Low-Energy Charged-Particle-Induced Fission*

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The excitation functions for the proton and deuteron induced fission of Th^{232} and U^{238} have been determined in the energy range of 3–12 MeV. The excitation functions were measured using direct counting with p,n junction counters, gross counting with a recoil catching technique, and radiochemical measurement of certain fission product nuclides. In addition, the ratio of the peak-to-valley values of the fission-fragment kinetic-energy spectrum were determined as a function of bombarding energy. Breaks in the ratio curve at certain bombarding energies are interpreted in terms of competition between fission and nucleon evaporation.

INTRODUCTION

THE papers of Tewes and James,[1] Butler,[2] Jones,[3] and Foreman[4] comprise the bulk of the experimental information which is available on the excitation functions and on the ratio of symmetric to asymmmetric fission in the field of low-energy charged-particle fission. Tewes and James studied, by radiochemical means, the fission of Th^{232} induced by protons which ranged in energy from 6.7 to 21.1 MeV. They characterized the excitation functions for both fission and the (p,xn) reactions. In addition to this, the ratio of the yield of Cd^{115} to that of Sr^{89}, as a function of bombarding energy, was measured. Since Cd^{115} is produced by symmetric fission, whereas Sr^{89} is formed from asymmetric fission, this ratio gives an indication of the relative amounts of symmetric and asymmetric fission. The findings of Butler and his co-workers, derived from a careful radiochemical study of Th^{232}, U^{238}, and Pu^{239} fissioned by proton bombardment, exhibited features which were not seen in the earlier work. A plot of the ratio of symmetric to asymmetric fission versus energy showed a number of maxima, while the ratio plotted by Tewes and James was a smoothly increasing function of energy. The maxima in Butler's plot were attributed to the onset of various (p,xnf) reactions. It was suggested that as the bombarding energy of the protons increased above the threshold for the (p,f) and (p,n) reactions, the fission resulted from only one reaction, (p,f). Symmetric fission becomes more probable with increasing excitation energy, thereby increasing the ratio. At some point the residual nuclei from the (p,n) reaction retained enough energy to fission. Such fissions from the (p,nf) reaction occured at a lower excitation energy than those from the (p,f) reaction at the same proton energy. As a consequence, the ratio of symmetric fission decreased. With further increase in proton energy the ratio again begins to rise.

Jones and his co-workers have presented mass-yield curves for the fission of U^{235} and U^{238} induced by protons in the energy range from 12 to 20 MeV. Foreman and his co-workers have reported the mass-yield curves and the excitation functions for the fission of Th^{232} by 15- to 43-MeV α particles and for the fission of U^{238} with 9 to 22-MeV deuterons.

All these earlier studies used cyclotron beams with a relatively large inherent spread in the particle energies. It seemed worthwhile to restudy low-energy charged-particle fission with the precisely defined energy beam from a Van de Graaff accelerator and with solid-state junction counters.

EXPERIMENTAL

The experiments described in this paper were performed using the Florida State University tandem Van de Graaff accelerator. The energy of the proton or deuteron beam is continuously variable from 3–12 MeV. The energy definition of the beam is 2–3 keV.[5]

Relative excitation functions were measured for U^{238} and Th^{232} fissioned by protons by a recoil catching technique. The normal "back-to-back" catching arrangement could not be used for bombarding energies below 6 MeV; the proton activation in the catcher was on the same order, or larger, than that of the fission product activity caught on the foil. It was also necessary in the recoil catching technique used to provide a large solid angle of detection to achieve maximum sensitivity for the low yields below 6 MeV. The arrangement which was used consisted of a cylindrical catching foil made of 0.001-in.-aluminum foil placed at backward angles to the beam. The beam entered along the axis of the cylinder and was not allowed to strike the catcher. The recoil products on the catcher were considered to come from the first 10 mg/cm^2 (approximate average range of fission fragments in uranium[6]) of a thick target (0.001 in.), and the energy of the protons giving the fragments on the catching foil was taken to be the

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[3] W. R. Jones, A. Timmick, J. H. Paehler, and T. H. Handley, Phys. Rev. **99**, 184 (1955).
[4] B. M. Foreman, W. M. Gibson, R. A. Glass, and G. T. Seaborg, Phys. Rev. **116**, 382 (1959).
[5] J. Nelson (private communication).
[6] J. B. Niday, Phys. Rev. **121**, 1471 (1960).

mean proton energy in this 10 mg/cm^2. The gross amount of activity on the foil was determined using windowless flow-proportional counting (for betas) and NaI scintillation counting (for gammas). A plot of the gross activity as a function of proton energy yielded the relative-excitation function. To determine whether or not neutrons were responsible for the fission observed at the lowest energies, targets were placed behind aluminum absorbers sufficient to stop the proton beam. No evidence of fission could be found in these targets.

