that it will be necessary to include contributions from other fundamental nuclear processes such as compound nucleus formation. At present it is not clear how the compound-nucleus and stripping processes interfere. In the case of (d,t) reaction one would expect a very small contribution from the compound-nucleus process, since in order for this process to contribute to the reaction, it would require the simultaneous appearance of two neutrons and a proton at the nuclear surface, all with sufficient energy to escape.

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Low-Energy Photofission Yields for U^{238}†

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The pattern of mass number dependence of fission yield has been determined in the photofission of natural uranium (U^{238}) by 16-Mev electrons from the linear accelerator. The effective photon energies were principally 9–14 Mev. Yields were determined by quantitative radiochemical isolation and close approximation to absolute β counting for 19 chains. The peak-to-valley ratio is 110, the highest found yet outside of low-energy neutron studies. A spike in fission yields was found near mass number 133. Yields were also obtained for a number of chains in photofission with 10-Mev electrons.

I. INTRODUCTION

As part of an extended study of the pattern of fission yields as a function of energy and nature of the nucleus undergoing fission, a detailed study has been made of the yields in the photofission of natural uranium (essentially the photofission of U^{238}). It is well known that low-energy fission gives principally asymmetric mass division, with peak yields near mass numbers 97 and 138, and a deep valley for masses near symmetric division ($\sim$117). The dependence of the peak-to-valley yields on the energy available to the fissioning nucleus has been the subject of considerable interest. An attempt was made to determine the energy dependence of the very low yields of symmetrical fission using the photons produced by electrons of 10 Mev and 16 Mev impinging on a cylinder of uranium salt. In addition, interest is attached to fission yields in the mass region 131-138 where a fission yield spike occurs, presumably related to closed-shell effects in the primary fission products.[1–4] Photofission yields for masses 132, 133, and 134 are correlated with yield for other fission processes.

II. EXPERIMENTAL PROCEDURE

A. Irradiation Procedure

The M.I.T. linear accelerator[5] accelerates electrons to a maximum energy of about 16 Mev, with those electrons reaching the end of the machine having a spread of energy of about ± 0.6 Mev. The machine terminates, in the target chamber, in a thin aluminum window through which the electrons can be passed essentially unimpeded. On rapid deceleration in a heavy metal target, the electrons give rise to a spectrum of photons ranging in energy from zero to the maximum electron energy. This diverging beam of x-rays is customarily the source of photons for irradiations.

Since the sample irradiated in this study was the salt of a heavy metal, it seemed more conservative of the photons to produce them in the immediate vicinity of the target nuclei by absorbing the electrons directly in the 20-gram uranium salt sample in a cylinder 10 cm long and 1.4 cm in diameter. The fission rate was increased by more than an order of magnitude employing this method over that of placing the sample in a lead-produced photon beam. Such an increase of

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

[2] Wiles, Smith, Horsely, and Thode, Can. J. Phys. **31**, 419 (1953).

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

[4] D. R. Wiles, Ph.D. thesis in Chemistry, Massachusetts Institute of Technology, November, 1953 (unpublished); D. R. Wiles and C. D. Coryell (to be published).

[5] P. T. Demos, Massachusetts Institute of Technology Laboratory for Nuclear Science and Engineering, Technical Report No. 50, May, 1951 (unpublished).

activity was necessary to carry out the study. Either uranyl nitrate hexahydrate or uranyl chloride was used as target. Irradiation times were usually one hour in length.

The linear accelerator was also run at approximately 10 Mev by disconnecting the last three sections. The machine was less reproducible in operation than at 16 Mev, and because of low intensity and low photofission cross section, much smaller amounts of fission products were produced.

Electron-induced fission is considered negligible compared to the fission from the x-rays (bremsstrahlung) produced as the electrons are slowed down.

