Radiochemical Studies of the (p,pn) Reaction in Complex Nuclei in the 80–450-Mev Range*†

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Excitation functions for the (p,pn) reaction have been determined in the energy range 82 Mev to 426 Mev for the target nuclei F^{19}, Cu^{65}, and Au^{197}. The absolute values are based on the excitation function for the reaction $C^{12}(p,pn)C^{11}$. These excitation functions exhibit a general decrease with energy and the cross sections lie between 122 mb for Au^{196} at 82 Mev and 23 mb for F^{18} at 426 Mev. An excitation function for the reaction $Al^{27}(p,3pn)Na^{24}$ is also presented. The results of Monte Carlo nuclear cascade calculations have been used to predict these (p,pn) excitation functions. The theoretical results are compared with experimental results of this and other reports. Agreement is obtained for the $F^{19}(p,pn)F^{18}$ excitation function and an extrapolation of the Monte Carlo results. The theoretical excitation functions are about one-half the experimental results for the $Cu^{65}(p,pn)Cu^{64}$ reaction and about one-third the experimental results for $Au^{197}(p,pn)Au^{196}$. The effects on the Monte Carlo calculations of a variation in the radius parameter are examined.

I. INTRODUCTION

NUCLEAR reactions induced by particles of kinetic energy of the order of 100 Mev and greater are generally regarded as proceeding in two steps. First, the bombarding particle passes through the nucleus sometimes initiating a cascade and sometimes, although less frequently, continuing through the nucleus with no interaction. Second, the residual nucleus de-excites by the emission of one or more particles (protons, neutrons, alpha particles, deuterons, etc.) and/or gamma rays. Attempts have been made by several authors[1–8] to calculate absolute cross sections for nuclear reactions involving complex nuclei using these ideas.

The (p,pn) reaction is a particularly suitable process for such calculations because, in this reaction, both stages are relatively simple. There have been numerous previous studies of such reactions. For example, Fig. 1 shows, for the $C^{12}(p,pn)C^{11}$ reaction, for the energy range 75–500 Mev, the recently reported cross sections of Crandall *et al.*,[9] their recalculations of the values of Aamodt,[10] and the results of Rosenfeld,[11] and Symonds *et al.*[12]

A summary of work done on (p,pn) reactions in this energy range in more complex nuclei[3,4,8,13–29] is presented in Table I. In many of the reported results, there has been little or no attempt to evaluate counting efficiencies carefully, and there is considerable uncertainty in the results. Further, with the exception of the reaction $C^{12}(p,pn)C^{11}$, there are very few complete ex-

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TABLE I. Summary of literature cross sections (80–500 Mev).

Reaction	Proton energy Mev	Cross section mb	Reference
$N^{14}(p,pn)N^{13}$	400	5.6	8
$F^{19}(p,pn)F^{18}$	280	28	8
	380	27	8
	400	26	8
	420	23	12
	450	22	13
$Na^{23}(p,pn)Na^{22}$	80	62	14
	100	60	14
$Sc^{45}(p,pn)Sc^{44m}$	80	100	15
	100	90	15
$Sc^{45}(p,pn)Sc^{44g}$	80	40	15
	100	35	15
$Fe^{54}(p,pn)Fe^{53}$	400	48, 45	8
$Co^{59}(p,pn)Co^{58m}$	80	96	16
	80	95	15
	100	80	16
	100	70	15
$Co^{59}(p,pn)Co^{58g}$	80	101	16
	80	60	15
	100	80	16
	100	50	15
$Co^{59}(p,pn)Co^{58}$	100	363	17
	170	185	17
	240	120	17
	370	121	18
$Ni^{58}(p,pn)Ni^{57}$	400	47, 52	8
$Cu^{63}(p,pn)Cu^{62}$	80	150	19
	100	120	19
	400	73, 64	8
$Cu^{65}(p,pn)Cu^{64}$	80	200	19
	90	190	19
	90	126	20
	100	150	19
	190	77	20
	280	69	8
	340	73	21
	380	68, 83	8
	400	67, 73, 71	8
	485	71	22
$Zn^{64}(p,pn)Zn^{63}$	100	120	3
	400	71, 66	8
$Ga^{69}(p,pn)Ga^{68}$	100	193	3
$As^{75}(p,pn)As^{74}$	100	102	23
	103	70	4
	170	90	4
	380	55	23
$Br^{81}(p,pn)Br^{80m}$	80	105	15
	100	90	15
$Br^{81}(p,pn)Br^{80g}$	80	85	15
	100	72	15
$Y^{89}(p,pn)Y^{88}$	100	150	24
	150	94	24
	180	120	24
	240	93	24
$Mo^{100}(p,pn)^{a}Mo^{99}$	280	79	8
	380	65	8
	400	79, 73	8
$Ag^{107}(p,pn)Ag^{106}$	340	130	25
$I^{127}(p,pn)I^{126}$	100	126	26
	170	64	26
	300	53	26
	480	70	26
$Cs^{133}(p,pn)Cs^{132}$	80	1120	27
	100	890	27
	150	570	27
	240	59	27
$Ce^{142}(p,pn)Ce^{141}$	380	87	28
	400	86	28
$Ta^{181}(p,pn)Ta^{180m}$	280	44	8
	380	62, 53	8
	400	46	8
$Th^{232}(p,pn)Th^{231}$	340	68	29
$U^{238}(p,pn)U^{237}$	100	93	29
	150	73	29
	200	68	29
	300	81	29
	340	85	29

