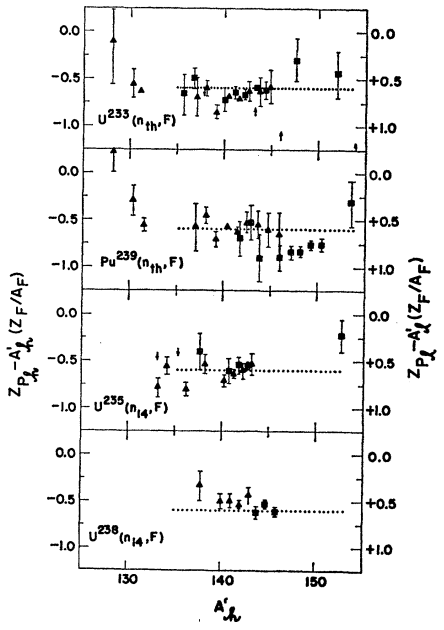

FIG. 1. Deviation from unchanged charge distribution. Triangles refer to empirical Z_P values for heavy fragments; squares refer to values for complementary light fragments. Dotted lines are for reference to show deviation of 0.6 charge unit.

that there is a deviation of ≈ 0.6 charge unit in favor of the light fragment for mass numbers made in high yield.[2] This average deviation is shown for reference in Fig. 1 by the dotted lines plotted at ordinates of $+0.6$ and -0.6 for the light and heavy fragments, respectively. The agreement of the data for these four fission processes with those from thermal neutron fission is remarkable. There are not enough data for mass numbers made in low yield (i.e., those near the valley or on the wings of the mass-yield curve) to deduce any general pattern.

The data in Fig. 1 for the mass-90 to mass-97 region for thermal neutron fission of Pu^{239} indicate that there is a larger deviation (≈ 0.8 charge unit as opposed to 0.6 charge unit). For fission of Pu^{239}, these mass numbers are no longer on a peak of the mass-yield curve, and indeed charge division may be greater for the more asymmetric splits. However, the effect may be only apparent. If the shape of the assumed charge distribution curve is no longer valid for these cases, then the deduced Z_P would be in error. Also, the uncertainty[23] of 0.3 to 0.4 mass unit for ν in the mass-90 region for thermal neutron fission of Pu^{239} causes a further uncertainty of 0.1 in the value of the ordinate of the data points in Fig. 1.

The implication of the deviation of ≈ 0.6 charge unit from unchanged charge distribution for low- and medium-energy neutron induced fission is that, at some time before scission during the fission act, a redistribution of charge occurs so that the light fragment has a higher charge density than the heavy fragment. For high-yield mass numbers, the redistribution amounts to an average transfer of ≈ 0.6 charge from the heavy fragment to the light fragment. This is consistent with the "neck model"[24–27] of fission in which light- and heavy-fragment cores are built around closed shells of nucleons, and in which the charge density of the heavy-fragment core is less than that of the light-fragment core and greater than that of the neck.

Predictions of Z_P

Since fractional yields have not been measured for most of the large number of fission-product nuclides produced in various fission processes, methods of predicting Z_P have been proposed by various investigators. Through the adoption of an isobaric charge dispersion curve, fractional yields can be inferred from predicted Z_P's. In this section, some of these methods will be compared with the experimental data for the four fission processes.

Two methods that are considered in detail in this paper are (1) the equal charge displacement postulate and (2) an adjustment of Z_P values from thermal neutron fission of U^{235}. A comparison of the predictions with the experimental data is shown in Fig. 2 with plots of $(Z_P-0.4A)$ versus A, a procedure that has been used by other authors.[28,29]

