

Excitation Functions of ( $p,2$  Nucleon) Reactions\*

P. P. STROHAL†† AND A. A. CARETTO, JR.

*Department of Chemistry, Carnegie Institute of Technology, Pittsburgh, Pennsylvania*

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Excitation functions for the ( $p,pn$ ) reaction have been determined in the energy range 250 Mev to 440 Mev for the target nuclei As<sup>75</sup>, Br<sup>81</sup>, Zr<sup>96</sup>, I<sup>127</sup>, Ce<sup>142</sup>, W<sup>186</sup>, and Re<sup>187</sup>. Both isomer products were determined in the case of the Br<sup>81</sup>( $p,pn$ )Br<sup>80</sup> reaction. Also, excitation functions were determined for the ( $p,2p$ ) reaction on Zr<sup>96</sup>, Ce<sup>142</sup>, and W<sup>186</sup> and for the ( $p,2n$ ) reaction on Zr<sup>96</sup>. All cross sections were measured relative to the Al<sup>27</sup>( $p,3pn$ )Na<sup>24</sup> excitation function. The ( $p,pn$ ) excitation functions are only moderately sensitive to energy showing an increase by about 30% for iodine, 20% for arsenic and cerium, and a 12% decrease for zirconium. The remaining excitation functions are flat. The ratio of  $\sigma_{p,pn}$  to  $\sigma_{p,2p}$  is  $1.62 \pm 0.09$ , for the three targets measured, at 440 Mev. The various mechanisms for the production of reactions of this type are examined in the light of present understanding. The suggestion is made that, despite the relative insensitiveness of these excitation functions to energy, ( $p,pn$ ) and ( $p,2p$ ) reactions may take place by more than one mechanism in this energy region. A comparison is made with the results of recent Monte Carlo nuclear cascade calculations applied to calculation of ( $p,pn$ ) reaction excitation functions.

## INTRODUCTION

IN the past several years, considerable work has been done in deducing and ascertaining the mechanism or mechanisms of high-energy nuclear reactions. It is now felt that the over-all distribution and yields of products resulting from high-energy nuclear reactions can be explained by the statistical nature of the initial nuclear cascade followed by nucleon evaporation until thermodynamic equilibrium is attained.<sup>1</sup> Monte Carlo calculations<sup>2-5</sup> of both phases of this process are in agreement with the general features of the experimentally observed results.

More recently, attention has been given to some of the "simpler" high-energy-induced nuclear reactions. Use of the word simple is meant as a measure of the complexity of the reaction in terms of numbers of particles entering into or leaving the reaction rather than the nature of the reaction mechanism.

Work on the measurement of excitation functions of ( $p,pn$ ) reactions has been done at several laboratories.<sup>6-10</sup> Simple reactions such as the ( $p,pn$ ) and ( $p,2p$ ), i.e., interactions in which  $A$  decreases by one unit and  $Z$  either remains the same or decreases by one, are of

particular interest, because in the energy range of several hundred Mev they account for several percent of the geometric cross section of the target nuclei, and are found to be largely energy independent. Reactions of this type have been interpreted as proceeding largely by the direct knock-on ejection of a proton or neutron at these incident energies.<sup>11</sup>

Most of the previous studies of these reactions have touched only lightly on the energy region of 100 to 400 Mev. It therefore seemed worthwhile to investigate in greater detail ( $p,pn$ ) and ( $p,2p$ ) reactions in this energy region.

Target nuclides for this study covered a large range of nuclear sizes and included: As<sup>75</sup>, Br<sup>81</sup>, Zr<sup>96</sup>, I<sup>127</sup>, Ce<sup>142</sup>, W<sup>186</sup>, and Re<sup>187</sup>. In the case of Br<sup>81</sup>, the cross section for the ( $p,pn$ ) reaction was measured for the production of both the 4.4-hr isomer and the 18-minute ground state of Br<sup>80</sup>. Only for the targets Zr<sup>96</sup>, Ce<sup>142</sup>, and W<sup>186</sup> was it possible to measure both the ( $p,pn$ ) and ( $p,2p$ ) reaction products. The ( $p,2n$ ) reaction on Zr<sup>96</sup> is the only reaction of this type measured. In all cases the criterion used for selection of targets was that the half-lives and radiations of the product nuclides be suitable for measurement with the available equipment.

## EXPERIMENTAL

All the irradiations were carried out in the circulating beam of the Carnegie Institute of Technology proton synchrocyclotron with proton energies of 250 to 440 Mev. The targets, whose total thickness varied between 35 and 100 mg/cm<sup>2</sup>, were irradiated for periods of 10 to 60 minutes. The incident proton energy was determined by placing the target at different radial distances in the circulating beam. The relative error of the spread in the energy of the proton beam was about  $\pm 2\%$  at 440 Mev to about  $\pm 5\%$  at 250 Mev.

All cross sections were measured relative to the cross section for the Al<sup>27</sup>( $p,3pn$ )Na<sup>24</sup> monitor reaction. The

<sup>11</sup> E. Belmont and J. M. Miller, Phys. Rev. **95**, 1554 (1954).

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† Presented in partial fulfillment of the Ph.D. degree in the Department of Chemistry, University of Zagreb, Zagreb, Yugoslavia.

‡ Present address: Institut Ruder Boskovic, Zagreb, Yugoslavia.

