Letters to the Editor

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High Energy Spallation and Fission Products of Uranium

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WE have bombarded natural uranium with 380-Mev helium ions in the Berkeley 184-inch cyclotron in order to study the distribution in yield to the radioactive products. After each of the irradiations, amounting to some 25 in number, the metallic uranium (2-cm×0.5-cm area and 0.1-cm thickness) was dissolved, and the yield in weighed chemical reactions of each of several isotopes was determined by means of counting experiments using a thin-window, bell-jar type Geiger-Mueller counter under conditions of known counting efficiency. Corrections were made for self-absorption, and for absorption by air and the mica window; back-scattering was insignificant in comparison with other factors. Conventional alpha-particle counters and an alpha-pulse analyzer[1] were used for the alpha-emitters. The yield for each isotope was determined relative to a standard isotope, the 12.8-day Ba^{140}, in order to compensate for the day-to-day variation in intensity of the helium ion beam. The results show what appears to be a continuous yield of radioactive products for the entire range of elements from the uranium region down to elements in the vicinity of atomic number 25. A plot of relative yield *vs.* the mass number of the radioactive products is shown in Fig. 1. The total cross section for the fission reaction (estimated from the area under the fission part of the curve) amounts to about 2×10^{-24} cm², but this value must be considered to be a very rough one, because of the unknown intensity of the helium ion beam. The errors indicated on this curve are estimated probable errors, arising from counting statistics, chemical operations, and correction factors applied.

There are several points of interest in connection with this distribution curve. The yield curve probably results from two essentially different types of reactions. Spallation reactions probably account for the products from the region of uranium down to those in the neighborhood of mass number 180–200, while the remainder of the curve is accounted for by the fission process. Isotopes with mass numbers 180–200 are more likely spallation products than fission products, and the small differences in yield in this region are not significant because of the low levels of radioactivity produced. Not shown on the curve are a number of alpha-emitting isotopes with mass number in the neighborhood of 210 which were observed to be present in relatively high yield; these are believed to be products of alpha-emitting radioactive chains[2] originating from heavier isotopes which are found in higher yield as can be seen from the curve.

The distribution of fission products seems to show only a single peak in the region of maximum yield. These results are of course very much in contrast with the yield curve for slow neutron fission of U^{235} with its very characteristic two peaks and deep valley in between.[3] This is not unexpected in view of the previously observed increased relative yield of fission products resulting from symmetrical cleavage as the energy of the bombarding particle is increased.[4–7] This single maximum is somewhat similar to that observed by Goeckermann and Perlman[8] in the fission of bismuth with 190-Mev deuterons, although the maximum, of course, occurs at a higher mass number. The data indicate, but are not sufficiently accurate to prove, that the maximum in the curve occurs at a mass number definitely less than one-half that of the initial compound nucleus formed in the reaction as is the case for bismuth fission, and thus it cannot be said with certainty whether fission is preceded by the emission of a number of neutrons as in the mechanism proposed by Goeckermann and Perlman. Since uranium can undergo fission as the result of small excitation energy, it is probable that some

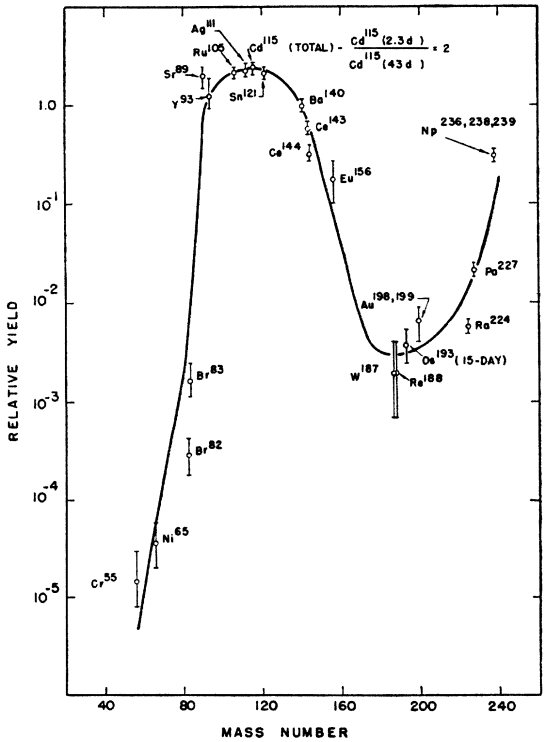

FIG. 1. Relative yield curve for products from 380-Mev helium ion bombardment of uranium.

of the fission results from reactions in which only a fraction of the total energy of the helium ion is utilized to form a fissioning compound nucleus of mass around 240, a mechanism similar to that of slow neutron fission results. Thus a number of mechanisms may prevail ranging from this one to that in which a number of neutrons are emitted before the fission process, giving rise to symmetrical cleavage as the most probable process, with the latter mechanism perhaps predominating in view of the shape of the distribution curve.

