Bombardment Energy and Fission Product Yield Pattern for Protons on Natural Uranium and U^{235}

W. H. JONES,* A. TIMNICK,† J. H. PAEHLER, AND T. H. HANDLEY
Oak Ridge National Laboratory, Oak Ridge, Tennessee

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Relative fission yields of ten nuclides produced by proton-induced fission of natural uranium and U^{235} were determined at several proton energies in the range 12 to 20 Mev. The expected trend toward symmetric fission at increased energies is observed. A statistical relation is used to correlate this shift in fission symmetry with the excitation energy of the compound nucleus.

A NUMBER of investigations in recent years[1–7] support the conclusion that symmetric fission is increasingly favored at higher excitation energies of the compound nucleus. This laboratory has previously reported a preliminary study[8] of the influence of bombardment energy on the proportions of Mo^{99}, Ag^{111}, and Ba^{140} formed during proton-induced fission of uranium. Those results led to a more extensive series of experiments, including the work on natural uranium[9] and U^{235}, herewith described, and similar studies now in progress on U^{233}, Th^{232}, and Th^{230}.

The target assembly, Fig. 1, consists of four foils of natural uranium, or two foils of electrodeposited U^{235}, each about 25×6 mm in size, interleaved with calculated thicknesses[10] of aluminum to form a stack. One bombardment was also made with two foils of electrodeposited natural uranium. The thickness of the pure natural uranium foil was 11 mg/cm², while the thickness of the electrodeposited natural uranium and the U^{235} was of the order of 4 mg/cm². Special purity aluminum was used as a base for the deposits. The foil stack was attached inside the target head against the 11-mil-thick aluminum window, directly in contact with the cooling water. This head was then positioned to intercept the internal proton beam near the maximum radius of the 86-inch fixed-frequency proton cyclotron.[11] The usual bombardment was 100 μa for an hour. Detuning of the beam was helpful in avoiding "burning" and crumbling of the uranium foils.

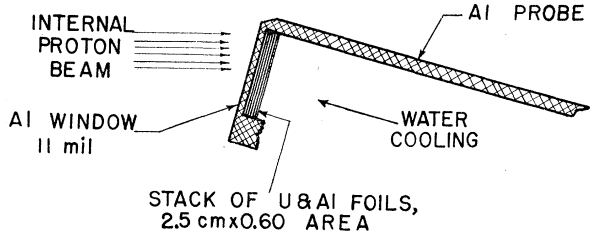

FIG. 1. Target assembly for proton bombardment of uranium foils.

The beam energy was determined in separate bombardment runs by producing the (p,n) and $(p,2n)$ reactions of Cu^{63} in a stack of copper foils.[11] Computation from known excitation curves[12] and the range-energy relations[10] for protons in copper gave 20.5±1.0 Mev as the beam energy. The energy of the protons reaching each uranium foil in the target stack was calculated from the beam energy thus established and the range-energy curves in aluminum and uranium. For bombardments of pure natural uranium, the first three foils were arranged to receive energies of ~18, 15, and 12 Mev, respectively, while the fourth foil was set beyond the proton range as a check on neutron fission background. For the electrodeposited natural uranium and U^{235}, two foils were arranged to receive energies of 20 and 16 Mev in a series of bombardments. In another series of bombardments of two foils of electrodeposited U^{235}, the first foil received 12 Mev, while the second was set beyond the proton range. The neutron fission background proved insignificant.

After the activity induced by a given bombardment run was allowed to diminish overnight, the target was disassembled and, in the case of the pure natural uranium, each foil was dropped into an individual flask containing nitric acid and silver as a carrier (the latter because of its strong absorption on glass). When dissolution was complete, the contents were diluted to 100 ml, and measured aliquots were used for determination, by radiochemical and counting methods, of the relative amount of each of the chosen fission products. The electrodeposited natural uranium and U^{235} foils were dropped into individual beakers containing nitric acid and warmed until active effervescence ceased. Each resulting solution was decanted, with its rinsings, to a volumetric flask.

* Research Participant from Emory University, Emory University, Georgia.

† On leave from Michigan State College, East Lansing, Michigan, 1952.