<sup>1</sup> J. M. Miller and J. Hudis, *Annual Reviews of Nuclear Science* (Annual Reviews, Inc., Palo Alto, 1959), Vol. 9, p. 159.<sup>2</sup> N. Metropolis, R. Bivins, M. Storm, A. Turkevich, J. M. Miller, and G. Friedlander, Phys. Rev. **110**, 185 (1958).<sup>3</sup> N. Metropolis, R. Bivins, M. Storm, J. M. Miller, G. Friedlander, and A. Turkevich, Phys. Rev. **110**, 204 (1958).<sup>4</sup> I. Dostrovsky, P. Rabinowitz, and R. Bivins, Phys. Rev. **111**, 1659 (1958).<sup>5</sup> I. Dostrovsky, Z. Fraenkel, and G. Friedlander, Phys. Rev. **116**, 683 (1959).<sup>6</sup> A. A. Caretto and G. Friedlander, Phys. Rev. **110**, 1169 (1958).<sup>7</sup> S. Markowitz, F. S. Rowland, and G. Friedlander, Phys. Rev. **112**, 1295 (1958).<sup>8</sup> I. M. Ladenbauer and L. Winsberg, Phys. Rev. **119**, 1368 (1960).<sup>9</sup> D. R. Nethaway and L. Winsberg, Phys. Rev. **119**, 1375 (1960).<sup>10</sup> H. P. Yule and A. Turkevich, Phys. Rev. **118**, 1591 (1960).

cross sections for this reaction at any given energy were based on the excitation function of Hicks *et al.*<sup>12</sup> This excitation function is in reasonably good agreement with that recently determined by Yule and Turkevich<sup>10</sup> and is at most 10% low in the region of 200 Mev. Both these determinations of the  $\text{Al}^{27}(p,3pn)\text{Na}^{24}$  excitation function are based on the absolute  $\text{C}^{12}(p,pn)\text{C}^{11}$  cross sections of Crandall.<sup>13</sup>

The targets were prepared by making a slurry of the target compound in acetone, placed in a 1-cm $\times$ 1-cm depression in a 3-mil aluminum foil, and dried to an even uniform mat. On top of the target compound were placed three aluminum foils, the middle one of which was used as monitor. The entire target assembly was wrapped in 1-mil aluminum foil and fastened on the end of the cyclotron probe. An identical area of target material and monitor foil were measured in the experiment. Misalignment of the aluminum monitor foil and target material by this technique was considered to produce less than a 5% effect as judged by the reproducibility from one run to another.

Recoil losses of the  $(p,pn)$  and  $(p,2p)$  products were considered negligible in accord with the experiments of Markowitz *et al.*<sup>7</sup> The contribution to the measured reaction yield from secondary particles is also considered negligible for the range of target thicknesses used in this work, again in agreement with the findings of other workers.<sup>7,10</sup> In some cases, i.e., zirconium, the value for the cross section was observed to be independent of large variations of target thickness, between 8 and 48 mg/cm<sup>2</sup>. Also, no effort was made to correct for the small contribution to the  $\text{Na}^{24}$  production in the monitor foil from secondary particles originating in the target assembly. This effect cannot be more than a few percent. Table I indicates the composition and thickness of the various targets used in the irradiations.

After irradiation the monitor foil and target material were removed from the target assembly and the disintegration rates of the various nuclides of interest were determined. The chemical procedures used are modifications of standard methods<sup>14-16</sup> and are described in the Appendix. These are procedures capable of achieving purification from all other impurities by factors of 10<sup>6</sup> or better. The final precipitates, mounted on small filter disks were counted under standard Tracerlab Geiger-Müller tubes in a known geometry. The chemical yields were usually determined gravimetrically. Decay curves were analyzed into their various components and for the radioactive nuclide sought, the disintegration rate for infinitely long bombardment was computed by

<sup>12</sup> H. G. Hicks, P. C. Stevenson, and W. E. Nervik, *Phys. Rev.* **102**, 1390 (1956).

<sup>13</sup> W. E. Crandall, G. P. Millburn, R. V. Pyle, and W. Birnbaum, *Phys. Rev.* **101**, 329 (1958).

<sup>14</sup> Atomic Energy Commission Report LA-1566c edited by J. Kleinberg, 1954 (unpublished).

<sup>15</sup> M. Lindner, University of California Radiation Laboratory Report UCRL-4377, 1959 (unpublished).

<sup>16</sup> W. W. Meinke, Atomic Energy Commission Report AEC-3084, 1951 (unpublished).

TABLE I. Target composition.

Target isotope	Abundance %	Chemical form	Target thickness (mg/cm <sup>2</sup> )
As <sup>75</sup>	100	As metal powder	35-50
Br <sup>81</sup>	49.4	NaBr	20-42
Zr <sup>96</sup>	2.8	Zr foil	8-48
I <sup>127</sup>	100	KI	20-30
Ce <sup>142</sup>	11.08	CeO <sub>2</sub>	25-30
W <sup>186</sup>	28.7	WO <sub>3</sub>	40-83
Re <sup>187</sup>	62.9	Re metal powder	65-130

corrections for length of bombardment, chemical yield, and counting efficiency. Corrections for self-absorption and scattering were made by the method outlined by Nervik<sup>17</sup> and air and counter window absorption correction factors were obtained by the procedure described by Rudstam.<sup>18</sup> For nuclides which emit  $\beta$ -rays of energy greater than about 0.70 Mev the effects of air-window absorption and self-scattering and self-absorption were usually negligible and the correction factors could be assigned the value of unity. Most of the data used for the calculation of counting efficiencies were taken from the compilations of Strominger *et al.*<sup>19</sup>

In all cases where the cross sections that were calculated were for nuclides in a decay chain [e.g.,  $(p,2p)$ ,  $(p,pn)$ , and  $(p,2n)$  reactions on Zr<sup>96</sup>] corrections were made for the growth and decay of the products during bombardment<sup>20</sup> and decay before chemical separation was achieved.