[a] Includes contribution from $Mo^{100}(p,2p)$ reaction.

citation curves in the region between 100 Mev and 400 Mev. It was thought desirable, therefore, to measure, for different mass numbers, some (p,pn) cross sections accurately in the energy range where the theory should apply and meson production does not complicate the situation too much. For this reason, excitation functions for the reactions $F^{19}(p,pn)F^{18}$, $Cu^{65}(p,pn)Cu^{64}$, and $Au^{197}(p,pn)Au^{196}$ have been determined in the energy range 82 Mev to 426 Mev. Cross sections were determined relative to the excitation function for the reaction $C^{12}(p,pn)C^{11}$. The radioactivities of F^{18}, Cu^{64}, Au^{196}, C^{11}, and Na^{24} were used to obtain the cross sections. The values obtained are compared with results obtained from cascade calculations combined with evaporation theory. As a byproduct of this study, the excitation function for the $Al^{27}(p,3pn)Na^{24}$ reaction has been determined, again based on the $C^{12}(p,pn)C^{11}$ cross section.

II. EXPERIMENTAL PROCEDURE

Targets were irradiated in the internal proton beam of the University of Chicago synchrocyclotron. Nominal proton energies were varied from 90 Mev to 450 Mev by suitable choice of the target radius.[30] The energy decrement due to radial oscillations has been estimated by Koch[31] and Rosenfeld and Warshaw.[32] These energy decrements ranged from 17 Mev at a nominal 90 Mev to 48 Mev at a nominal 450 Mev. All proton energies given in this paper are average energies calculated from the data of Koch.[31] Irradiations of the targets were performed for times varying from a few synchrocyclotron pulses to ten minutes, depending on the targets used.

In any given irradiation the extent of the (p,pn) reaction in a target material was compared to the production of Na^{24} from aluminum monitors bombarded simultaneously. The targets were constructed of three foils of target material, each 10 to 25 mg/cm^2 thick, surrounded on either side by three aluminum foils, each 4 mg/cm^2 thick. The target materials were poly-

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ethylene, Teflon, copper, and gold. Three foils were used for each group to compensate the center foil of the group for recoil losses and to protect it from the introduction of extraneous activities from recoils from adjacent foils. In the case of copper, only one foil was used due to the difficulty in processing, after irradiation, a target containing three copper foils. Here the recoil losses of Cu^{64} from the copper foil were investigated and found to be less than one percent of the total Cu^{64} activity at any energy. They were neglected in the cross section calculations.

After irradiation, the radially innermost 0.08 inch (leading edge) of the composite target was sliced off and discarded. The target was then cut into two adjacent sections each $\frac{3}{8}$ inch square. The first section was then $\frac{3}{8}$ inch closer to the center of the cyclotron than the second section. The center foils of each group of aluminum or target materials were weighed and then used for activity determination. The aluminum and Teflon foils were mounted directly on aluminum cards and the Na^{24} in the aluminum and the F^{18} in the Teflon were counted on the top shelf (two mm from the window) of methane flow end-window proportional counters.[33] In one experiment the annihilation gammas from the F^{18} in the Teflon were counted on a single channel scintillation counter. The polyethylene samples were mounted in the same way and the C^{11} annihilation gammas counted on the scintillation counter.

The copper targets were dissolved in nitric acid and copper was radiochemically separated from the other spallation products. In some cases a nickel sample was also isolated for use as a check on neutron induced activities. The gold foils were dissolved in aqua regia and radiochemically purified from the spallation and fission products of gold. Some platinum samples were also isolated as checks on neutron induced activities. The radiochemical procedures used for isolating copper and nickel radioactivities from the copper targets were those of Koch.[31] The others are given in the Appendix. The copper samples were normally counted on the proportional counters. Two samples however, were, in addition, counted in the annihilation region on the scintillation counter. All annihilation gamma counting was done with sufficient absorber to absorb all the positrons.