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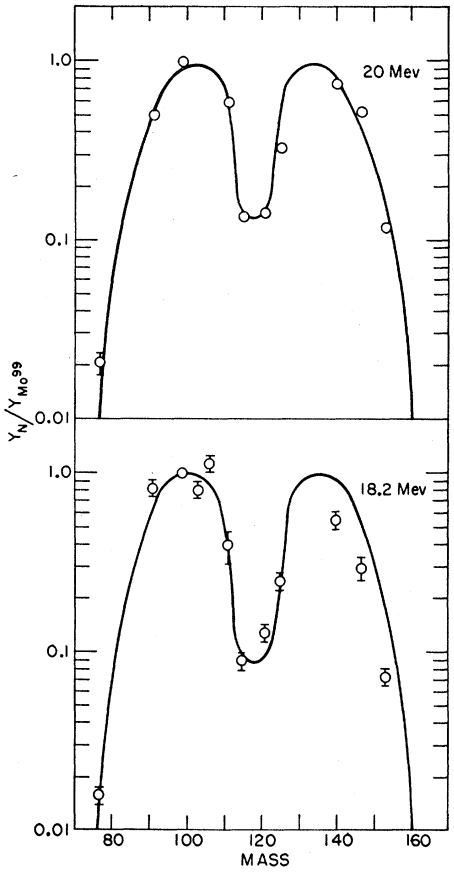

FIG. 2. Relative fission yields for natural uranium, 20 and 18.2 Mev.

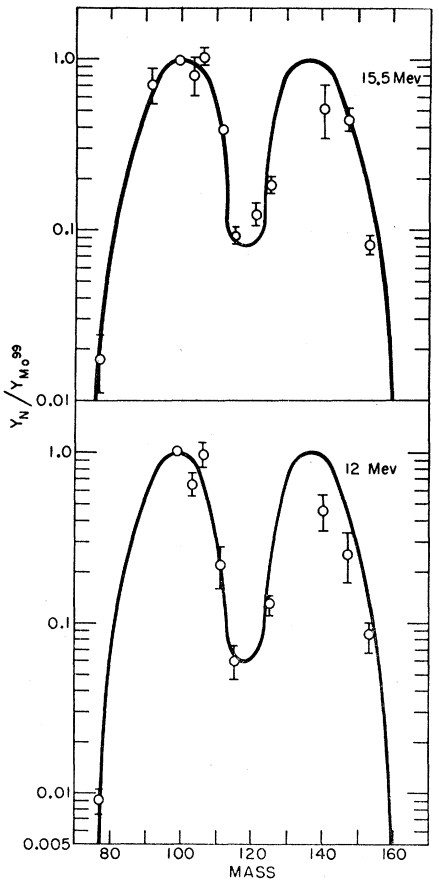

FIG. 3. Relative fission yields for natural uranium, 15.5 and 12 Mev.

On the basis of considerations of medium half-life, suitable intervals of mass, and practicability of the chemistry involved, the isotopes selected were, As^{77}, Y^{91}, Mo^{99}, Ru^{103}, Ag^{111}, Cd^{115}, Sn^{125}, Ba^{140}, Nd^{147}, and Sm^{153}. For most of these, the chemical procedures used were no departure from the standard methods described by Meinke,[13] the exceptions being molybdenum and the rare earths. For molybdenum, the ether extraction steps proved both tedious and inefficient; the use of silver nitrate as a final precipitant has been criticized.[14] The method adopted employs repeated α-benzoin-oxime[15] precipitations, a combination of scavenging operations, and final precipitation with lead nitrate.

The insoluble fluorides of the rare earths were isolated from the other elements present, then redissolved in a mixture of boric acid and concentrated nitric acid. After several scavengings,[16] the rare earths were precipitated as hydroxides, dissolved in hydrochloric acid and the solution introduced onto a column of Dowex-50 cation exchange resin, 250–500 mesh. This column, some 2 cm in diameter and 90 cm long, was maintained at 100°C by a steam jacket, after the design of Ketelle and Boyd.[17] A 5 percent solution of citric acid, kept sterile with a little phenol and adjusted very carefully with ammonia to *p*H 3.40, was used as the eluant. At this operating temperature an eluant throughput of 150 ml per hour was achieved by placing the reservoir two floors above the exchange column and reinforcing all connections. The receiving vessels were placed on a turntable controlled by an interval timer so set that each consecutive vessel received the column output for 30 minutes. Once routinized, this installation would dependably separate overnight the yttrium, samarium, and neodymium, in that order. Each was then precipitated as the oxalate, and the precipitate washed, transferred, and ignited to the oxide.