## RESULTS

The cross sections for the  $(p,pn)$  reactions, and  $(p,2p)$  and  $(p,2n)$  reactions where measured, for the target nuclides Br<sup>81</sup>, As<sup>75</sup>, Zr<sup>96</sup>, I<sup>127</sup>, Ce<sup>142</sup>, W<sup>186</sup>, and Re<sup>187</sup> are listed in Table II. The reactions with Zr<sup>96</sup> and Ce<sup>142</sup> are illustrated in Figs. 1 and 2. All the values given are averages of two or more determinations and the errors given are the deviations from the averages. Errors in these determinations arise from a number of sources. The error in the relative monitoring of the proton beam by use of aluminum foils and the possible misalignment thereof is only a few percent at any given energy. The absolute value of the  $\text{Al}^{27}(p,3pn)\text{Na}^{24}$  cross section is uncertain by about  $\pm 10\%$ . Uncertainties in counting efficiency, backscattering, air and window absorption, self-absorption and self-scattering were estimated to be about  $\pm 15\%$ . Errors resulting from the resolution of decay curves may be as large as 10%. The chemical yield determinations were accurate to 2-3% individually, but may be in error by 5% when multiple determinations are necessary in a parent-daughter

<sup>17</sup> W. E. Nervik and P. C. Stevenson, *Nucleonics* **10**, No. 3, 18 (1952).

<sup>18</sup> G. Rudstam, Doctoral thesis, University of Uppsala, Uppsala, Sweden, 1956 (unpublished), p. 6191.

<sup>19</sup> D. Strominger, J. M. Hollander, and G. T. Seaborg, *Revs. Modern Phys.* **30**, 585 (1958).

<sup>20</sup> W. Rubinson, *J. Chem. Phys.* **17**, 542 (1949).

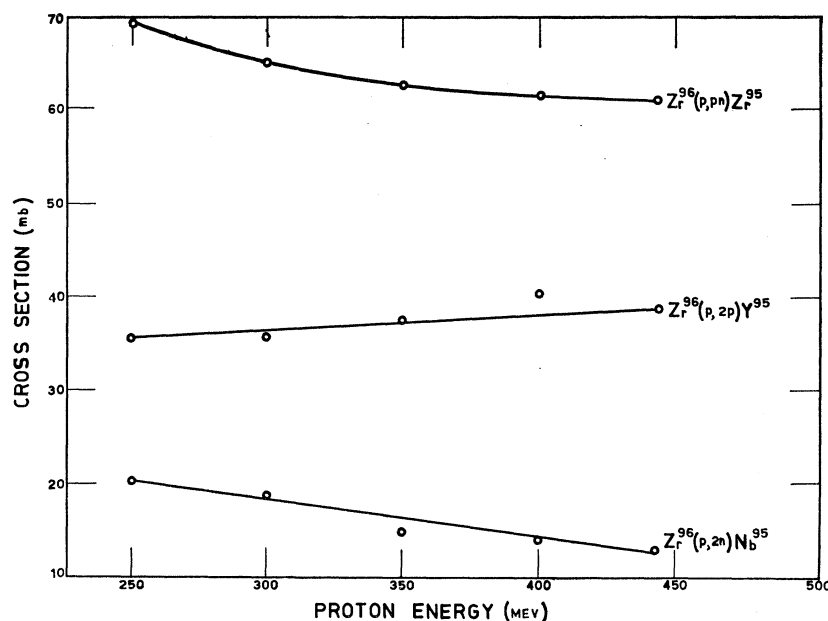


FIG. 1. Excitation functions for the (*p*,*pn*), (*p*,*2p*), and (*p*,*2n*) reactions on  $Zr^{96}$ .

chain. As a result of all these sources of error it is believed that the reported cross sections are accurate to about  $\pm 15\%$ . However, the ratios of the  $\sigma_{p,pn}/\sigma_{p,2p}$ ,  $\sigma_{p,pn}/\sigma_{p,2n}$ , and the  $Br^{80}$  isomer ratio are subject to only the last two sources of error mentioned and therefore should be reliable to about  $\pm 8\%$  as indicated in Table III.

With the exception of the (*p*,*pn*) reaction on  $As^{75}$ ,  $I^{127}$ , and  $Ce^{142}$ , the (*p*,*pn*) and (*p*,*2p*) reactions on the remaining elements are nearly independent of energy. In the case of  $As^{75}$  we report cross sections (see Table II) of 57 mb at 250 Mev with a gradual increase to 71 mb at 440 Mev. This compares with the data of Cumming<sup>21</sup> in which he quotes a value of 55 mb for

the cross section for the production of  $As^{74}$  by the (*p*,*pn*) reaction at 380 Mev. It is felt that further experiments should be performed to check the energy dependence of the (*p*,*pn*) reaction on arsenic.

The  $I^{127}(p,pn)I^{126}$  excitation function reported here has a cross section of 58 mb at 250 Mev and 77 mb at 440. This compares poorly with the experiments of Ladenbauer and Winsberg<sup>8</sup> who report  $(80.6 \pm 7.0)$  mb for this cross section at 250 Mev and  $(63.3 \pm 8.7)$  mb at 500 Mev with a continuous smooth decrease to a value of  $(46 \pm 4.3)$  mb at 6.2 Bev. Kuznetsova *et al.*<sup>22</sup> report values of  $(53 \pm 8)$  mb at 300 Mev and  $(51 \pm 10)$  mb at 660 Mev with a point at  $(70 \pm 9)$  mb at 480 Mev in fair agreement with the present data. The reason for

TABLE II. Summary of experimental results.