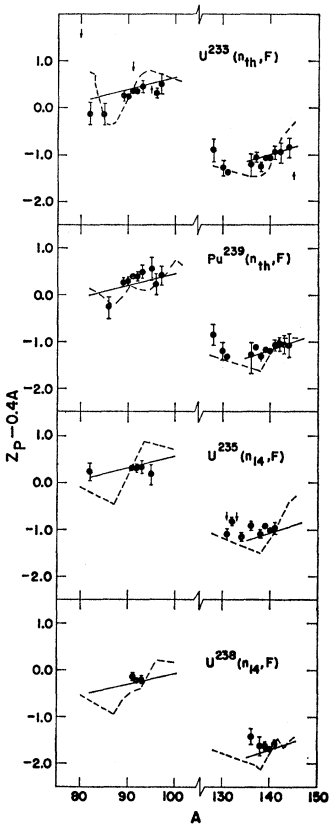

FIG. 2. Empirical Z_P values and proposed Z_P functions. Solid lines are the functions derived by the Coryell adjustment method. Dashed lines are the functions derived from the equal charge displacement postulate.

Fiedler and Herrmann,[28] using a new mass surface formula[30] and a modified version of the equal charge displacement postulate,[31] have reported a good fit of Z_P-versus-A data. Z_P values for Fig. 2 were calculated according to their prescription with the postulate applied after emission of neutrons. The number of neutrons emitted from fragments and the average mass numbers of the fissioning nuclei were estimated by the methods described in the preceding section and in the Appendix. The recommended Z_A values which were used were calculated by Mattauch and Thiele.[28,32]

Coryell *et al.*[29] have proposed a method which involves the use of Z_P's from thermal neutron fission of U^{235} as

[24] S. L. Whetstone, Phys. Rev. **114**, 581 (1959).
[25] R. B. Leachman, Trans. Am. Nucl. Soc. **5**, 19 (1962).
[26] R. B. Leachman, 1964 (private communication).
[27] P. Armbruster and H. Meister, Z. Physik **170**, 274 (1962) [English transl.: Atomic Energy Commission Report AEC-tr-5655, 1963 (unpublished)].
[28] J. Fiedler and G. Herrmann, Z. Naturforsch. **18a**, 553 (1963).
[29] C. D. Coryell, M. Kaplan, and R. D. Fink, Can. J. Chem. **39**, 646 (1961).
[30] L. A. König, H. Kümmel, W. Thiele, and A. Wapstra, Atomic Energy Research Establishment Report AERE-M-1078, pp. 35–36, 1962 (unpublished).
[31] L. E. Glendenin, C. D. Coryell, and R. R. Edwards, in *Radiochemical Studies: The Fission Products*, edited by C. D. Coryell and N. Sugarman (McGraw-Hill Book Company, Inc., New York, 1951); National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV, p. 489.
[32] J. Mattauch, 1964 (private communication).

reference values and then an adjustment of these values for other types of fission. The method was derived by taking the difference of Z_P's for two types of fission calculated by the equal charge displacement postulate. The need for explicitly calculating Z_A, which relies on assumptions about the mass surface, and the need for details about neutron emission, such as are described in the previous section and in the Appendix, are avoided. The following equations apply for this method[29]:

$$Z_P = Z_P^{\text{ref}} + \Delta Z_P$$
$$\Delta Z_P = 0.5(Z_F - Z_F^{\text{ref}}) - 0.21(A_F - A_F^{\text{ref}}) + 0.19(\nu_T - \nu_T^{\text{ref}}). \quad (3)$$

Superscripts "ref" apply to values from thermal neutron fission of U^{235}, A_F is in this instance the mass number of the initial compound nucleus, and ν_T is the average number of neutrons per fission including neutrons emitted from the compound nucleus before fission and neutrons emitted from fragments. In this paper values for ν_T are taken from Ref. 33. Z_P values apply to fission products after emission of neutrons.