All precipitates in this work were bonded into stainless steel planchets by the addition of a trace of

[13] W. W. Meinke, U. S. Atomic Energy Commission Report AECD-2738 (1949) (unpublished).

[14] N. E. Ballou *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 257, National Nuclear Energy Series, Plutonium Project Record Vol. 9, 1538.

[15] Another point not adequately made in the literature is the apparent instability of the 2 percent alcoholic solution of α-benzoin-oxime used as precipitant. Old solutions precipitate very poorly; two weeks is suggested as the age limit for this reagent.

[16] Reference 14, p. 1673.

[17] B. H. Ketelle and G. E. Boyd, J. Am. Chem. Soc. **60**, 2800 (1947).

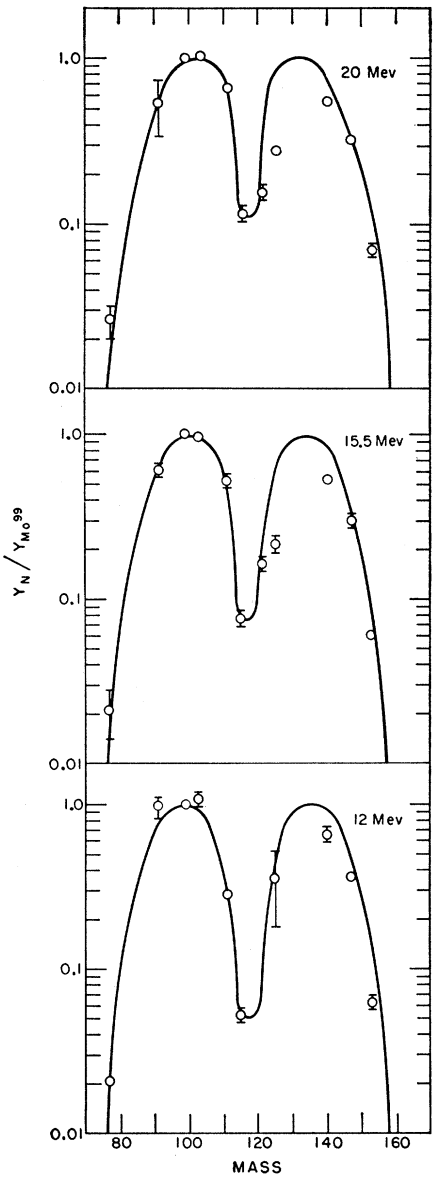

FIG. 4. Relative fission yields for uranium-235, 20, 15.5, and 12 Mev.

shellac to the alcohol used in transferring. These planchets were filed by number and their activities were counted at intervals of one, three, or seven days, over a period of several half-lives. For the most part, the resulting log-rate plots showed insignificant contamination, and the curves could be extrapolated to zero time with confidence. Mixed half-lives were involved in only three cases: Ru^{103}(40 day) *vs* Ru^{106}(365 day); Cd^{115}(54 hr) *vs* Cd^{115}(43 day); and Sn^{121}(27 hr) *vs* Sn^{123}(130 day) *vs* Sn^{125}(10 day).

As a basis for yield calculations, thermal neutron runs were made by exposing similar uranium foils in a nuclear reactor, followed by radiochemical and counting operations identical with those used for the cyclotron-irradiated samples. Calculation of proton fission yields was then made on the basis of the known yields[18] of the thermal neutron process, by the basic relation of relative yields[19]

$$\left(\frac{Y_N}{Y_{Mo}}\right)_p = \left(\frac{Y_N/Y_{Mo}}{A_N/A_{Mo}}\right)_n \left(\frac{A_N}{A_{Mo}}\right)_p .$$

Here the ratio of the yield of the nuclide N to the yield of molybdenum, in proton fission, is the quantity being determined on the left. The A's are the observed activities, corrected for chemical recovery. The complex parenthesis on the right is a constant for fixed operating conditions; it is evaluated by the thermal neutron runs referred to.