Proton energy (Mev)	Average cross sections (mb)				
	250	300	350	400	440
	( <i>p</i> , <i>pn</i> ) reactions				
$As^{75}(p,pn)As^{74}$	$57.4 \pm 6.6$	$60.7 \pm 5.6$	$63.4 \pm 4.8$	$66.0 \pm 3.3$	$71.0 \pm 7.0$
$Br^{81}(p,pn)Br^{80}$	$65.6 \pm 4.9$	$66.0 \pm 6.6$	$65.3 \pm 4.8$	$64.2 \pm 3.5$	$62.0 \pm 2.0$
$Br^{81}(p,pn)Br^{80m}$	$34.4 \pm 4.3$	$35.0 \pm 4.0$	$34.5 \pm 4.3$	$34.2 \pm 2.3$	$33.4 \pm 3.5$
$Br^{81}(p,pn)Br^{80g}$	$31.2 \pm 4.5$	$31.0 \pm 4.2$	$30.8 \pm 3.6$	$30.0 \pm 3.0$	$28.6 \pm 4.4$
$Zr^{96}(p,pn)Zr^{95}$	$69.8 \pm 5.4$	$65.7 \pm 4.4$	$63.3 \pm 2.0$	$61.4 \pm 3.6$	$61.3 \pm 2.4$
$I^{127}(p,pn)I^{126}$	$58.4 \pm 3.6$	$57.7 \pm 4.1$	$66.5 \pm 2.4$	$74.8 \pm 5.4$	$77.6 \pm 6.3$
$Ce^{142}(p,pn)Ce^{141}$	$70.9 \pm 1.0$	$73.2 \pm 3.3$	$83.8 \pm 4.5$	$82.6 \pm 4.0$	$82.5 \pm 6.2$
$W^{186}(p,pn)W^{185}$	$69.4 \pm 1.0$	$69.5 \pm 4.3$	$68.7 \pm 4.4$	$68.6 \pm 5.7$	$70.2 \pm 5.6$
$Re^{187}(p,pn)Re^{186}$	$67.1 \pm 2.2$	$67.5 \pm 6.7$	$67.4 \pm 5.0$	$66.8 \pm 4.6$	$68.0 \pm 3.7$
	( <i>p</i> , <i>2p</i> ) and ( <i>p</i> , <i>2n</i> ) reactions				
$Zr^{96}(p,2p)Y^{95}$	$35.8 \pm 3.6$	$35.0 \pm 4.2$	$37.4 \pm 4.6$	$40.4 \pm 3.2$	$37.8 \pm 2.4$
$Zr^{96}(p,2n)Nb^{95}$	$20.6 \pm 1.6$	$19.0 \pm 1.0$	$15.2 \pm 3.2$	$14.4 \pm 4.2$	$13.9 \pm 3.7$
$Ce^{142}(p,2p)La^{141}$	$22.5 \pm 1.4$	$30.4 \pm 3.2$	$47.6 \pm 2.7$	$54.3 \pm 3.8$	$50.9 \pm 4.8$
$W^{186}(p,2p)Ta^{185}$	$44.1 \pm 3.3$	$43.4 \pm 4.5$	$44.9 \pm 4.4$	$43.8 \pm 3.9$	$43.4 \pm 4.8$

<sup>21</sup> J. B. Cumming, Atomic Energy Commission Document, NYO 6141, 1954 (unpublished).

<sup>22</sup> M. Ia. Kuznetsova, V. N. Mekhedov, and V. A. Khalkin, J. Exptl. Theoret. Phys. (U.S.S.R.) **34**, 1096 (1958) [translation: Soviet Phys.—JETP **34**, 7, 759 (1958)].

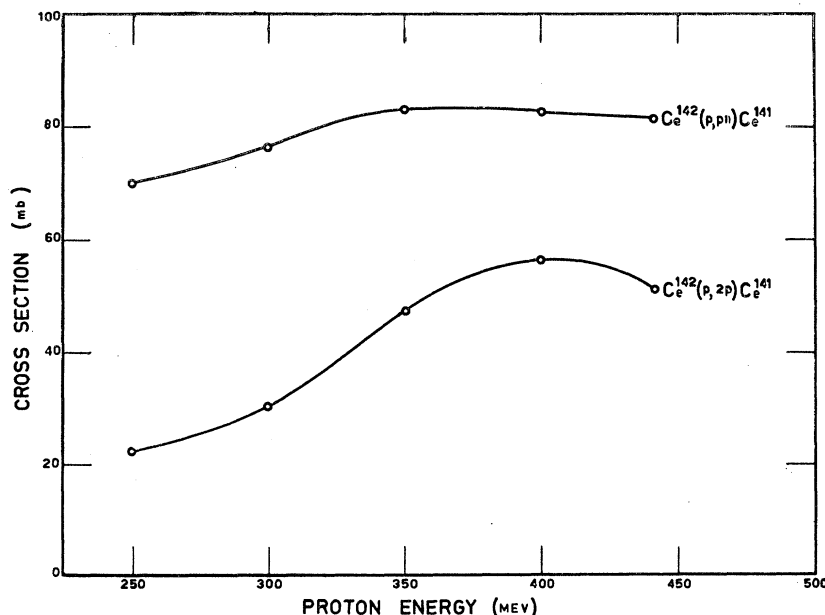


FIG. 2. Excitation functions for the  $(p,pn)$  and  $(p,2p)$  reactions on  $Ce^{142}$ .

the disagreement with the results of Ladenbauer seems obscure and awaits further experimentation. The discrepancies in the results reported here with other data could of course arise from many sources and it would be difficult to assess the trouble. One source of these discrepancies, however, can probably be traced to lack of uniformity among various authors in the values used for the  $Al(p,3pn)$  monitor cross section. Differences in reported cross sections by as much as 15% at various energies could be realized by using the  $Na^{24}$  cross-section data reported by Hintz and Ramsey,<sup>23</sup> Hicks *et al.*,<sup>12</sup> and Prokoshkin *et al.*<sup>24</sup> It is difficult to decide if these different values used for the  $Na^{24}$  cross section can be responsible for part of the discrepancies found in this work as compared with other authors, because frequently no clear statement as to which value was chosen for the  $Na^{24}$  cross section could be found in the literature.

The  $Ce^{142}(p,pn)Ce^{141}$  reaction has a cross section of  $(71 \pm 1)$  mb at 250 Mev which agrees with the data of Ware and Wiig<sup>25</sup> for the same reaction at 240 Mev. Also, the value of 82 mb at 440 Mev reported here compares well with the value of 86 mb at the same energy as reported by Caretto and Friedlander.<sup>6</sup>

## DISCUSSION

### $(p,pn)$ Reactions

Two general considerations should be made in discussing  $(p,pn)$  reactions in the energy interval 200 to 450 Mev.