The aluminum foils were counted on the second and third days after the bombardment when all other activities in the aluminum were less than one percent of the 15.06-hr Na^{24}. In general, all the radioactivities were measured at times when longer or shorter periods present contributed less than one percent to the counting rate. The half-lives and absorption characteristics were consistent with literature values except in the cases of F^{18} and Au^{196}.

For F^{18}, it was found in all cases that the half-life was somewhat shorter than the accepted value of 112 minutes.[34] Reported values of this half-life range from 107 to 115 minutes.[35–39] More recently, Jarmie[40] and Rayburn[41] have reported values of 111 minutes. The half-life used in this work was 110.2 minutes. It was determined as follows. Some radiochemically pure F^{18} was prepared by the irradiation of lithium fluoride with 420-Mev protons and chemical separation and purification of the fluoride. Two samples were prepared and counted on the proportional counter through various thicknesses of aluminum absorber. No trend of half-life with absorber thickness was seen, and a least squares analysis of the data yielded the value of 110.2 ± 0.2 minutes.

Au^{196} was identified by its gamma-ray spectrum. In this case, the half-life was found to be longer than the reported value of 5.6 days.[42,43] The half-life used in this work is 6.10 ± 0.02 days. It was determined from the decay curves of six samples followed for about eight half-lives. The samples were prepared by bombardment of gold with protons of various energies in the range 82 Mev to 426 Mev. An additional check on the half-life was performed by preparing another gold sample and following it for several half-lives, then performing an additional chemical purification step. No changes in specific activity and half-life were observed. Spectra in the range 0.1 to 1 Mev were taken on the sample about once every three days for the duration of the experiment. These spectra were plotted on semilog paper, and no change in the shape of the spectrum was seen after the first few days. At first there was a little activity in the range 0.5–1.0 Mev, but this died out rapidly, and was probably due to Au^{194} (40 hours).

III. TREATMENT OF DATA

All cross sections reported in this paper were measured relative to that of the $C^{12}(p,pn)C^{11}$ reaction using the Na^{24} produced in the aluminum foils to monitor the beam intensity from irradiation to irradiation. The values used for the (p,pn) cross section for C^{12} are those given by the solid curve of Fig. 1. In order to get numerical values for the cross sections of interest, there is needed, in addition, the counting efficiency for the nuclide investigated relative to that of the annihilation gammas of C^{11}. This was formally done by calculating disintegration rates for the counting conditions actually

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used, although it will be clear that in many cases the conversion factors cancel out.

The efficiency of measurement of annihilation gamma rays in our single channel spectrometer was determined with a Na^{22} solution standard obtained from the National Bureau of Standards. The standardization of this sample was given in terms of positron emission rate. The observed count was corrected for the contribution from the 1.277 Mev gamma of Na^{22} in the following manner. A standard Co^{60} source was measured under the same conditions as were the positron samples. This source has 1.17 Mev and 1.33 Mev gammas, which permit the estimation of the Compton contribution with an error of about two percent. The number of 1.277-Mev gammas per positron in the decay of Na^{22} was taken as the average of three literature values,[44–46] 1.12 γ/β^+. The resulting correction is about twelve percent of the annihilation counting rate, and hence is known sufficiently accurately.

The excitation functions of F^{18} and Cu^{64} were established on a relative basis by proportional counting. The Cu^{64} samples were corrected for the variation of the counting rate with sample weight by means of the scattering curve of Koch[31] who used identical sample preparation and counting conditions. These excitation functions of F^{18} and Cu^{64} were converted to an absolute basis by counting the annihilation radiation of several samples. The fraction of disintegrations proceeding by positron emission were taken as 0.97 for fluorine[47] and 0.19 for copper.[34] The disintegration rate of the Cu^{64} sample was also calculated from the proportional counting data by the method of Koch,[31] who measured the beta detection efficiency of Cu^{64} under the same conditions. The two methods agreed to within five percent.

The disintegration rate of the Au^{196} samples was determined from the photopeak efficiencies of its 331 kev and 354-kev gamma rays and the number of these gamma rays per disintegration. Branching ratios and conversion coefficients used are those of Thieme and Bleuler.[48] Photopeak efficiencies were determined from solution standards of I^{131}(364 kev) and Au^{198}(412 kev) obtained from the National Bureau of Standards and the assumption that photopeak efficiencies vary exponentially with gamma-ray energy.[49]

The counting efficiency of the Na^{24} in the proportional counter under our conditions was determined by comparison with that of Koch[31] and was found to be 0.538.

Cross sections for the reaction $Au^{197}(p,pn)Au^{196}$ were calculated on the assumption that the reaction proceeded directly to the ground state without the formation of any 14-hr Au^{196m}. Because of the time of radiochemical isolation of the gold, the error introduced by this assumption is small; if all the Au^{196} were formed via Au^{196m}, the calculated cross section would be 10% too high.