B. Absence of Appreciable Photoneutron Fission

That neutrons from the photoneutron effect were not contributing to the observed fissions was shown by the following experiments using Si^{31} from the $P^{31}(n,p)$ reaction to show the presence of fast neutrons. Identical "sandwiches," consisting of a small disk of red phosphorus between two disks of uranyl chloride, were irradiated, one in the linear accelerator, one in the fast-neutron beam of the cyclotron (14-Mev deuterons impinging on beryllium), and one in partially thermalized neutrons from the same source (the latter two samples being wrapped well with cadmium foil to minimize thermal fission of U^{235}). After irradiation, the phosphorus samples were counted directly, while the uranium samples were analyzed radiochemically for Ba^{139}. If the fast neutron spectrum for the cyclotron is the same as that from the $U^{238}(\gamma,n)U^{237}$ reaction at the linear accelerator, the activity induced in the P^{31} samples should be comparable after normalizing to a given fission rate by means of the Ba^{139} activities. Since the fast-neutron spectrum of the cyclotron probably contained many more neutrons of high energy than did the linear accelerator neutron spectrum, the cyclotron spectrum was changed drastically by surrounding the sample with 10 cm of paraffin, in order to bracket the actual spectrum. Table I summarizes the data from the experiments.

From the ratios of activities of Si^{31} to Ba^{139} it appears that very little Si^{31} activity was produced from $U^{238}\times(\gamma,n)U^{237}$ neutrons, and consequently few fissions (less than 0.8 percent) could be caused by neutrons from this source.

C. Analytical and Counting Procedure

After irradiation, the uranium samples were dissolved in a suitable quantity of water, and aliquots were analyzed radiochemically for the various fission products. Standard analytical procedures[6] were modified when necessary to provide for the large quantities of uranyl ion present. Barium analyses were performed on an aliquot of each irradiated sample in order to normalize the yields to that of 85-min Ba^{139} and to compare the individual irradiations.

TABLE I. Comparison of fast neutron effects with fission.

Irradiation	Ratio Si^{31}/Ba^{139} (counts/sec)
Cyclotron, fast neutrons	1680/1035 = 1.6
Cyclotron, slower neutrons	42.0/32.0 = 1.3
Linear accelerator	50/5500 = 0.009
Limit for photoneutron fission	<0.009/1.3

TABLE II. Summary of high-energy fission yield experiments (x-rays up to 16 Mev).

Nuclide	Fission yield, %	Reliability
2.40-hr Br^{83}	0.288	±0.056
31-min Br^{84}	0.511	±0.10
53-day Sr^{89}	3.67	±0.60
9.7-hr Sr^{91}	4.22	±0.48
2.7-hr Sr^{92}	3.46	±0.35
10.0-hr Y^{93}	5.29	±0.36
17.0-hr Zr^{97}	6.31	±0.23
68.1-hr Mo^{99}	6.06	±0.16
4.5-hr Ru^{105}	3.61	±0.17
13.0-hr Pd^{109}	0.224	±0.04
21.0-hr Pd^{112}	0.110	±0.01
5.0-hr Ag^{113}	0.0627	±0.0041
21-min Ag^{115}	0.0522	±0.0036
8.00-day I^{131}	4.43	±0.38
77-hr Te^{132}	5.78	±0.40
22.4-hr I^{133}	7.06	±0.40
85.0-min Ba^{139}	5.97[a]	...
12.8-day Ba^{140}	5.77	±0.33
33.0-hr Ce^{143}	5.32	±0.34

[a] Reference value to give integral of the yield-mass curve of 200 percent.

Final precipitates were filtered on tared filter papers, weighed, and then mounted on Scotch tape for counting. Victoreen Geiger-Mueller counters with thin mica end windows were used, and all counting was done at second shelf geometry.[7]

In order to correct the observed counting rates for absorption in the sample covering, air, and tube window, and for the many kinds of scattering exhibited by β rays, the data of Zumwalt[8] and Engelkemeir and co-workers[9] were used. It is estimated that the corrected activities are reliable to within ±10 percent with the use of such corrections. The corrected activities were computed over to saturation activities taking into account the chain relations, irradiation and decay times, and chemical yield, and then expressed in terms of the saturation activity of 85-min Ba^{139}.