(1) The gross features of the majority of  $(p,pn)$  excitation functions are essentially energy independent in this region. A number of minor exceptions may be noted. The arsenic and cerium excitation functions show gradual increases by about 20% between 250 Mev and 440 Mev. The iodine cross sections reported here show an increase of about 30%, while the zirconium  $(p,pn)$  reaction exhibits a decrease of about 12% in the same energy interval.

(2) The cross section for the  $(p,pn)$  reaction at a given energy plotted as a function of the target mass is illustrated in Fig. 3. Any theory concerning the mechanism of  $(p,pn)$  reactions should be able to explain the dependence of the  $(p,pn)$  reaction cross section on target mass number. Figure 3 illustrates the present experimental situation<sup>6,7,8,10,13,21,22,24,26</sup> with regard to this dependence for various  $(p,pn)$  reactions at 400 Mev.

TABLE III. Ratios of various  $(p,2p)$  reactions.

Proton energy (Mev)	250	300	350	400	440
$Br^{81}(p,pn)Br^{80m}/Br^{81}(p,pn)Br^{80g}$	$1.10 \pm 0.04$	$1.13 \pm 0.03$	$1.12 \pm 0.04$	$1.14 \pm 0.04$	$1.16 \pm 0.05$
$Zr^{96}(p,pn)Zr^{95}/Zr^{96}(p,2p)Y^{95}$	$1.95 \pm 0.20$	$1.88 \pm 0.08$	$1.53 \pm 0.50$	$1.56 \pm 0.04$	$1.62 \pm 0.03$
$Ce^{142}(p,pn)Ce^{141}/Ce^{142}(p,2p)Ce^{141}$	$3.15 \pm 0.08$	$2.40 \pm 0.01$	$1.76 \pm 0.01$	$1.52 \pm 0.03$	$1.62 \pm 0.08$
$W^{186}(p,pn)W^{185}/W^{186}(p,2p)Ta^{185}$	$1.57 \pm 0.01$	$1.60 \pm 0.04$	$1.53 \pm 0.50$	$1.56 \pm 0.04$	$1.62 \pm 0.03$

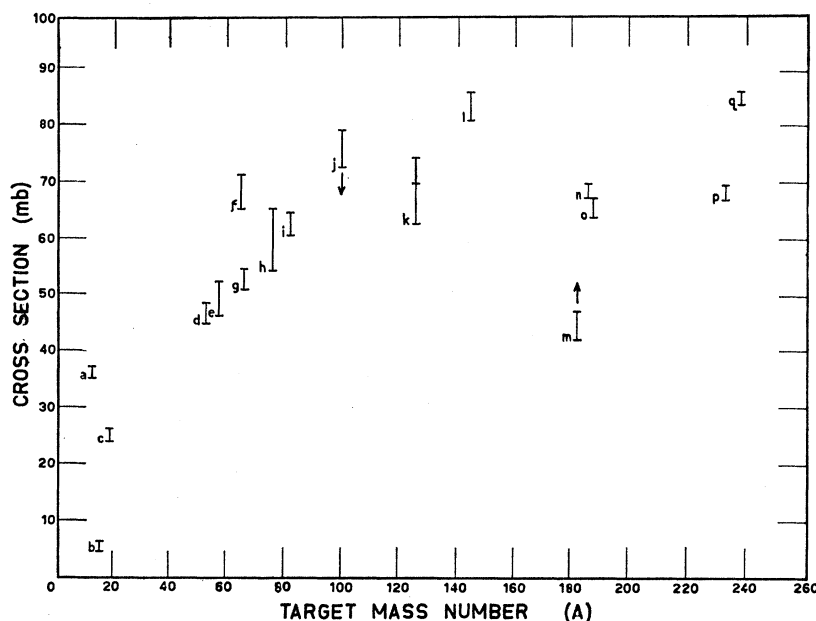
<sup>23</sup> N. M. Hintz and N. F. Ramsey, Phys. Rev. **88**, 19 (1952).

<sup>24</sup> Iu. D. Prokoshkin and A. A. Tiapkin, Soviet Phys.—JETP **5**, 148 (1957).

<sup>25</sup> W. Ware and E. O. Wiig (private communication).

<sup>26</sup> M. L. Lindner and R. N. Osborne, Phys. Rev. **103**, 378 (1956).

FIG. 3. Cross sections for ( $p, pn$ ) reactions at 400 Mev versus target mass number. Points are designated by target nucleus and reference: (a)  $C^{12,13}$ , (b)  $N^{14,7}$ , (c)  $F^{19,7}$ , (d)  $Fe^{54,7}$ , (e)  $Ni^{58,7}$ , (f)  $Zn^{64,7}$ , (g)  $Cu^{65,10}$ , (h)  $As^{75}$  (reference 21 and this work), (i)  $Br^{81}$  (this work), (j)  $Mo^{100,7}$  includes contribution of ( $p, 2p$ ) reaction, (k)  $I^{127}$  (references 8 and 22 and this work), (l)  $Ce^{142}$  (reference 6 and this work), (m)  $Ta^{181,7}$  only the  $Ta^{180}$  upper state was measured, (n)  $W^{186}$  (this work), (o)  $Re^{187}$  (this work), (p)  $Th^{232,24}$ , (q)  $U^{238,26}$



In view of the rather large errors quoted for ( $p, pn$ ) reaction cross sections, and the frequent large differences between cross sections quoted for the same reaction measured in different laboratories, it does not appear justifiable at this time to search for trends or find structure in the data illustrated here.

Before discussing these results further, it is useful to consider the possible mechanisms available to the ( $p, pn$ ) reaction in this energy interval. It does not appear likely that compound nucleus mechanisms can play a significant role at energies above 100 Mev. The results of the Monte Carlo cascade calculations<sup>2</sup> indicate that the fraction of all events leading to compound nucleus formation decreases with increasing energy and increases with atomic weight. Thus, those authors report that, for 100-Mev incident protons compound nucleus formation occurs in 6% of the total inelastic interactions for copper and in 30% of all inelastic interactions for uranium. At 200 Mev, however, the fraction of all inelastic events leading to compound nucleus has decreased to less than 1% for copper and about 8% for uranium.

