Fission and Total Reaction Cross Sections for 22-Mev Protons on Th^{232}, U^{235}, and U^{238}

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Absolute excitation functions for (p,f) reactions in Th^{232}, U^{235}, and U^{238} were measured by radiochemical techniques; at 21.5 Mev, the cross sections are 0.83, 1.31, and 1.28 barns, respectively (±15 percent in each case). Cross sections for (p,xn) reactions in U^{238} were found to be quite small. The total reaction cross sections correspond to a nuclear radius of $(1.55\pm0.1)A^{\frac{1}{3}}\times10^{-13}$ cm. Some advantages of determining the nuclear radius by total reaction cross sections are pointed out, and the discrepancy between electromagnetic and nuclear determinations of the nuclear radius is discussed.

MEASUREMENTS of cross sections for neutron-induced fission on various target isotopes and for various incident neutron energies are widely scattered through the literature. Excitation functions (i.e., cross sections as a function of energy) for charged-particle-induced fission have been measured by Jungerman[1] and by Jungerman and Wright[2] using alpha particles and deuterons. Tewes and James[3] have measured the Th(p,f) excitation function, though not on an absolute scale. No absolute cross sections have been reported, however, for proton-induced fission.

Fission cross sections in the medium energy region are especially interesting because, as will be shown, they are essentially a measure of the total reaction cross section, a quantity which, while of great importance in nuclear reaction theory, is generally difficult to measure. This quantity is of more basic importance when determined for protons than for deuterons or alphas, since, in the latter cases, the interpretation is complicated by the structure of the incident particle.

In addition to the fact that excitation functions for (p,f) reactions are of considerable interest, the experimental problems in carrying out the measurements have been greatly simplified by the recent studies of fission mass distributions from proton-induced fission.[3–6] As a result of this work, fission cross sections can be measured by determinations of the radiochemical yield of a single fission product. Measurements of excitation functions for proton-induced fission in Th^{232}, U^{238}, and U^{235} using the internal, circulating 22-Mev proton beam of the ORNL 86-inch cyclotron were therefore undertaken.

The experimental methods used for measurements of excitation functions have been described previously.[7] The energy of the incident protons was lower (21.9 Mev) and somewhat less homogeneous (full width at half-maximum $\simeq$ 0.7 Mev, maximum energy = 22.4 Mev) than in the previous work. Relative excitation functions were determined by bombardment of foil stacks of natural uranium metal, thorium metal, and U^{235}-oxide plated on platinum, with appropriate absorbers interspersed. The stopping powers of uranium and thorium were determined experimentally relative to that of copper (they were about 65 percent of the copper stopping power). The fission product used most consistently was 17-hr Zr^{97} (in equilibrium with 73-min Nb^{97}), although several checks were made with 85-min Ba^{139} and 9.7-hr Sr^{91}.

Absolute cross sections were determined in separate runs employing copper foils. The current was then obtained from the known cross section for the $Cu^{63}(p,n)$ reaction[8] in the region of its broad maximum at about 13 Mev. A large number of determinations (25 for U^{238}, 13 for Th^{232}, and 8 for U^{235}) was carried out. As a check on the method, measurements were made of the cross section for production of Na^{22} by proton bombardment of magnesium and compared with the measurement of that cross section by Batzel and Coleman[9] in the region of its broad minimum near 20 Mev. The magnesium cross section obtained by comparison with copper was larger than the value from reference 9 by 20 percent; in order to take this discrepancy into account, cross sections measured by the copper comparison were reduced by 10 percent.

The maximum error in the ratio of the cross section for production of Zr^{97} to the $Cu^{63}(p,n)$ cross section is estimated to be about 20 percent. The uncertainty in the absolute cross sections on which the measurements are based should not be larger than 20 percent, and errors due to energy uncertainties in the comparison of the cross sections are very small. Errors in the determination of fission yields from the smooth curve (this was necessary in the case of U^{235} and U^{238}) are generally about 5 percent in the regions of Zr, Sr, and Ba; neither studies of fission fine structure[10] nor the direct determi-

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[1] J. Jungerman, Phys. Rev. **79**, 632 (1950).
[2] J. Jungerman and S. C. Wright, Phys. Rev. **74**, 150 (1948).
[3] H. A. Tewes and R. A. James, Phys. Rev. **88**, 860 (1952).
[4] Fowler, Jones, and Paehler, Phys. Rev. **86**, 71 (1952).
[5] Jones, Timnick, Handley, and Paehler (private communication).
[6] Timnick, Handley, and Paehler (private communication).
[7] Cohen, Newman, Charpie, and Handley, Phys. Rev. **94**, 620 (1954).
[8] S. N. Ghoshal, Phys. Rev. **80**, 939 (1959).
[9] R. E. Batzel and G. H. Coleman, Phys. Rev. **93**, 280 (1954).
[10] See, for example, A. C. Pappas, Atomic Energy Commission Report AECU 2806 (unpublished). Other references are listed therein.

nations of the yields of these nuclides in thermal fission[11] or proton-induced thorium fission[4] give any indication that their yields are abnormal. Considering the fact that the Ba and Sr checks agree well with the Zr data, it is believed that uncertainties in the fission yields could not contribute errors larger than 8 percent to the absolute fission cross sections. While the sum of these effects gives a rather large maximum possible error (~50 percent), the *probable* error in the absolute cross section for U^{238} is less than 15 percent. The errors in the ratios of the cross sections for U^{235} and Th^{232} to that of U^{238} are considerably smaller.