III. RESULTS

The saturation activities are directly proportional to the fission yields. The spread of masses (83 to 143) is great enough that the data can be used to estimate the normalizing constant necessary to give summation of the yield-mass curve to 200 percent. To define the mass

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

[7] See reference 6, Introduction to Part I, p. 4.

[8] L. R. Zumwalt, Oak Ridge National Laboratory Unclassified Report Mon C-397, September 14, 1949 (unpublished).

[9] Engelkemeir, Seiler, Steinberg, and Winsberg, reference 6, Paper 4.

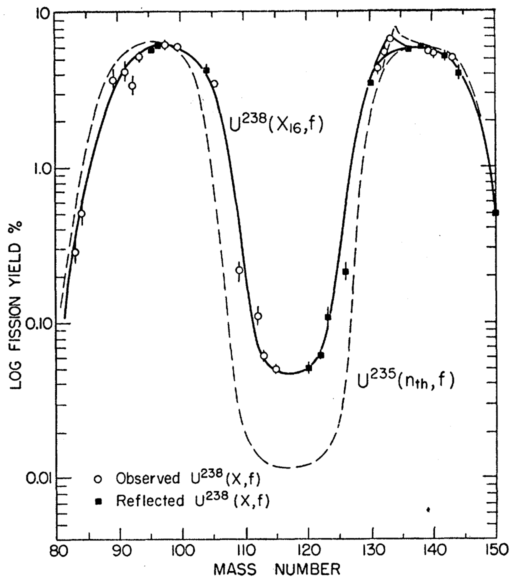

FIG. 1. Yield-mass curve for the fission of U^{238} by photons of 16-Mev maximum energy. Curve for thermal fission of U^{235} for comparison.

yield curve more sharply for this purpose, it was convenient to assume arbitrarily that an average of 3 neutrons is emitted per fission, so that for the relative yield of each mass number A, a reflected point could be plotted of the same yield at mass number 238-A-3. Normalization to 200 percent total yield was achieved by assigning 85-min Ba^{139} the fission yield of 5.97 percent for the 16-Mev experiments and the yield of 5.87 percent for the 10-Mev experiments.

Table II and Fig. 1 summarize the experimental yields of 19 species from the fission of U^{238} by x-rays of maximum energy approximately 16 Mev. The values reported represent an average of at least three individual determinations of each species. In the last column of the table is an estimate (the standard deviation) of the reliability of the separate fission yields. For reference purposes, the smoothed, yield-mass curve for thermal fission of U^{235} taken from Manhattan Project data[6] is also plotted in Fig. 1 as a broken line.

Intensities were not high enough to get the yield of 2.3-day Cd^{115}. Experiments were directed to 3-hr Cd^{117} but trouble was encountered with a 40-min isomer and isomeric indium daughters,[10] so useful yield data were not obtained.

Table III presents the experimental yields of 13 species from the fission of U^{238} by x-rays of maximum energy approximately 10 Mev. The estimated errors of these yields are somewhat greater than those of the higher energy irradiations because of the very low activity obtained from the samples. Those yields for which no estimate of error is given were single observations.

[10] Coryell, Lévêque, and Richter, Phys. Rev. **89**, 903 (1953).

It was of interest to establish the distribution of photon energies producing the fissions. This can be obtained as the product of the photon spectrum $N(E)$ and the fission cross-section function $\sigma(E)$. The thin-target photon energy distribution function of Schiff[11] is used as a starting point. Production of x-rays by degraded electrons in the target leads to a considerable softening of the spectrum from the thin target spectrum. Dr. I. Halpern helped us treat the case of axial incidence of an electron beam on a narrow cylinder, for which it was shown that the correction factor on a $1/E$ spectrum is $1+\ln[7(E_0-E)/4]$, where (1/7) g/cm² is the target thickness before electrons and thus potential hard x-rays are lost from the cylinder, and 4 Mev cm²/g is the effective stopping power of the uranium salt. It is assumed that the Schiff spectrum should be corrected by the same factor.