Other mechanisms available for the ( $p, pn$ ) reaction are:

(a) The pure knock-on process involves the collision of the incident particle with a neutron in which both collision partners are knocked out of the nucleus and in which the nucleus is left with insufficient residual excitation energy to allow any nucleon evaporation. This latter restriction implies that the "hole" left by the removal of a neutron must not be sufficiently deep in the nuclear potential well so that nucleon evaporation will ensue.

The knock-on process may also take place wherein the incident particle engages in a scattering collision

with a nucleon with which it shares a large fraction of its energy. This can lead to a small cascade after which both collision partners leave the nucleus carrying with them almost all the incident excitation energy. This process is again restricted to those cases in which the residual excitation energy is less than 10 Mev. This nonideal knock-on mechanism is probably not a very likely process.

(b) Another process would be one in which the incident proton makes an elastic collision with a nucleon in such a way that one collision partner is scattered out of the nucleus with almost all the incident excitation energy. Sufficient excitation is imparted to the struck nucleon so that after many internal collisions, an additional nucleon is finally evaporated. This process can take place either by a proton being knocked out followed by neutron evaporation, or less likely, a neutron being knocked out followed by proton evaporation. In either case, the initial collision must deposit about 10–20 Mev in the nucleus.

(c) The ( $p, pn$ ) reaction product could be produced by a reaction path involving the emission of deuterons, i.e., the ( $p, d$ ) reaction. These so-called pickup processes are well known at low energies<sup>27</sup> but are not considered too important here. They have been shown to be unimportant at 300 Mev.<sup>28</sup> Heidmann<sup>29</sup> theoretically predicts an  $E^{-6}$  dependence of the pickup process with energy.

(d) Reaction paths involving mesons may take place at incident energies above pion-production thresholds. Reactions such as ( $p, 2p\pi^-$ ) and ( $p, pn\pi^0$ ) are possible. It is not considered likely however that these mecha-

<sup>27</sup> W. Selove, Phys. Rev. **101**, 231 (1956).

<sup>28</sup> W. Hess and B. Moyer, Phys. Rev. **101**, 337 (1956).

<sup>29</sup> J. Heidmann, Phys. Rev. **80**, 171 (1950).

nisms can play a very important role in the energy region up to 440 Mev where the incident energy is only about twice the threshold for meson production. This is substantiated by comparison with the results of the Monte Carlo calculations<sup>3</sup> in which “ $(p, pn)$ ” reactions involving pions were found to be negligible at 0.46 Bev but contribute about half at 1.84 Bev.

Therefore, the mechanisms remaining to be discussed in connection with the results reported here are those listed as (a) and (b). A study of Cu<sup>64</sup> recoils from Cu<sup>65</sup> in this energy region, to be reported in a subsequent paper,<sup>30</sup> indicates that at least two different reaction mechanisms must be operative. This is somewhat surprising in view of the nearly energy-independent shape of this excitation function.<sup>10,31</sup> The reaction path probably involves mechanisms (a) and (b) in varying amounts. This fact should be kept in mind when considering the mechanisms of  $(p, pn)$  reactions. Ultimately one would expect both knock-on mechanisms and the mechanism involving  $(p, p')$  events followed by nucleon evaporation [mechanism (b)] to lead to smaller cross sections with increasing energies in this energy region. However, this would be expected to be a slow variation with energy related to the increasing mean free path in nuclear matter of the incident particle. In the small energy region between 250 Mev and 440 Mev one might expect increasing, decreasing, and constant valued excitation functions depending on the relative contributions of mechanisms (a) and (b) as a function of energy.

It has been suggested<sup>2,6,7,10</sup> that low-energy-deposition reactions such as the  $(p, pn)$  may take place predominantly in the nuclear surface. Another suggestion, made by Markowitz<sup>7</sup> and expanded by Grover<sup>32</sup> and Benioff,<sup>33</sup> is that nuclear shell structure plays an important role in knock-on reactions. Unfortunately, as was previously mentioned, it is difficult to ascertain the reasonableness of these suggestions with the available data. The data reported here are part of a continuing program of such studies in this laboratory. It is felt that once a large number of  $(p, pn)$  cross sections are measured, as accurately as possible, one should be able to ascertain the role that nuclear structure plays in these reactions.

Yule and Turkevich<sup>10</sup> report a comparison of their experimentally determined Cu<sup>65</sup> $(p, pn)$ Cu<sup>64</sup> reaction with the theoretically determined number of events from Monte Carlo calculations of Metropolis *et al.*<sup>2</sup> and evaporation calculations of the residual excited nuclei. Their calculations indicate that the knock-on process increases with energy for both Cu<sup>65</sup> and Au<sup>197</sup> while mechanism (b) [ $(p, p')$  followed by evaporation] decreases with increasing energy. The results reported here indicate that  $(p, pn)$  excitation functions on dif-

ferent targets may increase slightly, decrease slightly, or remain essentially energy independent. If these effects are indeed correct then this implies varying contributions of reaction mechanisms (a) and (b) for different mass targets. The calculations of Yule<sup>10</sup> on Cu<sup>65</sup> and Au<sup>197</sup> do not indicate relative changes in the contributions from the various possible mechanisms. It may be, however, that the Monte Carlo calculations are insensitive to such effects. Also, the number of cascades that were calculated to lead to the  $(p, pn)$  product by the various reaction paths were statistically small. Thus, one would not expect these Monte Carlo calculations to be of sufficient accuracy to indicate these trends.