The absolute cross section for $U^{238}(p,f)$ was determined to be 1.28 barns at 21.5 Mev. The ratios of the cross sections for Th^{232} and U^{235} to that of U^{238} at this energy were found to be 0.65 ± 0.06 and 1.02 ± 0.10, respectively. The thorium value is based largely on the Zr^{97} fission yield from reference 4, which, after remaining at roughly 4.15 percent for proton energies from 9 to 19.5 Mev, suddenly falls to 3.63 ± 0.09 percent at 21.1 Mev. If this jump is assumed not to be real so that the fission yield remains at about 4.15 percent, the ratio of the Th^{232} to U^{238} cross sections becomes 0.57.

The excitation functions for fission in U^{238} and Th^{232} are shown in Figs. 1 and 2, normalized to the above absolute values. In order to determine the total reaction cross sections, it is necessary to obtain some estimate of cross sections for competing reactions. The principal reactions to be considered are (p,xn), i.e., (p,n), $(p,2n)$, $(p,3n)$, etc. Tewes and James[4] have measured the (p,n) and $(p,3n)$ cross sections for Th^{232}; their excitation functions indicate that $(p,3n)$ is predominant at 21 Mev, and they found its cross section to be about 35 percent of the fission cross section. Their measurement is indicated in Fig. 2. Measurements of the (p,n) and $(p,3n)$ cross sections and excitation functions for U^{238} were made by comparing yields of Np^{236} and Np^{238} with those of Zr^{97}. Since the neptunium determinations require carrier-free radiochemistry, the results are somewhat inaccurate. Chemical yields from the organic-solvent extractions were estimated by counting the activity in aliquots from repeated extractions from both the solvent and acid phases. Several other tests were made to ascertain that the neptunium recovery efficiency was high. The final activities were identified as Np^{236} and Np^{238} (in the high-energy and low-energy foils, respectively) by half-life, absorption characteristics, and rough determinations of the gamma-ray spectrum. The $(p,3n)$ excitation function is shown in Fig. 1. Its shape indicates that it is the predominant (p,xn) reaction at 21.5 Mev. Its absolute value at that energy, 32 ± 13 mb, represents an essentially negligible difference between the fission cross section and the total reaction cross section for U^{238}. The (p,n) cross section was found to be 4 ± 2 mb at 14 Mev, and somewhat less at higher energies.

The fact that the fission cross section is considerably smaller in Th^{232} than in U^{238} was also found in the case of alpha- and deuteron-induced fission; it now seems

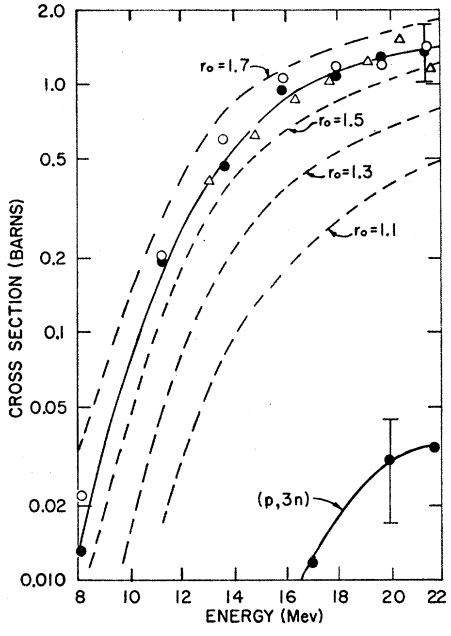

FIG. 1. Fission and $(p,3n)$ excitation functions for protons on U^{238}. The dashed curves are the theoretical total reaction cross sections for various nuclear radii.