With the exception of the 20-Mev data on natural uranium, each energy was represented by two to four bombardments differing by not more than 0.7 Mev, and the chemical determinations for each of these were performed in duplicate. Of the 183 individual determinations made, the results in 16 instances were rejected on the basis of unexplained extreme deviations. The remaining values gave at 20.0, 18.2, 15.5, and 12.0-Mev energies the yield curves shown in Figs. 2–4, in which the ordinate is the molybdenum-based yield ratio found for each nuclide, plotted against its mass number as abscissa. The average difference of individual values from the mean is indicated for each point if it exceeds the diameter of the circle used. These mean deviations plotted were found to be approximately 0.8 of the standard deviation, as determined by checks made in a number of representative cases. The fit of

TABLE I. Fission product yields, percent.

Mass	Natural uranium Proton energy, Mev 20	18.2	15.5	12	Mass	Uranium-235 Proton energy, Mev 20	15.5	12
75;161	0.02	0.02	0.02	0.01	75;159	0.04	0.03	0.03
77;159	0.09	0.07	0.08	0.05	77;157	0.12	0.09	0.09
79;157	0.23	0.19	0.16	0.13	79;155	0.23	0.18	0.20
81;155	0.39	0.39	0.32	0.28	81;153	0.42	0.34	0.37
83;153	0.62	0.71	0.59	0.51	83;151	0.69	0.61	0.68
85;151	0.95	1.11	1.04	1.04	85;149	1.01	0.92	1.16
87;149	1.43	1.65	1.70	1.71	87;147	1.52	1.37	1.92
89;147	1.94	2.37	2.39	2.38	89;145	1.90	1.97	2.55
91;145	2.50	3.02	3.00	3.32	91;143	2.47	2.51	3.51
93;143	3.01	3.66	3.70	4.04	93;141	3.00	3.03	4.09
95;141	3.52	4.04	4.21	4.66	95;139	3.59	3.47	4.35
97;139	4.07	4.36	4.49	5.02	97;137	4.08	3.89	4.39
99;137	4.63	4.64	4.68	5.18	99;135	4.48	4.18	4.39
101;135	4.63	4.64	4.68	5.13	101;133	4.48	4.18	4.26
103;133	4.54	4.50	4.49	4.77	103;131	4.48	4.06	4.00
105;131	4.44	4.32	4.07	4.04	105;129	4.44	3.72	3.34
107;129	4.12	3.90	3.56	3.11	107;127	4.08	3.26	2.77
109;127	3.66	3.06	3.00	2.23	109;125	3.63	3.20	2.09
111;125	2.73	1.81	1.68	1.14	111;123	2.91	2.13	1.25
113;123	1.13	0.52	0.51	0.47	113;121	1.79	0.68	0.29
115;121	0.64	0.45	0.41	0.32	115;119	0.51	0.31	0.23
117;119	0.62	0.42	0.39	0.31	117;117	0.51	0.31	0.23
118;118	0.61	0.42	0.39	0.31				

[18] Reference 14, Appendix B, p. 2003.
[19] Reference 14, Part V, 1368 (1951).