One failure of the Monte Carlo calculations of Metropolis *et al.*<sup>2,3</sup> as well as by those of Rudstam<sup>18</sup> and Meadows<sup>34</sup> is that the cross-section values predicted for the  $(p, pn)$  reaction product are factors of 2 or 3 lower than the experimental values. It has been suggested<sup>1,10</sup> that this discrepancy arises from the use of a sharp nuclear boundary in these calculations thus giving an incorrect number of low-energy depositions. However, it is not clear that making use of a diffuse nuclear boundary will lead to varying contributions of different reaction mechanisms with energy.

### $(p, 2p)$ Reactions

The same mechanisms that apply to the  $(p, pn)$  reaction can apply to the  $(p, 2p)$  reaction. In the case of mechanism (b) one would expect a preference for neutron evaporation due to the coulomb barrier against proton emission.

It is of interest to compare the  $(p, pn)$  to  $(p, 2p)$  cross-section ratios for the three nuclides studied in this work. Table III lists these values. It can be noted that for Zr<sup>96</sup> this ratio has a value of 1.95 at 250 Mev and decreases regularly to a value of 1.62 at 440 Mev. For Ce<sup>142</sup> the ratio has a value of 3.15 at 250 Mev and a value of 1.62 at 440 Mev. However, for  $A=186$ , the W<sup>186</sup> $(p, pn)$ W<sup>185</sup> to W<sup>186</sup> $(p, 2p)$ Ta<sup>185</sup> reaction cross-section ratio is constant with a value of  $1.58 \pm 0.05$  throughout this energy region. It is interesting to note that the  $\sigma_{p, pn}$  to  $\sigma_{p, 2p}$  ratio for the three targets measured is  $1.62 \pm 0.09$  at 440 Mev. It is believed that the constancy of the  $(p, pn)$  to  $(p, 2p)$  cross-section ratio for W<sup>186</sup> with increasing energy is strong evidence for the knock-on mechanism. The knock-on mechanism is the only mechanism of those considered which would predict that the ratio of  $\sigma_{p, pn}$  to  $\sigma_{p, 2p}$  should be essentially energy-independent in this energy region. If the  $(p, pn)$  and  $(p, 2p)$  reactions took place by mechanism (b) one would expect, that as the spectrum of residual excitation energy changed with increasing incident energy, the probability of evaporation of a neutron versus a proton would not remain constant. It is thus concluded that the knock-on mechanism predominates for the  $(p, pn)$  and  $(p, 2p)$  reactions on W<sup>186</sup> between 200 to 440 Mev.

<sup>30</sup> E. Merz and A. A. Caretto, Jr. (to be published).

<sup>31</sup> D. W. Barr, Atomic Energy Commission Document, UCRL 3793, Berkeley, California, 1957 (unpublished).

<sup>32</sup> J. R. Grover (private communication).

<sup>33</sup> P. A. Benioff, Phys. Rev. **119**, 324 (1960).

<sup>34</sup> J. W. Meadows, Phys. Rev. **91**, 885 (1953).

**( $p, 2n$ ) Reaction**

The only ( $p, 2n$ ) reaction measured in this work is the  $Zr^{96}(p, 2n)Nb^{95}$  reaction. It can be noticed that the ( $p, 2n$ ) reaction is smaller than the ( $p, pn$ ) by a factor of at least 3.5 at all energies and smaller than the ( $p, 2p$ ) by a factor of 1.5 to 2 over the energy interval. This is not surprising if one considers that both the ( $p, pn$ ) and ( $p, 2p$ ) reactions can take place in one step via the knock-on process. The ( $p, 2n$ ) reaction on the other hand must involve a two-step process in which a neutron is evaporated. However, as was suggested by Winsberg,<sup>8</sup> one would expect about an order of magnitude difference between so called one-step and two-step processes. It is surprising that the cross section for this reaction is so close to that of the ( $p, 2p$ ) and ( $p, pn$ ) cross sections. Possibly this is evidence that there is a high probability for the ( $p, pn$ ) reaction in this energy region to take place by a two-step process such as mechanism (b).

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**APPENDIX**

In this Appendix all the chemical procedures that were used are briefly discussed. These are modifications of standard methods.<sup>14-16</sup> Also, that fraction of the disintegrations that were detected by beta radiation for each nuclide is designated.

**Arsenic**

The arsenic target was dissolved in aqua regia with the addition of carriers for the elements Se down to Cu. Se was removed by evaporating the solution to dryness and converting the solution to 1*N* in HCl and the addition of  $NH_2OH \cdot HCl$ . Addition of 1*N* KI removed the remainder of the Se and  $I_2$ . The supernate solution was then made 3*N* in HCl and  $As_2S_3$  precipitated with  $H_2S$ . As and Ge were dissolved in dilute KOH, leaving CuS insoluble. Ge was removed by distillation from 6*N* HCl in a slow stream of chlorine.  $As_2S_3$  was precipitated from the remaining solution and mounted for counting.

The 17.5-day  $As^{75}$  was detected by counting beta particles, assumed to be in 62% abundance.

**Bromine**

The NaBr target was dissolved in distilled water and the volume made up to 100 ml. A known aliquot was removed for subsequent chemistry and counting. Hold back carriers of Se, As, Zn, Cu, Ni, and Fe were added. AgBr was precipitated by addition of  $AgNO_3$  solution, dissolved, reprecipitated, washed, and mounted for counting. The 4.4-hour  $Br^{80m}$  and the 17.6-min  $Br^{80}$  were detected by counting beta particles. The abundance of beta particles was assumed to be 95% in all  $Br^{80}$  samples and for  $Br^{80m}$  in which  $Br^{80}$  was present in equilibrium.

**Zirconium**

It was necessary to achieve a fast separation of niobium, zirconium, and yttrium from the target. The zirconium target was dissolved in concentrated HF with the addition of hold back carriers of Sr, Br, Na, Se, As, Ge, Zn, Cu, Ni, and carriers for niobium and yttrium.  $YF_3$  was immediately removed from solution.  $BaCl_2$  solution was added and  $BaZrF_6$  was precipitated and washed with water twice. Niobium was removed by passing the solution through a Dowex-2 ion exchange column. The Nb was washed from the column with 3*N* HCl. The niobium was purified and precipitated by adding the column eluate to a solution of concentrated HCl with  $NH_4OH$  and tartaric acid. One percent cupferron solution was added and the solution filtered, washed with 10% HCl, and finally washed with water.

