In terms of these functions, the counting rate R is given as

$$R=\{\tfrac{2}{3}|I_t|^2+\tfrac{1}{3}(|I_1+I_2|^2-\sqrt{2}I_2I_3)\}\Omega, \quad \text{(A-14)}$$

keeping only the leading term in I_3 and dropping all factors of 2π, etc., since we are not concerned with the absolute normalization of R. Factor Ω is the phase-space factor which depends on the pair of variables observed. If E_1 and ψ_1 are observed, as in Figs. 1 and 2,

$$\Omega=\frac{2\pi M^2k^2p_1}{M+k-p_1\cos\psi_1}dE_1d\Omega_1. \quad \text{(A-15)}$$

The following constants were employed in the numerical evaluation of these functions:

$$\mu=139.63 \text{ Mev},$$
$$M_n/\mu=6.2786,$$
$$\alpha/\mu=0.3274,$$
$$\beta/\mu=1.54,$$
$$[\mu-(M_n-M_p)-B_0]/M_n=0.1449.$$

In employing the effective-range expansion, we used the first two terms, with $r_0=2.65$ f.

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Charge Distribution in the Fission of Uranium Isotopes Induced by 20–40 Mev Helium Ions*

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The primary yields of Br^{82}, I^{130}, La^{140}, Pr^{142}, and I^{135} have been accurately determined for the medium-energy (20–40 Mev) helium-ion-induced fission of U^{233}, U^{235}, and U^{238}. These accurate primary yield data have been correlated with the constant-charge-ratio rule for nuclides away from the neutron shells and give a smooth, but different, distribution curve for nuclides of 83 neutrons.

INTRODUCTION

THE details and mechanism of the primary nuclear charge distribution of fission fragments in higher energy fission have largely been inferred from studies involving the determination of primary yields of various fission products. In principle, if primary yield data for enough of the nuclides of a given mass chain could be obtained, a charge distribution curve could be constructed, the maximum of which would define Z_p, the most probable charge for that mass. A comparison of Z_p with that predicted by the various theories describing the nuclear arrangement at the moment of fission can yield valuable information on this important phenomenon. Therefore, the construction of such distribution curves based on primary yield data has been the goal of the majority of previous investigations. Since the necessary experimental conditions for the determination of the charge distributions of every fission product mass do not exist, it becomes necessary to assume that the charge distribution is essentially independent of mass to correlate data for different masses.

Charge distributions in fission were first considered for low-energy (thermal neutron) fission of U^{235} by Glendenin, Coryell, and Edwards[1] (and later modified by Pappas[2]). They obtained the most probable charge, Z_p, by postulating equal beta-decay chain lengths for the light and heavy fragments. This postulate is usually referred to as the equal-charge-displacement rule (E.C.D.).

Another hypothesis was proposed by Goeckermann and Perlman[3] to obtain Z_p values which would best correlate their primary yield data obtained from bismuth fission induced by 190-Mev deuterons. This hypothesis assumes that fission at high energies is so rapid that the charge distribution in the fragments is essentially the same as in the fissile nuclide, i.e., a constant-charge ratio (C.C.R.).

Steinberg and Glendenin[4] have adequately discussed these rules in a summary concerned with the radiochemical data on the fission process. Additional primary yield data obtained by mass spectrometric and radiochemical methods have appeared for neutron fission of

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[1] L. E. Glendenin, C. D. Coryell, and R. A. Edwards, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 52, National Nuclear Energy Series, Plutonium Project Record, Vol. 9.
[2] A. C. Pappas, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 7, p. 19; also Atomic Energy Commission Report AECL-2806, September, 1953 (unpublished).
[3] R. H. Goeckermann and I. Perlman, Phys. Rev. **76**, 628 (1949).
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U^{233}, U^{235}, U^{238}, and Pu^{239}.[5-11] The medium- and high-energy fission data have been obtained mostly by radiochemical techniques.[12] Hicks and Gilbert[13] studied U^{238} bombarded with 19–190 Mev deuterons, 70–340 Mev protons, and 50–380 Mev helium ions and found that the C.C.R. rule gave the best fit. Wahl[9] found the E.C.D. rule to be acceptable for the fission of U^{235} with 14-Mev neutrons. Gibson,[14] Thomas,[15] and Foreman[16] presented some charge distribution data from 20–40 Mev helium ions on Np^{237}, U^{233}, and Th^{232} and found that although the C.C.R. rule gave the best single fit, a mixture of the two rules might also correspond to the true situation. Data obtained by Alexander and Coryell[17] and U^{238} and Th^{232} bombarded with 13.6-Mev deuterons seemed to correlate best with the E.C.D. rule. Pate *et al.*[18,19] claimed that Th^{232} bombarded with 8–80 Mev protons is best correlated by the E.C.D. rule. Levrukhins and Krasavina[20] studied the fission of U^{238}, Th^{232}, and Bi induced by 480-Mev protons and found the C.C.R. rule was supported. A very interesting anomaly appears in the data of Porile and Sugarman[21] which shows the E.C.D. rule to correlate best for bismuth with 450-Mev protons and the C.C.R. rule to correlate best for tantalum with 450-Mev protons. Unfortunately, the experimental difficulties in obtaining the necessary data are great, and many of these apparent discrepancies can best be explained in light of the poor accuracy of much of the older high-energy data.