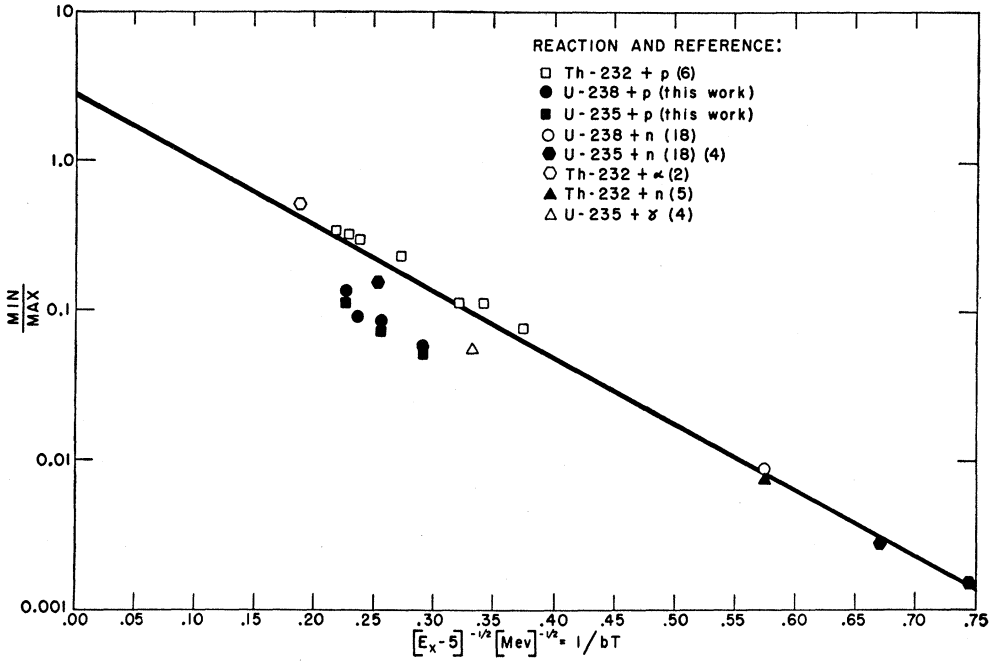

FIG. 5. Fission symmetry as a function of excitation energy.

these points is not always good, as seen, but it is considered that the yield distribution picture at these energies is fairly represented.

In completing these curves the assumptions of symmetric yield and no fine structure were adhered to. Mirror points, though not shown on the completed figures, were equally weighted in their construction, with the curves reflected on mass 118 for natural uranium, and on 117 for U^{235}. With mirror points omitted, partial failure of the above assumptions seems suggested; these assumptions have nevertheless been retained on the basis of inconclusive evidence to the contrary.

For percentage yields, the yield ratios from each of these curves were summed over the complete range from mass 75 to the midpoint. A factor was determined for bringing this total to 100 percent, and each curve was converted from yield ratios to percentage yields by applying its factor. From the resulting curves (not shown), Table I was obtained by reading for the odd mass numbers the percentage yields thus averaged.

A statistical approach to the fission process[8,20] predicts how the relative mass distribution changes with nuclear excitation energy. For interpreting along these lines the observed trend toward symmetric fission at higher energies, the relation plotted in Fig. 5 was formulated. Here the logarithmic ordinate is the minimum-to-maximum yield ratios, as taken from both our work and from other nuclear fission reactions as recorded in the literature. In setting up the abscissa, the quantity (E_x-5) is first evaluated, where E_x is the sum of the bombarding particle energy plus the energy with which it is bound in the compound nucleus. The binding energies are calculated from semiempirical mass tables.[21] The 5 Mev is subtracted as being the energy expended in distorting the nucleus to the point of fission; this value correlates with the γ-ray fission threshold of about 5 Mev. The square root of this (E_x-5) is thus of the nature of nuclear temperature, and the abscissa, with its negative exponent, is proportional to the reciprocal temperature of the distorted nucleus. When the proportionality constant is evaluated on the basis of energy level densities as estimated by Weisskopf,[22] the straight line as drawn corresponds to the relation

$$Y_{\min}/Y_{\max}=2.8\exp[-2.9/T].$$

Since the relative probability of two states differing in energy by an amount ΔE is $\exp[-\Delta E/T]$, it is thus suggested that 2.9 Mev is the additional energy required to produce symmetrical, in preference to asymmetrical fission.

The authors wish to acknowledge the original suggestions and continued interest of J. L. Fowler of this Laboratory. Authors W. H. Jones and A. Timnick are grateful for the opportunity of participating in this cooperative program.

[20] P. Fong, Phys. Rev. **89**, 332 (1953).

[21] N. Metropolis and G. Reitwiesner, U. S. Atomic Energy Commission Report NP-1980, 1950 (unpublished).

[22] V. F. Weisskopf (private communication to J. L. Fowler).