Figure 2 shows in curve A (broken line) the Schiff thin-target spectrum for 16-Mev electrons, in arbitrary units. Curve B is the spectrum corrected to cylindrical thick target by the factor given above. Curve C is the fission cross-section function determined by Ogle and McElhinney[12] for x-rays on U^{238}. Curve D (again in arbitrary units) is the product of curves B and C and represents the relative number of fissions caused by photons of energy E. It is seen that there is a broad maximum from 9 to 14 Mev, and the effective x-ray energy might be called 12 Mev.

A similar treatment cannot be applied easily to the data for 10-Mev electrons because of the poorer control of the beam energy and the closeness to the threshold of the fission reaction. The effective energy is certainly below 10 Mev, and perhaps about 8.5 Mev.

IV. DISCUSSION

The most prominent feature of the curve for the fission of U^{238} by x-rays up to 16 Mev (Fig. 1) is the fact that the valley for symmetrical fission has risen only about sixfold over the very low value observed for

TABLE III. Summary of low-energy fission yield experiments (x-rays up to 10 Mev).

Nuclide	Fission yields, %	Reliability
2.40-hr Br^{83}	0.300	±0.045
31-min Br^{84}	0.411	±0.074
9.7-hr Sr^{91}	4.44	
17.0-hr Zr^{97}	5.11	
68.1-hr Mo^{99}	4.94	
13.0-hr Pd^{109}	0.0854	±0.020
21.0-hr Pd^{112}	0.042	±0.002
8.00-day I^{131}	3.76	±0.16
77-hr Te^{132}	5.58	±0.80
22.4-hr I^{133}	6.80	±0.60
85.0-min Ba^{139}	5.87[a]	...
12.8-day Ba^{140}	5.77	±0.47
33.0-hr Ce^{143}	5.94	

[a] Reference value to give integral of the yield-mass curve of 200 percent.

[11] L. I. Schiff, Phys. Rev. **83**, 252 (1951).
[12] W. E. Ogle and J. McElhinney, Phys. Rev. **81**, 344 (1951).

thermal fission of U^{235}. Indeed, the ratio of the peak-to-valley yields is 110 (6.3/0.052) in $U^{238}(x_{16},f)$ compared to 600 in $U^{235}(n_{th},f)$. Low intensities precluded determination of the yields of silver and cadmium activities in the experiments with low energy (up to 10 Mev) x-rays, but Table III shows that the yields of Pd^{109} and Pd^{112} are lower by a factor of 2.5 than for the higher-energy x-rays.

Previous work on photofission has taken place only at higher beam energies. Spence[13] reports a peak-to-valley ratio of $\sim$24 for U^{235} irradiated with x-rays from a betatron operating at 22-Mev maximum energy. Schmitt and Sugarman[14] obtained a peak-to-valley ratio, defined by 17-hr $Zr^{97}/(2.3\text{-day } Cd^{115m}+43\text{-day } Cd^{115})$, of 20 for U^{238} in the x-ray beam of a betatron operating at 22 Mev, a ratio of 10 at 48 Mev, and a ratio of 7 at 100 Mev. The peak-to-valley ratio of 110 in this work lies far above any simple extrapolation of the betatron data to lower energies. It appears, therefore, that an increase in yield of symmetrical fission sets in just above 16 Mev, a small amount of which lowers the ratio greatly. Cross-section measurements[12,15] show that most of the photofission for bremsstrahlung of maximum energy above 20 Mev occurs in the giant resonance of about 18 Mev. Schmitt and Sugarman[14] have pointed out that a high-energy cross-section tail extends above 48 Mev, giving symmetric fission.

Turkevich and co-workers[16] have presented a rough empirical correlation of peak-to-valley ratio against the excitation of the compound nucleus undergoing fission, irrespective of the nature of this nucleus. The ratio for low-energy photofission of U^{238} does not fit well on this curve.