In this situation, it seemed desirable to attempt accurately to determine the absolute yields of specific "shielded" and "guarded" nuclides (I^{130}, Br^{82}, La^{140}, and Pr^{142}) from medium-energy helium-ion-induced fission of U^{233}, U^{235}, and U^{238} by improved radiochemical techniques.[22-25]

EXPERIMENTAL

The particular experimental procedure and conditions for bombardments, chemistry, and counting have been described previously.[26] The bombardment energies in this research were adjusted so that the mass yield curves for U^{235},[26] U^{233}, and U^{238} [27] could be used in correlating the primary yield data so obtained.

The shielded isotopes selected for this study were I^{130} and Br^{82} whose yields should not be affected by any closed shells; Pr^{142} and La^{140} which have 83 neutrons and whose yields, therefore, might be expected to show effects associated with the 82-neutron closed shell; and I^{135} which lies on the 82-neutron closed shell. By measuring the yields of these isotopes from the fission of U^{238}, U^{235}, and U^{233} at various helium ion energies, data were obtained under circumstances where the most probable charge for various masses varied over a wide and significant range.

The selection of the above isotopes was made primarily on the basis of the accuracy with which each could be assayed. The low activity due to Br^{82} was accurately determined by allowing all other shorter lived isotopes of bromine to decay out, and counting the resulting, longer lived Br^{82} activity in a low-level 2π flow counter, described by Griffioen and Cobble.[25]

A unique method for determining small activities of I^{130} in the presence of very large amounts of masking radiation from I^{131}, I^{133}, and I^{135} was developed. A calibrated 5-in. well-type NaI(Tl) scintillation crystal[22] was used as a "summing spectrometer." Fortunately, the decay[28] of the I^{130} gives the highest energy gamma-ray "sum" peak of all the observable iodine isotopes produced by fission. Using multichannel pulse-height analysis and a NaI(Tl) crystal detector, it was possible to measure activities of $\sim$100 counts/min of I^{130} in the presence of 10^6–10^7 counts/min of other iodine isotopes. The efficiency with which the 2.34-Mev sum peak was counted was 9.2%. This sum peak is assigned to I^{130} by its characteristic energy and half-life. Integration of its area allows the determination of I^{130} activity to $\pm$10%. This method is believed to be superior to previously used methods based on resolution of gross beta-decay curves.

The La^{140} activity was measured in a $2\pi\beta$ proportional counter as described and calibrated by Gunnink and Cobble.[24]

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The most difficult activity to assay was that due to Pr^{142}. The method used involved an accurate determination of the separate Pr^{145} and Pr^{143} activities (by 2π proportional counting), and accurate gross counting of the praseodymium sample at the time when the Pr^{145} activity had largely disappeared and the Pr^{142} activity was still detectable.

The I^{135} activity was determined by resolution of the gamma decay curve of I^{133} (the Xe^{135m} daughter having essentially the same gamma-decay energy). The activity attributed to I^{135} was then corrected for photopeak efficiency and intensities from decay scheme data. The significance of the cross section so obtained will be discussed later.

RESULTS

The primary yield data for the isotopes studied from the fission of U^{238}, U^{235}, and U^{233} at the various energies, as obtained in this research, are summarized in Tables I–II. The total chain yields, and the number of neutrons evaporated, $\bar{\nu}$, for U^{235} were those reported by Gunnink and Cobble[26]; the total chain yields and $\bar{\nu}$ for U^{238} and U^{233} are those obtained by Colby, Shoaf, and Cobble.[27] As will be discussed, the C.C.R. rule appears to provide the best correlation for the primary yield data; therefore, the last column in the tables lists the $\Delta Z = Z - Z_p$ of the isotope, as calculated by the C.C.R. rule for the particular compound nucleus and excitation energy.

TABLE I. Primary yield cross sections for helium-ion-induced fission of U^{238}.