The zirconium was purified by solution of the  $BaZrF_6$  in concentrated  $HNO_3-H_3BO_3$  solution.  $Zr(OH)_4$  was then precipitated by addition of  $NH_4OH$ . The precipitation cycle was repeated twice and the zirconium converted to  $BaZrF_6$ . This was redissolved,  $Zr(OH)_4$  reprecipitated and ignited to  $ZrO_2$  and mounted for counting.

The  $YF_3$  was dissolved in  $HNO_3$  and saturated  $H_3BO_3$ , and Zr carrier was added. HF was added to precipitate  $YF_3$ . This was saved for the determination of the yttrium chemical yield. La carrier was added to the solution and  $LaF_3$  precipitated twice. The solution was made basic with  $NH_4OH$  to precipitate  $Zr(OH)_4$ . The Zr from yttrium was then converted to  $ZrO_2$  and mounted for counting.

The 10.5-minute  $Y^{95}$  was determined by counting its 65-day  $Zr^{95}$  daughter. This latter nuclide was assumed to be associated with beta particles in 100% of the disintegrations. The 35-day  $Nb^{95}$  was also assumed to decay by beta particles in 100% of the disintegrations.

**Iodine**

The KI target was dissolved in water and distilled by the addition of concentrated  $H_2SO_4$  collecting the

distillate in 1*N* NaOH. The distillate was made acid with H<sub>2</sub>SO<sub>4</sub> and the iodine oxidized with NaNO<sub>2</sub>. I<sub>2</sub> was extracted with CCl<sub>4</sub> and back extracted with NaOH solution. The extraction was repeated 3 times. Finally I<sub>2</sub> was reduced with SO<sub>2</sub> in acid solution. The SO<sub>2</sub> was removed from the solution by blowing air through it and then AgI was precipitated by the addition of AgNO<sub>3</sub>. The 13.3-day I<sup>126</sup> was determined by assuming that 45.3% of the disintegrations involved beta particles.

### Cerium

The CeO<sub>2</sub> target was dissolved in concentrated HCl with the addition of several crystals of KI to aid the dissolution. The I<sub>2</sub> produced was reduced with 30% H<sub>2</sub>O<sub>2</sub>. Ten milligrams of La carrier were added, the solution made to a known volume, and a known aliquot was reserved for the cerium determination by standard analytical methods. The cerium and lanthanum were precipitated by addition of HF, dissolved in boric and nitric acids, and reprecipitated as hydroxides. Solution of the hydroxides in 8*N* HNO<sub>3</sub> was followed by oxidation of the cerium with BrO<sub>3</sub><sup>-</sup> and extraction of the Ce in the plus-four state in methyl isobutyl ketone according to the procedure outlined by Glendenin *et al.*<sup>35</sup> The 3.7-hour La<sup>141</sup> was allowed to completely decay to the 33.1-day Ce<sup>141</sup> daughter. The latter activity was chemically isolated from the lanthanum fraction and the Ce<sup>141</sup> counted. The Ce<sup>141</sup> was assumed to be associated with beta particles in 100% of its disintegrations.

### Tungsten

The WO<sub>3</sub> target was dissolved in 2*N* NaOH and diluted to a known volume. A 50% aliquot was taken for the tantalum chemistry, the remaining portion re-

served for tungsten. The tantalum procedure is as follows: Tantalum, rare earth, and zirconium carriers were added and the rare earth fluorides were precipitated by addition of HF. The solution was scavenged with a rare earth precipitation two more times. The tantalum was then extracted into methyl isobutyl ketone from a nitric acid, HF solution. The organic layer was washed two times with 6*N* HNO<sub>3</sub> and HF. The tantalum was back extracted with saturated H<sub>3</sub>BO<sub>3</sub> solution. Tungsten carrier was added and then the solution was made strongly acid with HNO<sub>3</sub> to precipitate WO<sub>3</sub>. This was mounted for counting the 75.8-day W<sup>185</sup> daughter of 50-minute Ta<sup>185</sup>.

The tungsten fraction was purified by adding Zr, rare earth, and Ta carriers and then scavenging with a Fe(OH)<sub>3</sub> precipitation. The Fe(OH)<sub>3</sub> precipitation was repeated twice. The solution was made strongly acid with HNO<sub>3</sub> and heated in a water bath. WO<sub>3</sub> precipitated and was dissolved in 2*N* NaOH. The acid precipitation of WO<sub>3</sub> and dissolution in NaOH was repeated two times. The final precipitate was washed with water and mounted for counting. The 0.430-Mev beta particles of 75.8-day W<sup>185</sup> were assumed to be present in 100% of the disintegrations.

### Rhenium

The rhenium targets were dissolved in HNO<sub>3</sub> together with hold back carriers of Ta, W, rare earths, and Zr. Re<sub>2</sub>S<sub>7</sub> is precipitated by passing H<sub>2</sub>S through the boiling solution for 15 minutes. The Re<sub>2</sub>S<sub>7</sub> was dissolved in 1 M NaOH and one drop of 30% H<sub>2</sub>O<sub>2</sub>. An Fe(OH)<sub>3</sub> scavenge was then carried out twice. Re<sub>2</sub>S<sub>7</sub> was then reprecipitated from 12*N* HCl solution and the precipitate was washed with CS<sub>2</sub> and water. The precipitate was mounted for counting. The 88.9-hour Re<sup>186</sup> was assumed to be associated with beta particles in 92% of the disintegrations.

<sup>35</sup> L. E. Glendenin, E. P. Steinberg, K. F. Flynn, and R. F. Buchanan, *Ann. Chem.* **27**, 59 (1955).