IV. EXTRANEOUS CONTRIBUTIONS TO THE VARIOUS ISOTOPES

Secondary protons or neutrons can produce the product of the (p,pn) reaction by (p,pn) or $(n,2n)$ processes. In the case of $Cu^{65}(p,pn)Cu^{64}$, the product may also be made by the reaction $Cu^{63}(n,\gamma)Cu^{64}$. Koch[31] found secondary contributions to the reaction $Ni^{64}(p,n)Cu^{64}$ in one-mil nickel foil to the extent of 10% at 416 Mev and smaller contributions at lower energies. Since the (p,pn) cross section is much larger than the (p,n) cross section, no appreciable secondary contribution to the (p,pn) cross section was anticipated. On the basis of Koch's work, it was estimated that secondary contributions from protons would increase about 50% if the number of target foils was doubled. Experiments with such targets at 426 Mev showed no increase in cross section, and it is concluded that contributions from particles born within the target are negligible.

TABLE II. Summary of experimental results.

Average proton energy Mev	Average cross sections (mb) $F^{19}(p,pn)F^{18}$	$Cu^{65}(p,pn)Cu^{64}$	$Au^{197}(p,pn)Au^{196}$	$Al^{27}(p,3pn)Na^{24}$
82	46.0±1.5	108.4±4.2	121.6±9.8	11.7±0.4
110	38.5±1.3	93.6±3.7	···	10.9±0.4
134	···	74.6±2.9	···	10.9±0.4
139	···	···	91.2±7.4	···
144	30.3±1.0	···	···	···
168	···	65.8±2.6	···	10.1±0.3
176	25.8±0.8	···	···	···
196	24.7±0.8	64.3±2.5	···	9.92±0.3
210	···	···	73.6±6.0	···
225	24.3±0.8	57.9±2.3	···	10.1±0.3
263	24.6±0.8	55.0±2.1	···	11.2±0.4
282	···	···	71.0±5.7	···
330	23.9±0.8	55.9±2.2	···	11.7±0.4
426	23.1±0.8	51.6±2.0	70.5±5.7	11.9±0.4

The possible contribution from neutrons abundantly present in an accelerator area was investigated by examining the formation of the (p,pn) products as a function of location in the target. Within the target, the proton flux decreases exponentially with increasing distance from the center of the machine; the first section of the targets that were used usually had five times the activity that the second section had. The neutron flux is not expected to vary appreciably with the radius. The variation with radius of the (p,pn) products was compared to the variation with radius of a species having a high-energy threshold and which would not be produced by low-energy neutrons (Ni^{57} from copper and Pt^{188} from gold). The data obtained set an upper limit of two percent to the fraction of

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(p,pn) activity being due to neutrons in the first section of the target and ten percent in the second section.

The neutron pick-up or (p,d) reaction can also contribute to the (p,pn) products. Our experiments clearly include the contributions from this process. Selove[50] has presented data for the pick-up process for 95-Mev protons on carbon. These data can be used to estimate the cross section for deuteron production leading to C^{11} to be about 8.2 mb. Crandall[9] reports the cross section for $C^{12}(p,pn)C^{11}$ to be 65 mb, and hence the (p,d) process in this case contributes less than twelve percent to the (p,pn) products at 95 Mev. The (p,d) reaction should be a smaller fraction for targets of higher mass since it is thought to occur on the nuclear surface. Evidence for this is presented by Morrison[51] for the (n,d) reaction. Heidmann[52] expects theoretically that the pick-up cross section should decrease about as the inverse sixth power of the bombardment energy. Hence, it is felt that (p,d) processes do not contribute appreciably to the (p,pn) products in the 100–450-Mev range.

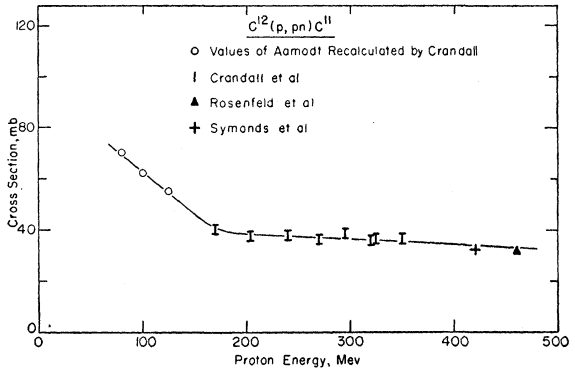

FIG. 1. Excitation function for the reaction $C^{12}(p,pn)C^{11}$. The smooth curve is that used as a basis for calculating cross sections in this work.