There is considerable interest in fine structure in fission yields, in particular in the spike around mass 134 discovered in thermal neutron fission by Thode and Graham,[17] and its change with fissioning nucleus and energy. Two sources are credited for this spike: enhanced neutron emission[1,3] from primary fission fragments with a few neutrons (odd number) above 82, and *a priori* selectivity[2–4,18] in fission for closed shell structures (nuclei with 82 neutrons or 50 protons). The analysis of Pappas[3] indicates that for thermal neutron fission of U^{235}, the two sources contribute approximately equally. It is generally considered that increasing the energy of fission should lower the selectivity for closed-shell structures.

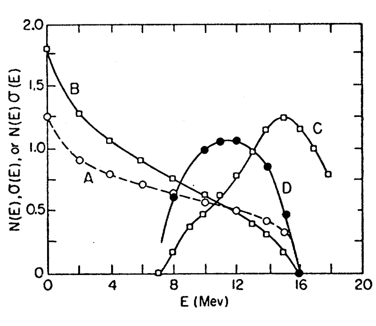

FIG. 2. Photon intensity, cross section, and photofission yield, dependence on energy. *A*, Schiff thin-target spectrum; *B*, photon spectrum corrected for target shape (see text); *C*, Ogle-McElhinney cross-section curve; *D*, product of *B* and *C*, representing photofission energy distribution.

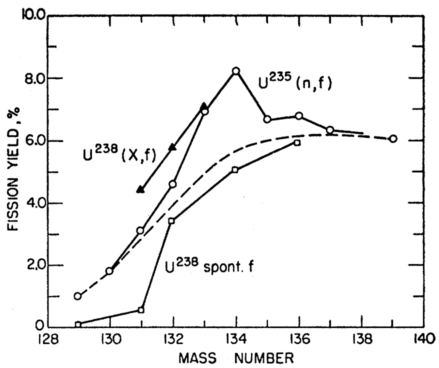

FIG. 3. Comparisons of fine structure in fission yields for different types of fission. Broken line, hypothetical smooth curve for $U^{235}(n,f)$.

Evidence for the spike is still present in the photofission data for 8.0-day I^{131}, 77-hr Te^{132}, and 22.4-hr I^{133} as shown in Fig. 1, and exhibited in more detail in Fig. 3, in comparison with other results. The broken line in Fig. 3 is the smoother curve for U^{235} thermal neutron fission,[6] a hypothetical shell-independent line. The circles represent mass-spectrometric data of Thode and co-workers[2] for xenon and cesium thermal fission products of U^{235}, normalized to 3.1 percent for chain 131, with Ba^{139} added. The squares represent Wetherill's data[19] for spontaneous fission of U^{238} based on a yield of 6.0 percent for Xe^{136}. The triangles represent the data from this work.

The spontaneous fission reaction represents tunnelling through the barrier without excess energy, and shows narrower peaks. A pronounced shoulder is apparent at mass 132. The curve for $U^{235}(n,f)$ shows the spike at mass 134. The more fragmentary data for photofission show that a spike occurs for it at or beyond mass 133, and in addition, that the peaks are wider, as seen also from Fig. 1. Data for the complimentary peak around mass number 101 would be helpful in determining how much of the selectivity effect remains.

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[13] R. W. Spence, Unclassified Brookhaven National Laboratory Conference Report BNL-C-9, July, 1949 (unpublished).
[14] R. A. Schmitt and N. Sugarman, Phys. Rev. **89**, 1155 (1953).
[15] G. C. Baldwin and G. S. Klaiber, Phys. Rev. **71**, 3 (1947).
[16] Turkevich, Niday, and Tompkins, Phys. Rev. **89**, 553 (1953).
[17] H. G. Thode and R. L. Graham, Can. J. Research **A25**, 1 (1947).
[18] Glendenin, Steinberg, Inghram, and Hess, Phys. Rev. **84**, 860 (1951).
[19] G. W. Wetherill, Phys. Rev. **92**, 907 (1953).