Energy (Mev)	Isotope	Primary cross section (mb)	Total chain cross section (mb)	Percent chain yield (%)	ΔZ
39.9	Br^{82}	0.075	4.8	1.56	2.35
39.9	I^{130}	7.0	44	15.8	1.2
39.9	La^{140}	4.2	50.0	8.4	1.21
33.8	Br^{82}	0.038	3.8	1.0	2.5
33.8	I^{130}	3.78	37	10.2	1.42
33.8	La^{140}	1.8	43	4.2	1.48

TABLE II. Primary yield cross sections for helium-ion-induced fission of U^{235}.

Energy (Mev)	Isotope	Primary cross section (mb)	Total chain cross section (mb)	Percent chain yield (%)	ΔZ
39.9	Br^{82}	0.20	7.0	2.86	1.92
39.9	I^{130}	17.1	48.5	35.2	0.55
39.9	La^{140}	8.5	40	21	0.52
39.9	Pr^{142}	0.67	35	1.9	1.71
33.8	Br^{82}	0.12	4.8	2.5	2.05
33.8	I^{130}	11.1	37	30	0.78
33.8	La^{140}	4.3	34	12.6	0.78
33.8	Pr^{142}	0.48	33	1.45	1.96
28.2	Br^{82}	0.064	2.8	2.3	2.15
28.2	I^{130}	4.5	22.0	20.5	0.91
28.2	La^{140}	2.7	23	11.7	0.89
28.2	Pr^{142}	0.22	22	1.0	2.10

DISCUSSION

In the E.C.D. rule it is assumed that the two fission fragments are formed with neutron to proton ratios, n/p, such that maximum stability for the system is obtained. This rule reduces to equal beta-decay chain lengths for the light and heavy fragments. The C.C.R. rule assumes that the fission fragments will have the same neutron to proton ratio as the original fissioning nucleus. A most probable charge for each mass can be empirically defined on the basis of one or the other of these rules, and the experimental primary yield data can be so correlated. In order for one to make calculations from these two rules and in order for the data to have the widest applicability, the assumptions and information which are required are summarized as follows:

Equal Charge Displacement Rule

(1) The charge distribution function has the same shape for all mass numbers; (2) the target isotopes and excitation energies under consideration are so similar that the charge distribution is assumed independent of the $Z+N$ of the fissioning nucleus; (3) the charge distribution is symmetrical about the most probable charge, Z_p; (4) the average number of neutrons emitted, $\bar{\nu}$, per fission event must be known; (5) the number of neutrons emitted before and after fission must be established; (6) a distribution of post-fission neutrons between the light and heavy fragments must be assumed; and (7) a method for determining the most stable charge, Z_A, for a given mass chain must be selected.

Constant-Charge Ratio Rule

(1) The charge distribution has the same shape for all mass numbers (the same as E.C.D.); (2) the target

TABLE III. Primary yield cross sections for helium-ion-induced fission of U^{233}.

Energy (Mev)	Isotope	Primary cross section (mb)	Total chain cross section (mb)	Percent chain yield (%)	ΔZ
40.5	Br^{82}	0.59	6.5	9.09	1.62
40.5	I^{130}	22.1	49	45.1	0.10
40.5	La^{140}	10.9	35	31	0.01
40.5	Pr^{142}	1.41	23	6.1	1.20
34.5	Br^{82}	0.45	7.0	6.4	1.79
34.5	I^{130}	16.0	41	39.1	0.32
34.5	I^{135}	4.4	49	9.0	−1.67
34.5	La^{140}	8.9	34	26.2	0.28
34.5	Pr^{142}	0.63	24	2.6	1.48
29.0	Br^{82}	0.20	4.8	4.2	1.85
29.0	I^{130}	8.6	22	39	0.42
29.0	La^{140}	6.2	23	26.9	0.38
29.0	Pr^{142}	0.38	18	2.1	1.60
25.3	Br^{82}	0.106	2.8	3.8	1.92
25.3	I^{130}	4.93	13.5	36.1	0.55
25.3	I^{135}	2.3	18	12.8	−1.45
25.3	La^{140}	3.3	16.5	20.1	0.52
25.3	Pr^{142}	0.23	12	1.9	1.71

isotopes and excitation energies under consideration are so similar that the charge distribution is independent of the $Z+N$ of the fissioning nucleus (the same as E.C.D.); (3) the charge distribution is symmetrical about the most probable charge, Z_p (the same as E.C.D.); (4) the average number of neutrons emitted per fission event must be known (the same as E.C.D.); and (5) the postfission neutrons are assumed to be distributed between the fragments in proportion to the masses of the fragments.