V. EXPERIMENTAL RESULTS

A summary of the experimental results is given in Table II. This lists for the different proton bombarding energies (column 1), the average values of the (p,pn) cross sections obtained for F^{19}, Cu^{65}, and Au^{197} (columns 2, 3, and 4). Also given are the data we obtained for the formation cross section of Na^{24} from Al^{27} (column 5). At 82 Mev and 426 Mev, these average cross sections were obtained from the results of two or three bombardments. Each bombardment yielded two values of the cross section, one from each section of the target. At intermediate energies there was ordinarily only one bombardment. The two cross sections from a given bombardment were usually less than two percent apart

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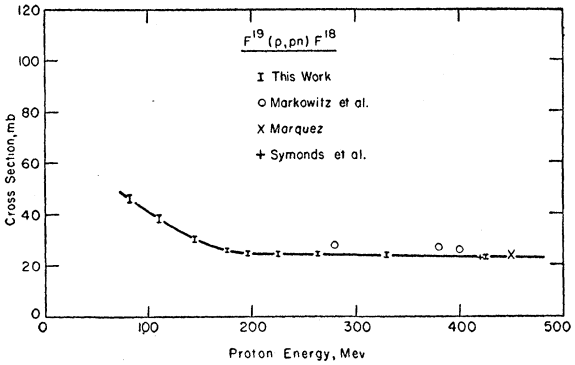

FIG. 2. Experimental cross sections as a function of energy for the reaction $F^{19}(p,pn)F^{18}$. Comparison with other published work is indicated.

for fluorine runs and four percent apart for copper runs. The gold runs sometimes gave a spread of 8%, although at 282 and 426 Mev, agreement was better than one percent. In almost all cases the cross sections calculated from different bombardments at the same energy agreed within two percent. The errors indicated in Table II are statistical combinations of the standard deviations of the errors in the individual factors determining the ratios of the cross section to that of the $C^{12}(p,pn)C^{11}$ reaction. The errors in this cross section are not included.

The numbers listed in Table II are to be found in graphical form in Figs. 2, 3, 4, and 5, together with the literature values from Table I. For the reaction $F^{19}(p,pn)F^{18}$, it will be seen that our results are in excellent agreement with those of Marquez.[13] Agree-

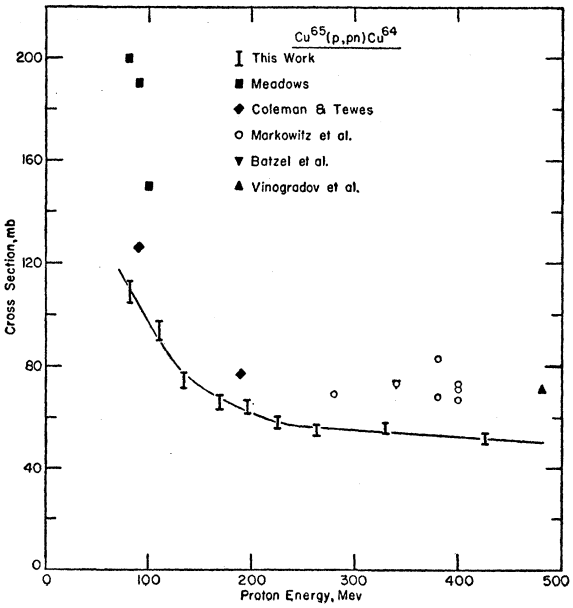

FIG. 3. Experimental cross sections as a function of energy for the reaction $Cu^{65}(p,pn)Cu^{64}$. Comparison with results of other published work is indicated.

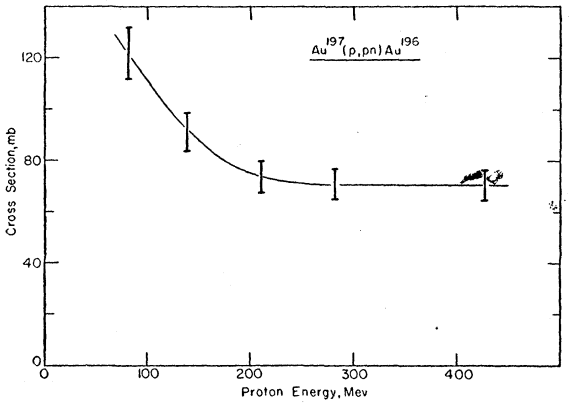

FIG. 4. Experimental cross sections as a function of energy for the reaction $Au^{197}(p,pn)Au^{196}$.

ment with the results of Markowitz *et al.*[8] is not as good, for their later numbers[8] are about 10% high.