Solid-state *p,n* junction counters were used for fission-fragment counting during bombardment to measure relative-excitation functions for U^{238} and Th^{232} fission by protons and dueterons. For these measurements 50-Ω-cm junction counters were prepared.[7] The usefulness of the junction counter method was limited to energies above 6 MeV, since the very low cross sections below that energy gave too low a counting rate.

The pulses from the junction counter were amplified and then analyzed on the Technical Measurement Corporation 256-channel analyzer.

For the system $U^{238}+p$, the absolute cross sections were determined at 9.25 and 11.45 MeV by standard radiochemical means. The nuclides whose fission yields were determined were: Nb^{95},[8] Zr^{97},[8,11] Mo^{99},[8] Te^{132},[8] Pd^{109},[9] Cd^{115},[10] Ba^{140},[9] I^{131},[11] I^{133},[11] I^{135},[11] Ru^{105},[12] and Br^{82}.[11] From these yields plus the yields of the complementary fission products, a mass yield curve was constructed from which the fission cross section was calculated. The relative-excitation function curve was normalized to these absolute values. For $Th^{232}+p$, $Th^{232}+d$, and $U^{238}+d$, the relative-excitation functions were normalized to the data of Tewes and James[1] and Huizenga.[13]

The experimental arrangement used for the measurement of the ratio of asymmetric to symmetric fission consisted of a 50-Ω-cm *p,n* junction counter placed at right angles to the beam. The output pulses from the counter were amplified and then analyzed in a Technical Measurement Corporation 256-channel analyzer. From the graph of the count-rate versus energy, the ratio of the height of the high-energy peak to that of the valley

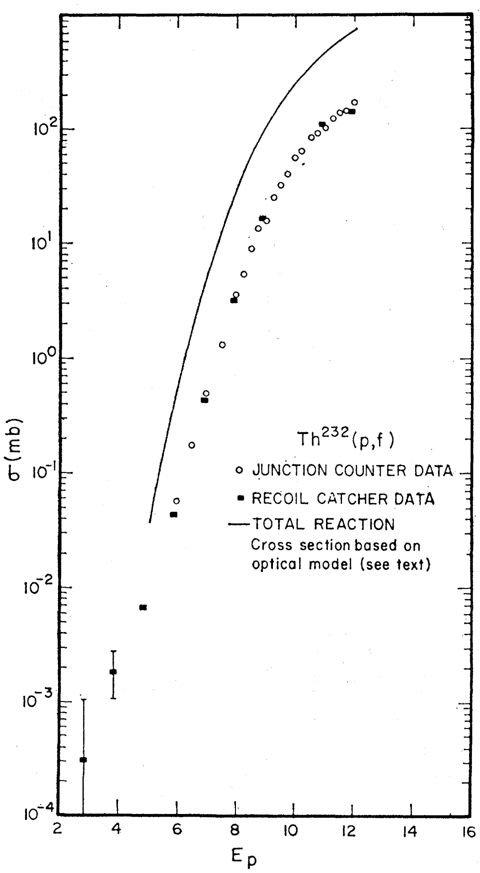

FIG. 1. $Th^{232}(p,f)$ excitation function.

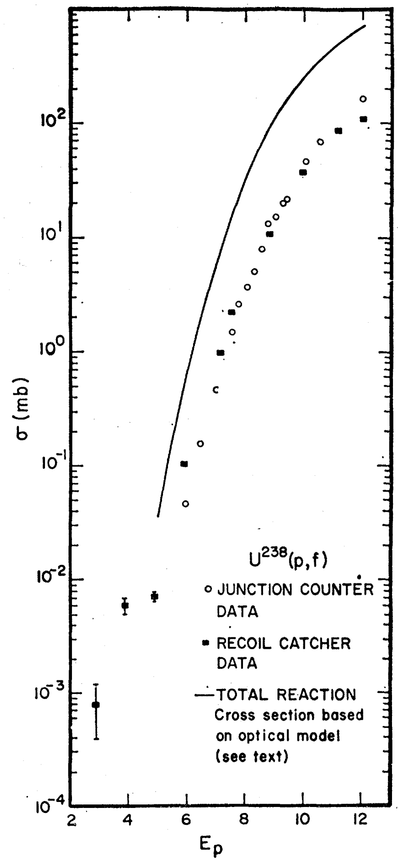

FIG. 2. $U^{238}(p,f)$ excitation function.