If a deviation of 0.6 charge unit is assumed for mass numbers made in high yield from thermal neutron fission of U^{235}, the following equations apply:

$$Z_{P_l}^{\text{ref}} = A'(Z_F/A_F) + 0.6, \quad Z_{P_h}^{\text{ref}} = A'(Z_F/A_F) - 0.6.$$

After inserting the proper values for Z_F and A_F and estimating A' by using the ν's from Eq. 2, we obtain

$$Z_P^{\text{ref}} = 0.4237A - 2.19 \quad (\text{for } 82 < A < 100)$$

and

$$Z_P^{\text{ref}} = 0.4331A - 6.06 \quad (\text{for } A > 135). \quad (4)$$

Thus, Eq. 3 can be used with Eq. 4 to estimate Z_P values for mass numbers made in high yield. At the present time, not enough measurements have been made for low-yield mass numbers to make such generalizations.

The comparison of the two methods of predicting Z_P in Fig. 2 shows that the Coryell adjustment method predicts Z_P's better than the equal charge displacement postulate. For those mass regions in which both methods can be compared, the average deviation from the experimental points of Z_P's predicted by the adjustment method is 0.11 charge unit, whereas that of Z_P's predicted by the equal charge displacement postulate is 0.24 charge unit.

It should be pointed out that the application of the Coryell adjustment method to fractional yield data from charged-particle induced fission of moderate excitation energy, such as those of Colby and Cobble,[34] is not successful. Conversely, unchanged charge distribution, applied after emission of neutrons, which was used to describe data from charged-particle induced fission,[34] does not explain the data from 14-MeV neutron induced fission.

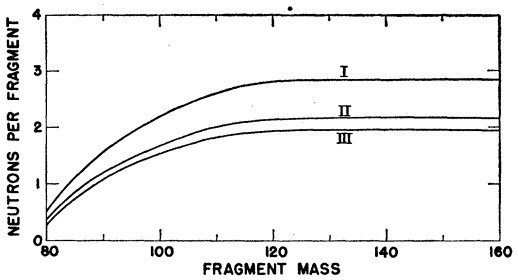

FIG. 3. Average number of prompt neutrons as a function of mass. Curve I represents the average of the curves determined by Britt and Whetstone. Curves II and III are the estimated curves for fission of U^{235} and U^{238} with 14.6 MeV neutrons.

APPENDIX: ESTIMATION OF ν FOR 14-MeV NEUTRON FISSION

Britt and Whetstone[35] have measured neutron emission from fragments produced in fission of U^{233} and Th^{230} with 29.5- and 25.7-MeV alpha particles. The average of the four neutron-yield-versus-A curves is represented by curve I in Fig. 3. Since these fission processes occur from uranium and plutonium nuclei with initial excitation energies between 20 and 25 MeV, and since 14-MeV neutron fission of U^{235} and U^{238} occurs from uranium nuclei with initial excitation energies of about 20 MeV, we shall use for the latter cases neutron emission curves which are similar to curve I, but which are normalized on the average number of neutrons emitted from fragments. These numbers have been calculated by Britt and Whetstone[35] for their cases, and the average of the four numbers is 4.93. For estimating this number for 14-MeV neutron fission of U^{235} and U^{238}, consider the following:

$$U^A + n(14.6 \text{ MeV})$$
$$\rightarrow U^{A+1}(E_1^*) \xrightarrow{-n} U^A(E_2^*) \xrightarrow{-n} U^{A-1}(E_3^*) \xrightarrow{-n}.$$
$$\downarrow f_1 \qquad \downarrow f_2 \qquad \downarrow f_3$$

The average excitation E^* of each compound nucleus that can undergo fission is calculated from the nuclear masses of König *et al.*[36] with the assumption that the average kinetic energy of neutrons evaporated from the uranium nuclei is 1.2 MeV.[35] The average number of neutrons emitted from fragments per fission (f_1,f_2,f_3) of each of the three nuclei was estimated by the method of Hanna and Clarke[37] in which ν is taken as 2.40 for a uranium nucleus with 6.0-MeV excitation energy and is then increased by 0.142 for each additional MeV of excitation. The fraction of the total num-