In the case of $Cu^{65}(p,pn)Cu^{64}$ the present results are lower than those of almost all previous investigators. The discrepancy with earlier work such as that of Meadows,[19] Batzel,[21] and Vinogradov[22] are probably not to be taken too seriously because counting efficiencies and monitor cross sections were poorly known then. The discrepancies with Coleman and Tewes[20] and Markowitz *et al.*[8] are more serious since comparable care was exercised by these authors to determine counting efficiencies. When these discrepancies became apparent, upon publication of the work of Markowitz *et al.*,[8] a cooperative experiment was arranged with these authors which established that the source of the divergence was in the beta counting efficiency used for Cu^{64}. This counting efficiency, under the conditions used in the present work, was then checked by comparison of the annihilation radiation of a Cu^{64} sample with a new National Bureau of Standards Na^{22} standard. The result was in adequate agreement with the value used in the present work.

Figure 5 shows the excitation function for the reaction $Al^{27}(p,3pn)Na^{24}$ along with that of Hicks,[53] whose cross sections also depend on the absolute $C^{12}(p,pn)C^{11}$ cross sections of Crandall.[9] The agreement is reasonably good, the difference in the two sets of results probably being due to differences in counting efficiencies.

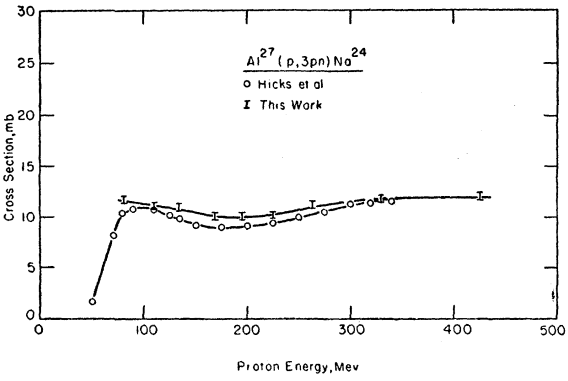

FIG. 5. Excitation function for the reaction $Al^{27}(p,3pn)Na^{24}$. Comparison of experimental results obtained in this work with literature values of Hicks et al. (see reference 53).

VI. THEORETICAL CROSS SECTIONS

On the two-stage theory of nuclear reactions at high energies, the cross sections of such simple processes as the (p,pn) reactions studied here should be calculable by combining the results of the intranuclear cascade and nuclear evaporation theory. The recent cascade calculations of Metropolis *et al.*[6] provide data on the residual nuclei and their excitation energies for protons in the energy range of interest here and target nuclei Al^{27}, Cu^{64}, Ru^{100}, Ce^{140}, Bi^{209}, and U^{235}.

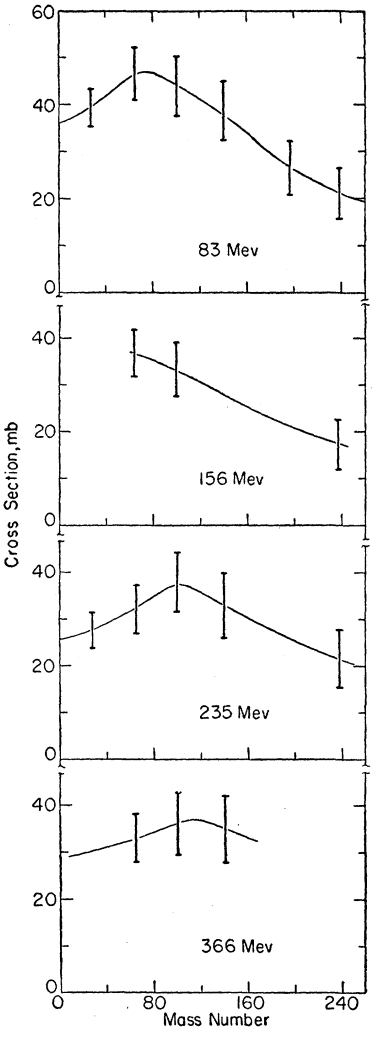

FIG. 6. Theoretical (p,pn) cross sections versus mass number at 83 Mev, 156 Mev, 235 Mev, and 366 Mev.

In performing the evaporation calculations, the binding energies of the actual nuclei involved were used.

[53] H. G. Hicks, P. C. Stevenson, and W. E. Nervick, Phys. Rev. 102, 1390 (1956).

These were taken from Wapstra[54] for aluminum, from Way[55] for copper, from Cameron[56] for ruthenium, cerium and gold, and from Huizenga[57] for uranium. The actual evaporation treatment was based on the formulas summarized by Morrison,[51] with the specific widths chosen being those applicable[51] to a radius parameter of 1.4×10^{-13} cm. The treatment of the odd-even effect on the density of states was likewise that of Morrison.[51]

The results of these calculations are summarized in Fig. 6. This shows, for energies between 83 Mev and 366 Mev for which intranuclear cascade data is available, the dependence of the predicted (p,pn) cross section on the mass number. The errors indicated reflect only the statistical uncertainties connected with the Monte Carlo results. The figure shows that the dependence of the calculated (p,pn) cross sections on mass number is not large. There appears to be a slight