[7] P. F. Donovan [private communication (Notes by P. B. Weiss)].

[8] E. M. Scadden and N. E. Ballou, National Academy of Sciences, NAS-NC-3009 (1960).

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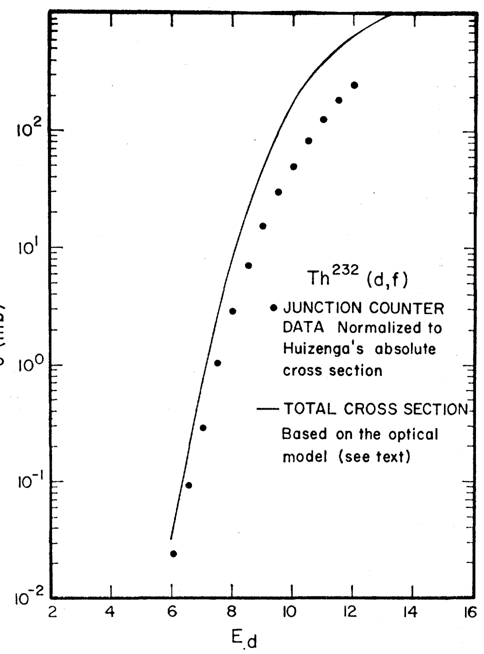

FIG. 3. $Th^{232}(d,f)$ excitation function.

was determined. The targets which were used consisted of either uranium tetrafluoride or thorium tetrachloride which had been vaporized onto thin (0.00025 in.) aluminum foil. Their thickness ranged from 40 to 200 $\mu g/cm^2$. The targets were bombarded with protons from the tandem Van de Graaff at energies 250 keV apart from 6 to 12 MeV.

RESULTS AND DISCUSSION

Shown in Figs. 1–4 are the fission excitation functions for the reactions $Th^{232}+p$, $U^{238}+p$, $Th^{232}+d$, and $U^{238}+d$, respectively, compared to calculated curves for the total reaction cross section. The open circles represent data taken from p,n junction counter measurements, and the closed circles, from the recoil catching data. The smooth curves, shown here for comparison, are the total reaction cross sections for protons and deuterons on U^{238} computed using an optical model nuclear potential of the form

$$V=\frac{U+iW}{1+e^{(r-R)/a}},$$

together with a Coulomb potential assumed to be that given by a sphere of uniform charge density and radius R. The parameters used in the calculation were: $U=-48.5$ MeV; $W=-9.00$ MeV; $a=0.63$ F; and $R=9.4$ F. The calculation was carried out using the "ABACUS 2" program.[14]

A typical fission-fragment kinetic-energy spectrum is shown in Fig. 5. The energies of the fragments corresponding to the peaks are approximately 65 and 90 MeV. The ratio of the counting rate at the high-energy peak to that in the valley is plotted as a function of energy in Fig. 6 for the system $U^{238}+p$. The error limits shown in Fig. 6 were calculated from the statistical counting error in both of the counting rates from which the ratio, was determined. Several runs were made and consistent results were obtained.

Figure 6 exhibits two significant features. First, a larger break is observed in the curve which may be attributed to the onset of second chance fission, i.e., fission after a neutron has been emitted from the compound nucleus. The location of the major break is in agreement with the work of Butler[2] as can be seen from the following summary:

Major break		Minor break	
Butler	This work	Butler	This work
6.5±0.5 MeV	6.8±0.1 MeV	None	8.35±0.1 MeV 10.6 ±0.1 MeV[15]

Secondly, two smaller breaks are observed in the curve. The position of these breaks are noted in the above summary. Such smaller variations in the ratio may be explained qualitatively in the following manner: Symmetric fission requires a higher energy than asymmetric fission, as can be seen from increased preference for symmetric fission at higher bombarding energies. Therefore, it seems reasonable to assume that the onset

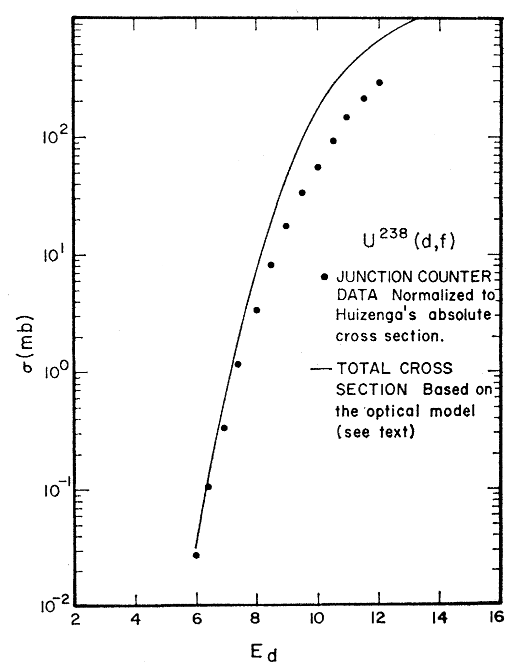

FIG. 4. $U^{238}(d,f)$ excitation function.