The use of both of these rules requires that the average number of emitted neutrons per event be known. In the case at hand, these were known experimentally from previous work.[26,27] The assumption in the C.C.R. rule that postfission neutrons are emitted in proportion to their fragment masses is considerably reinforced (at least for those fission product masses investigated in this research) by the recent work of Whetstone[29] on neutron emission from the fragments in the spontaneous fission of Cf^{252}.

An attempt to correlate the data by the E.C.D.

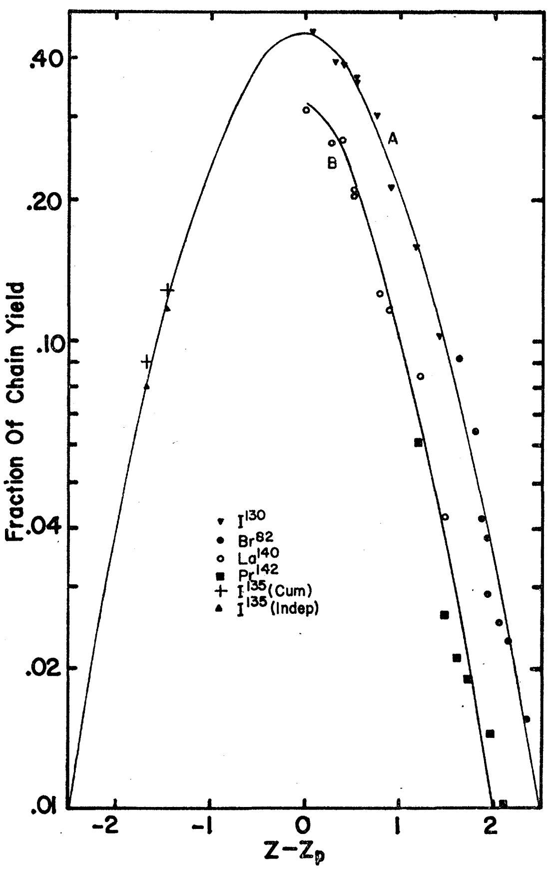

FIG. 1. Charge distribution curves from the medium-energy, helium-ion-induced, fission of the uranium isotopes assuming the constant-charge ratio (CCR) rule.

[29] S. L. Whetstone, Jr., Phys. Rev. **114**, 581 (1959).

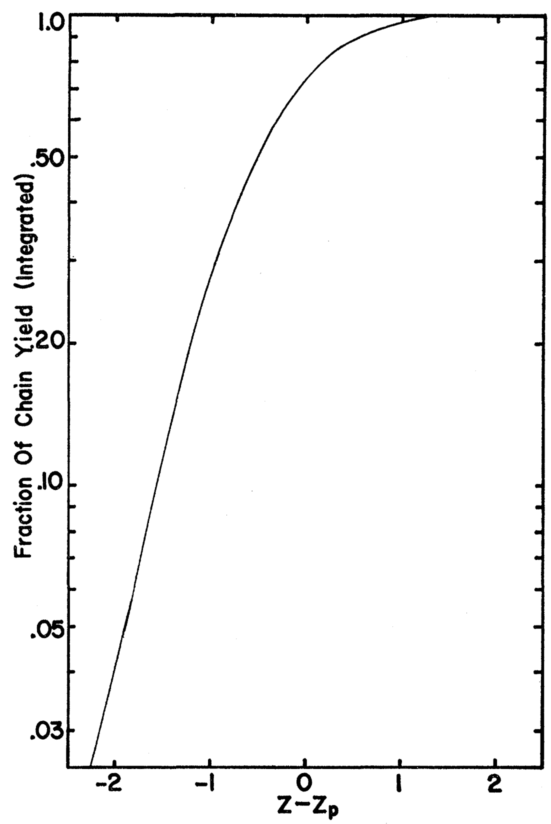

FIG. 2. Integrated fractional chain yield as a function of distance from the most probable charge, Z_p.

rule assuming the emission of one prefission neutron,[30] equal division between fragments for the postfission neutrons, and using the Z_A values of Coryell[31] was entirely unusuccessful.