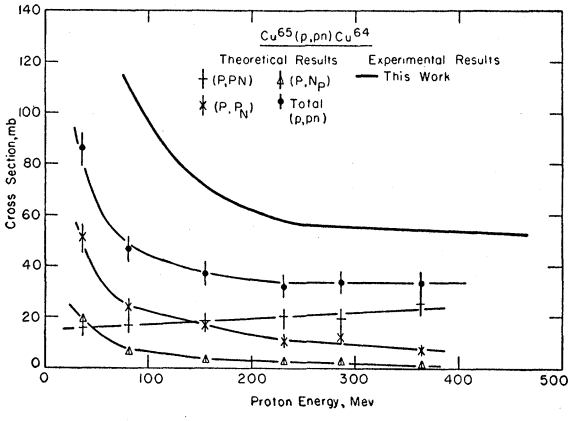

FIG. 7. Comparison of calculations on the $Cu^{65}(p,pn)Cu^{64}$ reaction with experimental results. The contributions of the separate mechanisms to the calculated results are indicated.

maximum in the middle mass region, but its energy dependence is not clear.

Comparison of the results of these calculations with the experimental data reported in this paper are shown for Cu^{65} and Au^{197} in Figs. 7 and 8, respectively. The gold data are compared with a calculation based on the intranuclear cascades computed for Bi^{209}. Indicated in these two figures are the separate contributions to the calculated cross sections of the three possible reaction paths leading to a (p,pn) product:

1. a (P,PN) cascade leaving a nucleus with too little excitation to evaporate any other particles.

2. a (P,P') cascade followed by neutron evaporation. This is indicated in the figures by the symbol (P,P_N').

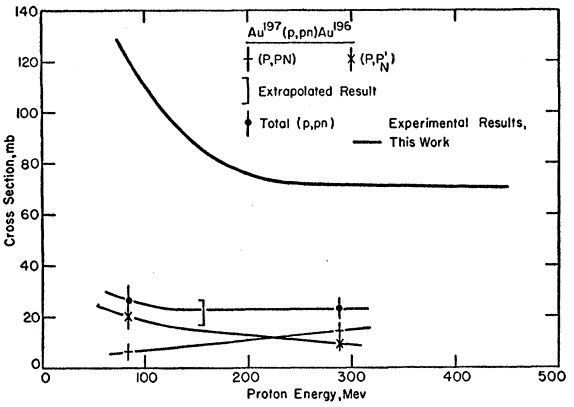

FIG. 8. Comparison of calculations on the $Au^{197}(p,pn)Au^{196}$ reaction with experimental results. The contributions of the separate mechanisms to the calculated results are indicated.

3. a (P,N) cascade followed by proton evaporation. This is indicated in the figures by the symbol (P,N_P).

A theoretical curve for the (p,pn) reaction in fluorine is not presented since no intranuclear cascades were run for any element lighter than Al^{27} and evaporation calculations would also be suspect here. Reasonable extrapolation of the mass trends of Fig. 6, however, lead to predicted (p,pn) cross sections for mass 19 in very good agreement with the experimental data presented in Fig. 2.

Examination of Figs. 7 and 8 as well as comparison of the literature values of Table I with the calculations as summarized in Fig. 6 indicate that, except for the case of F^{19}, the calculated (p,pn) cross sections are lower than experimental ones by factors of between two and four. The energy dependence in the case of the calculated copper curve in this energy range appears to be the same as that of the experimental data, but the calculated gold curve is too flat. In the case of uranium, inclusion of fission competition in the calculation would lower the calculated cross sections even more and make the discrepancy with experimental results greater.

The failure of this type of calculation to predict (p,pn) cross sections has been previously noted by Rudstam[4] for the $As^{75}(p,pn)As^{74}$ reaction at 170 Mev and 103 Mev, and Meadows[3] for the $Cu^{65}(p,pn)Cu^{64}$ reaction at 100 Mev. Both these authors had much more limited nuclear cascade data to use in their calculations. More recently Markowitz *et al.*[8] have shown that this discrepancy is even larger at higher energies: the (p,pn) cross sections found experimentally at 3 Bev are factors of 5 to 25 larger than predicted by evaporation theory and the cascade calculations of Metropolis *et al.*[6]

Because of the very simple nature of the evaporation processes contributing to (p,pn) reactions it is hardly likely that the general systematic lack of agreement with experimental results can be attributed to this part

[54] A. H. Wapstra, Physica **21**, 367 (1955).
[55] K. Way *et al.*, Atomic Energy Commission Report TID-5300, June, 1955 (unpublished).
[56] A. G. W. Cameron, Atomic Energy of Canada Limited, Chalk RiverProject Report CRP-690, March, 1957 (unpublished).
[57] J. R. Huizenga, Physica **21**, 410 (1955).