[14] The authors wish to thank Dr. Elliot Auerbach of the Nuclear Engineering Department, Brookhaven National Laboratory, for his generous assistance in this calculation.

[15] This break has subsequently been seen in a radiochemical peak-to-valley determination. [E. F. Meyer (private communication)].

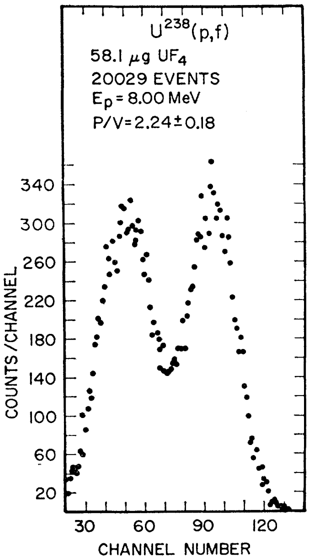

FIG. 5. Typical fission-fragment pulse-height spectrum.

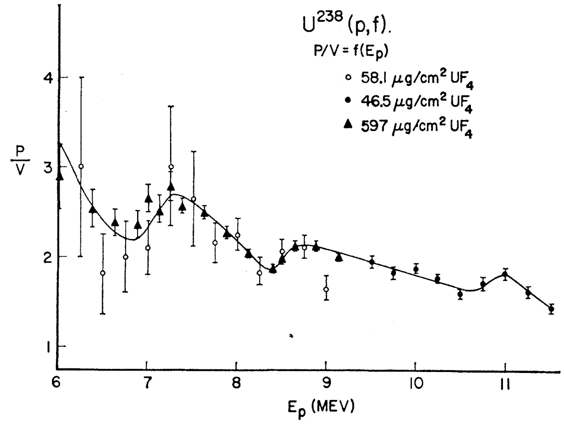

FIG. 6. High-energy peak-to-valley ratio as a function of proton energy for $U^{238}(p,f)$.

of any exit channel from the compound system will compete predominantly with symmetric fission. Between 6.8 and 8.3 MeV, the principal competitive reactions are (p,n), (p,f), and (p,nf). The neutron may be considered to be always emitted with a certain average value of the kinetic energy and, when fission subsequently occurs, the ratio of asymmetric to symmetric fission is expected to decrease gradually. However, when the $(p,2n)$ reaction first becomes a major competitor with the (p,nf) reaction, the second neutron is emitted most probably in those cases where the kinetic energy of the first neutron was lower than the average value. Consequently, the emission of this second neutron competes predominantly in the case where symmetric fission would be more important. A similar line of reasoning may be applied at any other threshold.

If the threshold for the $(p,2n)$ reaction is calculated[16] for U^{238}, and the most probable kinetic energy (1.1 MeV)[17] of the two neutrons emitted is added to this, the value is 9.03 MeV $(Q+K.E._1+K.E._2=6.83+1.1+1.1)$. This is roughly in agreement with the first small break observed. There are two possible reaction thresholds which might be responsible for the second small breaks: Those for the $(p,p'n)$ and the $(p,3n)$ reactions. The Q values for these reactions are 6.00 and 13.55 MeV. Because of the high-Q value, it seems reasonable to discard the $(p,3n)$ reaction as a possibility. In order for this break to be due to the $(p,p'n)$ threshold, the proton would have to carry off 3.5 MeV $(10.6-6.0-1.1)$. This is a little larger than three times the neutron energy, which is not unreasonable since proton emission is hindered by the Coulomb barrier. Thus, it is likely that the $(p,p'n)$ threshold is responsible for this break. Additional work on the $Th^{232}+p$ reaction indicates the same behavior. There the minor breaks seem to occur at 9.5 and 11.25 MeV. The (p,nf) threshold energy (7.7±0.1 MeV) obtained in this manner is in good agreement with the value obtained by Butler (8.0±0.5 MeV). Additional work is in progress on Th^{232}.

ACKNOWLEDGMENTS

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[16] B. M. Foreman, UCRL-8223, 1958 (unpublished).
[17] J. M. B. Lang and K. T. LeCouteur, Proc. Phys. Soc. (London) **A67**, 586 (1954).