[33] Argonne National Laboratory Report ANL-5800, 2nd ed., 1963 (unpublished).
[34] L. J. Colby and J. W. Cobble, Phys. Rev. **121**, 1410 (1961).
[35] H. C. Britt and S. L. Whetstone, Phys. Rev. **133**, B603 (1964).
[36] L. A. König, J. H. E. Mattauch, and A. H. Wapstra, Nucl. Phys. **31**, 18 (1962).
[37] G. C. Hanna and R. L. Clarke, Can. J. Phys. **39**, 967 (1963).

ber of fissions resulting from each of the three uranium nuclei and the weighted average mass numbers of the fissioning nuclei were calculated from the Γ_n/Γ_f ratios of Vandenbosch and Huizenga.[38] For fission of U^{235} and U^{238} with 14.6-MeV neutrons, the average numbers of neutrons emitted from fragments per fission were estimated from these values to be 3.74 and 3.40, respectively, giving curves II and III of Fig. 1. The average mass numbers of the fissioning nuclei are 235.2 and 238.0. The neutron-emission curve of Whetstone[39] for 14-MeV neutron fission of U^{235} is quite different in shape from the curves considered above and has much larger uncertainties, but it gives essentially the same values of ν in the mass regions of interest.

[38] R. Vandenbosch and J. R. Huizenga, in *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy*, 1958 (United Nations, Geneva, 1958), Vol. 15, pp. 284–294.

[39] S. L. Whetstone, Phys. Rev. **133**, B613 (1964).

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Exact Multiple-Scattering Analysis of Low-Energy Elastic K^--d Scattering with Separable Potentials*

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A calculation of the K^--d low-energy elastic-scattering cross sections is carried out with the intent of determining the importance of multiple scatterings. Under the assumption that the two-body interactions are S-wave nonlocal separable potentials of the Yamaguchi form, an expression for the scattering amplitude in terms of a set of one-dimensional integral equations for each partial wave is derived. The derivation does not take into account Coulomb forces or the n-p and K^--$\bar{K}^0$ mass differences. A transformation from real to complex dummy variables that allows a rapid numerical computation of the solution to the integral equations for the scattering amplitude is presented and discussed. With the use of the Humphrey and Ross kaon-nucleon scattering lengths, the elastic angular distribution and cross section, as well as the total cross section, are calculated for incident kaon lab momenta of 105, 194, and 300 MeV/c. The results of the multiple-scattering calculation for the elastic cross section are two to three times smaller than the impulse approximation results throughout this momentum range. The multiple-scattering corrections to the impulse approximation for the total cross section are small ($\lesssim 10\%$) only at the largest momentum used.

I. INTRODUCTION

THE intent of this theoretical investigation is to determine the contribution of the multiple scattering terms in low-energy K^--deuteron elastic scattering. By multiple scattering (MS) terms we mean that part of the expansion of the K^--d t matrix beyond the sum of the free K^--n and K^--p t matrices. By low-energy we mean those center-of-mass energies which lie above the threshold for deuteron breakup and below about 70 MeV. Our attention will be focused for the most part on K^--d elastic scattering, but the formalism developed may be applied with varying degrees of modification to K^+-d, π-d, or N-d scattering (elastic or inelastic), or any scattering problem in which the forces are of short range and the target may be considered to be a two-particle composite.

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In the past two types of approximations have been applied to the MS series for scattering from deuterons. In the first of these the motion of the neutron and proton are treated adiabatically and the recoil of these particles when struck by the incident particle is neglected; i.e., the incident particle is considered to be scattered by two potentials a fixed distance apart, this distance being averaged according to the deuteron ground-state probability distribution after the scattering amplitude has been calculated. Here it is found that for several types of simplified two-particle t matrices the MS series for the scattering amplitude may be summed into a closed form. Such simplifications included nonlocal separable (NLS) t matrices,[1] zero-range t matrices,[2,3] and t matrices whose matrix elements off the energy shell,[1,4] or in all but one

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[4] T. B. Day, G. A. Snow, and J. Sucher, Nuovo Cimento **11**, 637 (1959); Phys. Rev. **119**, 1100 (1960).