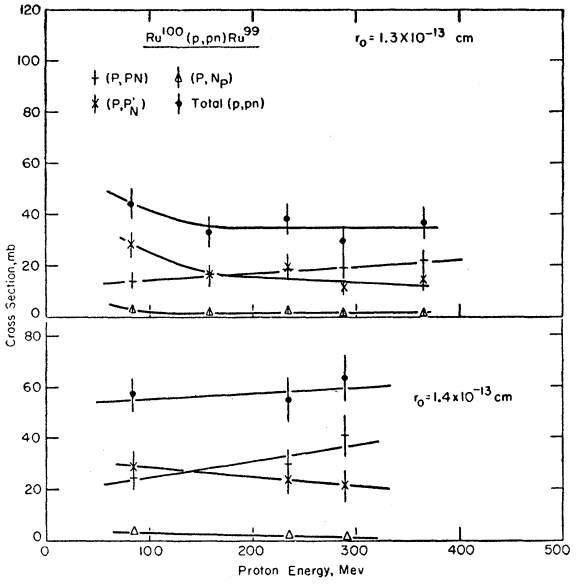

FIG. 9. Comparison of calculations on the $Ru^{100}(p,pn)Ru^{99}$ reaction with radius parameters of 1.3×10^{-13} cm and 1.4×10^{-13} cm.

of the calculation. The reason is rather to be sought in inadequacies in the treatment of the cascade portion of the calculation. The (p,d) process, not taken into account in the calculation, could increase the cross section at the lowest energies, but arguments presented above indicate it should be negligible for complex nuclei in most of our energy range.

It has been suggested[7,21] that this inadequacy of the cascade calculations of Metropolis *et al.*[6,7] to predict correctly the probability of small energy depositions in spite of their success in correlating much of the data on energetic encounters of high-energy particles with complex nuclei, is connected with the use, in these calculations, of a sharp nuclear boundary. Although no cascade calculations based on a more realistic nuclear density distribution are available, Metropolis *et al.*[6] did study some cascades produced in Ru^{100} by protons using both $r_0=1.3\times10^{-13}$ and 1.4×10^{-13}. The results of these latter cascades have been combined with an evaporation calculation carried out in a similar way to that described above. The result for the two radii parameters are presented in Fig. 9. It is seen that the calculated (p,pn) cross section from the $1.4\times10^{-13}A^{\frac{1}{3}}$ cascades is increased to almost 60 mb for Ru^{100} from a value of about 35 mb calculated from cascades with the smaller radius. The energy dependence is flatter in the former case, but pick-up reactions at the lower energies might make it more realistic. It should be remembered, however, that the average excitation energies for all cascades calculated from this larger radius parameter are only 75–80% of those with the smaller parameter, and so would lead to discrepancies in the predictions of the yields of more typical high-energy reactions.[6]

These results thus lend some encouragement to the idea that a more realistic cascade calculation involving perhaps even a denser central part to the nucleus than that used by Metropolis *et al.* but also a diffuse edge extending to larger distances than they used may provide the proper fraction of low-energy events to explain simple reactions of the (p,pn) type as well as more typical high-energy events.

APPENDIX

Chemical Procedures

The radiochemical procedures used for separating copper and nickel samples from the other spallation products of copper are those of Koch.[31] The separations of platinum and gold samples from the spallation and fission products of gold are described below. The terms "acid" and "acidic" refer to hydrochloric acid with the hydrogen ion concentration being about $2M$.

Gold

The gold target foils were dissolved in aqua regia. Gold carrier and platinum carrier were added, and the solution made acidic. Holdback carriers of iron, ruthenium, rhodium, palladium, iridium, mercury, arsenic, and molybdenum were added. The gold was extracted into ethyl acetate, the organic layer being washed twice with acid. The aqueous layer was reserved for the platinum chemistry. The ethyl acetate layer was boiled nearly to dryness, acid was added, and the remaining ethyl acetate boiled off. Sulfur dioxide was passed into the hot acid solution to precipitate metallic gold. The gold was dissolved in aqua regia and recycled twice using the same procedure. The final precipitate was metallic gold.

Platinum

To the platinum solution from the gold procedure was added additional gold carrier. This was then extracted with ethyl acetate. Excess sodium hydroxide was added and the precipitate centrifuged off. The supernate was made acidic. Solid hydroxylamine hydrochloride and solid stannous chloride were added, and the red $PtCl_4^{=}$ complex was extracted into ethyl acetate. The platinum was recovered into the aqueous layer by evaporating off the ethyl acetate, and the sample recycled twice with the addition of iridium, ruthenium, rhodium, and ferric holdback carriers. The final precipitate of platinum metal was prepared by the addition of powdered magnesium to an acid solution of platinum.