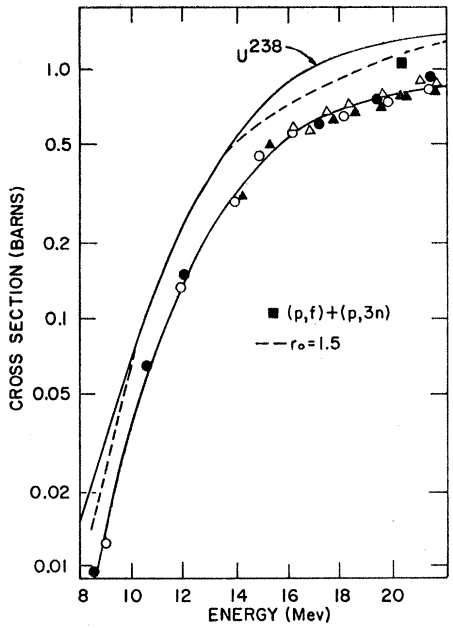

FIG. 2. Fission excitation function for protons on Th^{232}. The point labeled $(p,f)+(p,3n)$ is based on the ratio of $(p,3n)$ to (p,f) cross sections from reference 4. The dashed curve is the theoretical total reaction cross section for $r_0=1.5$, and the line labeled "U^{238}" is from Fig. 1.

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

quite evident that this is due to less effective competition of (p,xn) reactions in the case of U^{238}.

The principal quantity of theoretical interest that can be derived from these measurements is the nuclear radius. The total reaction cross section can be expressed as a product of the geometric cross section [approximately $\pi(R+\lambda\!\!\!^{-})^2$], a Coulomb factor, and a "sticking probability." At energies well above the Coulomb barrier, the Coulomb factor merely takes into account the fact that the path of an incident charged particle approaching a nucleus is hyperbolic rather than rectilinear. The exact quantum mechanical results can be reproduced with reasonable accuracy by a simple classical calculation from Newtonian mechanics.[12] This calculation is quite insensitive to the distribution of charge inside the nucleus. The "sticking probability" has a maximum value of unity and is well known to approach that value closely for the cases under consideration. Furthermore, it will be seen that the interesting feature of these measurements is the large value of the cross section; this feature would be unchanged if the "sticking probability" were less than unity. Measurements of total reaction cross sections at energies well above the Coulomb barrier therefore have a considerable advantage over determinations of the nuclear radius by Coulomb barrier penetration[13] [such as the half-life—energy relationship in alpha decay and low-energy (p,n) excitation functions] or by total neutron cross sections[13] which are difficult to interpret theoretically. The interpretation of the nuclear radius determined from total reaction cross sections is very straightforward, and essentially classical—when an incident particle passes within that distance of the center of the nucleus, a nuclear reaction takes place.

Figure 1 shows the theoretical total reaction cross section for $r_0=1.1$, 1.3, 1.5, and 1.7 (nuclear radius $=r_0A^{\frac{1}{3}}\times10^{-13}$ cm), calculated as described by Shapiro *et al.*[14] Comparison of these curves with the data indicates that $r_0=1.55\pm0.1$. Considering the uncertainties involved, $r_0=1.5$ is within the expected error, but values lower than about 1.3 seem to be completely out of the question. The same conclusion is obtained from the U^{235} and Th^{232} cross sections. It is interesting to note that this value of r_0 is in excellent agreement with the only other existing measurement of a total reaction cross section at energies well above the Coulomb barrier, Kelly's determination of the (p,n), $(p,2n)$, and $(p,3n)$ cross sections for bismuth.[15]

The fact that this value of r_0 is considerably larger than that found from the electron scattering[16] and from μ-mesonic x-ray fine structure[17] measurements (although it is in general agreement with determinations by other nuclear methods)[13] is at first surprising. However, this discrepancy between the determinations of the nuclear radius by electromagnetic and nuclear methods may perhaps be explained as follows: Electromagnetic measurements determine the radius at which the absolute value of the nuclear wave function becomes different from zero; since the nuclear wave function determines the spatial distribution of the nucleons considered as points—there is nothing in the wave function which indicates that a point nucleon is the center of a nuclear force—electromagnetic experiments determine the spatial distribution of these "point" nucleons. Nuclear experiments, however, determine the radius at which the incident particle first experiences a nuclear force. This happens when it comes within the range of the nuclear force of the "point" nucleons in the target nucleus. Thus, radius determinations by nuclear experiments should be larger by the range of nuclear forces than determinations by electromagnetic experiments. For the heaviest nuclei, this represents a difference of about 25 percent, which approximately accounts for the discrepancy.

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[12] J. M. Blatt and V. F. Weisskoff, *Theoretical Nuclear Physics* (John Wiley and Sons, Inc., New York, 1952).

[13] Summarized in reference 12, p. 15.

[14] M. M. Shapiro, Phys. Rev. **90**, 171 (1953); Feshbach, Shapiro, and Weisskoff, U. S. Atomic Energy Commission Report NYO 3077 (unpublished).

[15] E. Kelly, University of California Radiation Laboratory Report UCRL 1044 and private communication.

[16] Hofstadter, Fechter, and McIntyre, Phys. Rev. **92**, 978 (1953).

[17] V. L. Fitch and J. Rainwater, Phys. Rev. **92**, 789 (1953).