The fission yield curve definitely extends to higher and lower mass numbers than is the case for the fission of uranium with lower energy particles. At the low energy end the products down to the region of atomic number about 27 seem definitely to be fission products, but the source of the $_{24}Cr^{55}$ is doubtful in that the uranium may have contained sufficient iron impurity to account for the small yield of this isotope. It may be noted that if there are fission products with mass number as low as about 55 it should be expected that there be heavy fission products extending up to about mass number 180, and hence joining with the region of spallation products.

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[1] See, e.g., A. Ghiorso, A. H. Jaffey, and H. P. Robinson, "An alpha-pulse analyzer apparatus," Plutonium Project Record, **14B**, 17.3 (1948) (to be issued).
[2] A. Ghiorso, W. W. Meinke, and G. T. Seaborg, Phys. Rev. **74**, 695 (1948).
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[6] A. S. Newton, Phys. Rev. (in press).
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[8] R. H. Goeckermann and I. Perlman, Phys. Rev. **73**, 1127 (1948).

Cross Section for the Reaction $C^{12}(\gamma,n)C^{11}$

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PURE graphite has been irradiated by x-rays from the betatron operating at 50 and 98 Mev at measured quantum intensities. The number of (γ,n) processes occurring per unit time was determined by observation of the C^{11} activity with a calibrated Geiger-Müller counter.

The quantum intensity was determined for both 50- and 98-Mev betatron operation first by calibrating an ionization chamber monitor at low level with an absolute x-ray spectrum analyzer designed by one of the authors,[1] and second by a careful measurement of the relative ion chamber response from the low level operation to the high levels used in the C^{12} bombardment. While this second calibration step seems simple and straightforward, it was found somewhat difficult because of the large intensity range covered (about 10^5), the desired accuracy (about 5 percent), and the fluctuating nature of the betatron x-ray beam. The spectrum analyzer, used to calibrate the ion chamber at low level in terms of the absolute quantum intensity, counts electron-positron pairs ejected from a thin target of chosen material and weight. This analyzer first uses a collimator which defines a beam aperture and, by means of a magnetic field region, clears the beam of charged particles. A pair-forming target, magnetic field, and Geiger-counter array then serves to register electron and positron events; finally, an analyzing circuit registers electron-positron events of given total energy, which is, therefore, essentially the pair spectrum. This instrument at present registers pairs in energy channels 15 Mev in width; the number of quanta in each energy channel is obtained from the pair data, the pair-forming target thickness, and the cross section for pair production. This last cross section is now known theoretically and experimentally.[1]

The spectrographic graphite powder irradiated was held in an aluminum mount in a layer of uniform thickness and known area. The mount was accurately positioned so that the graphite layer subtended exactly the same part of the beam subtended by the pair radiator.

After irradiation for a known time at nearly constant and known intensity, all the powder was formed into a pellet of standard geometry; the subsequent decay of the 20-minute C^{11} activity was followed. The geometry, counting arrangements, and correction methods for absorption, self-absorption, and back-scattering were the same as those used for studies of relative yields x-ray induced nuclear reactions.[2] After the counting measurements were completed, the graphite was weighed and the target thickness calculated. The activity was calculated to saturation bombardment, and it was corrected for the geometrical efficiency of the end-window counter. This efficiency (34 percent of the total solid angle) was determined with the aid of a thin weighed U_3O_8 deposit—the UX_2 beta-rays of which were counted through an absorber sufficiently thick to remove the soft UX_1 radiations.

Results of the measurements are shown in Table I. The numbers in column II are calculated by dividing the number of pairs per minute in the 15-Mev wide band (centered at 30 Mev) by the width, and then dividing by the cross section for pair production at 30 Mev and also by the pair-forming target thickness. The quantity, σ, shown in the fourth column is essentially the ratio of column III to column II and represents the product of cross section by the level width of the γ-n reaction,

TABLE I.

I X-ray energy (Mev)	II No. of quanta/ Mev minute (at 30 Mev)	III No. of (γ,n) processes/min. mg atom cm^{-2}	IV $\sigma=\frac{\text{III}}{\text{II}\times 6.02\times 10^{20}}$
98	4.74×10^{7}	4220	1.48×10^{-25} Mev cm^2
98	4.74×10^{7}	4220	1.48×10^{-25} Mev cm^2
50	1.24×10^{7}	1090	1.46×10^{-25} Mev cm^2