It was interesting to find that the data obtained in this research correlated very well with the C.C.R. rule (see Fig. 1). It can be noted that the data for nuclides away from the closed shells (I^{130} and Br^{82}) show a smooth correlation (curve A in Fig. 1); and the data for nuclides next to the 82 neutron closed shell (La^{140} and Pr^{142}) show a separate, but also smooth, correlation (curve B in Fig. 1). Other investigators[9,14,15,26,32] had previously noted abnormally low yields for isotopes next to the 82-neutron closed shell and had debated the possibility of closed shell perturbations. The unperturbed distribution (curve A in Fig. 1) integrates to a value of 0.96 instead of unit total chain yield. Figure 2 shows the integrated fraction of a chain yield as a function of the distance from the most probable charge, $Z-Z_p$. This curve was normalized to unity and is convenient for correcting directly any given cumu-

[30] R. Vandenbosch, T. D. Thomas, S. E. Vandenbosch, R. A. Glass, and G. T. Seaborg, Phys. Rev. **111**, 1358 (1958).
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[32] Y. Y. Chu, Ph.D. thesis, University of California Radiation Laboratory Report UCRL-8926, November, 1959 (unpublished).

lative cross section to a total chain cross section for the isobar under investigation.

The perturbed distribution curve (curve B in Fig. 1) suggests that the nuclides formed with 83 neutrons (before postfission neutron emission) will on the average "boil off" approximately 25% more neutrons than those formed with greater than 83 or less than 81 neutrons. The primary nuclides formed with 82 neutrons will tend to boil off fewer neutrons on the average and, therefore, their primary yields after postfission neutron emission should be higher than predicted by yield systematics. It follows that the primary yields after neutron emission for fragments with 81 neutrons should again be lower than predicted. It is also possible that a similar effect might occur at the 50-neutron closed shell.

If the above description is correct, then the assumption that one obtains a smooth charge distribution curve for the yields of a series of isobars would only be valid when such a series does not extend across closed shells.

It can be seen that the most probable charge for mass 135 for the higher energy bombardments on U^{233} (in the case of I^{135}) gives large negative values for $\Delta Z = Z - Z_p$. This provides an opportunity to obtain primary yields with negative values for ΔZ. Since the yield of I^{135} is a cumulative yield, a correction must be made for the amount of I^{135} produced by beta decay from Te^{135} and Sb^{135}, which accounts for 10% of the total I^{135} observed activity. The results for the measured primary yield show good agreement with the unperturbed charge distribution (curve A in Fig. 1).

It might be expected, from arguments previously advanced, that the primary yield of this isotope, which has 82 neutrons, should be abnormally high, due to the fact that a related isotope of 82 neutrons would boil off $\sim$25% more neutrons than the average fission fragments. However, the actual yield for I^{136} is only about one sixth of the yield of I^{135} because it is 0.7 charge unit farther from Z_p. 25% of $\frac{1}{6}$ of the I^{135} yield amounts to an increase of only approximately 4% and thus the I^{135} yield falls on the unperturbed distribution curve (curve A in Fig. 1).

One can, however, also conclude from these data that the yield of a fission fragment before postfission neutron-emission is essentially unaffected by the closed shells.

Since only a few mass numbers were studied, it was not expected that the data would be conclusive in elucidating all the various aspects of charge distribution. They do, however, provide accurate primary yield corrections necessary for determining fission mass yield curves. It should also be noted that the supposition that the C.C.R. rule is more realistic than the original E.C.D. rule is supported by the fact that a large primary yield correction is necessary to obtain smooth mass-yield curves for the I^{133} and Ba^{140} isotopes.[26,27]

Since this work was completed, Chu[32] has reported mass spectrometric data on charge distribution in this 20–40-Mev excitation region for U^{235} and U^{236}. These results also indicate that the E.C.D. rule in its original form does not explain the charge distribution systematics for medium-energy fission when the effect of discontinuities at the closed shells on the Z_A function are considered (a conclusion previously drawn from this research). It was proposed, however, that if a continuous and linear Z_A function such as that given by Friedlander and Kennedy[33] were used and, further, if the number of emitted neutrons were adjusted to a value lower than experimentally observed for this energy region, then a reasonable fit for the data could be obtained. (The data from the present research is also in agreement with this modified E.C.D. procedure.) It was noted that these mass-spectrometric data plotted according to the C.C.R. rule also gave good agreement. The best agreement was obtained by varying the parameters and assumptions in the E.C.D. rule. The conclusions from this work favor an interpretation lying between the C.C.R. and modified E.C.D. rules. It is, however, quite significant that the data presented are very well correlated by the C.C.R. rule and that the small but real deviations might equally well be ascribed to certain invalid initial assumptions, e.g., that a charge distribution has the same shape for all mass numbers. This is also possibly the case in this research.

It is, therefore, concluded that the data presented in this paper support the present findings and that the correct interpretation of them lies near the C.C.R. rule but that some contribution from the E.C.D. rule presists.

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[33] G. Friedlander and J. W. Kennedy, *Nuclear and Radiochemistry* (John Wiley & Sons, Inc., New York, 1955), p. 50.