Study of (p,n) Reactions between 100 and 400 MeV*†

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Excitation functions of (p,n) reactions have been determined for proton energies between 100 and 400 MeV using targets of Sc^{45}, Ni^{64}, Rh^{103}, Te^{130}, and W^{186}. The energy dependence of the cross sections was found to vary from $E^{-1.31}$ to $E^{-0.75}$ over this range of mass numbers. This variation is apparently related to the increase in attenuation of the incident and outgoing nucleon waves with increasing target mass number. The experimental cross sections were compared with Monte Carlo cascade calculations based on a simple model which did not take refraction of the incident protons into account and which made use of a square-well nuclear potential. Serious deviations were observed between the calculated and the experimental energy dependencies, indicating the necessity of employing a more sophisticated model than one based on simple intranuclear nucleon-nucleon cascades.

INTRODUCTION

NUCLEAR reactions induced with protons of energies equal to or greater than 100 MeV are currently regarded as proceeding through a two-step mechanism.[1,2] In the first step the incident particle initiates a direct fast cascade of intranuclear nucleon-nucleon collisions. The second step involves the de-excitation of the resulting residual nucleus by emission of particles and/or electromagnetic radiation. Attempts to calculate absolute spallation cross sections taking into account both steps of this mechanism have been made by several authors.[3–8]

In the light of this two-step mechanism, the (p,n) reaction is particularly interesting in that severe energetic restrictions are imposed on the system. The direct cascade is limited to the specific case of the ejection of a single neutron and the capture of the incident proton. Meson interactions can be considered negligible at energies less than about 400 MeV. The second, or evaporation step, must be restricted to electromagnetic de-excitation. An alternative mechanism would be the formation of a compound nucleus from which a single neutron would be evaporated. Such a mechanism need not be considered because of the large excitation energy that would be deposited into the compound nucleus.

In view of this two-step mechanism, the cross section of (p,n) reactions should be related, as a first approximation, to the probability of two-body breakup by a pure nucleon knock-out interaction. In actuality, it is more reasonable to assume that the mechanism involves charge-exchange scattering processes in which the incident proton is converted into a neutron while traversing the nuclear matter. Valentin *et al.*[9] have interpreted their measurements of the cross section of (p,n) reactions on light nuclei ($A \leqslant 19$) at 155 MeV in terms of charge-exchange scatterings in which the selective excitation in the (p,n) products of isobaric analog states of the ground and low-lying states of the target nucleus is enhanced. In this case, severe energetic restrictions are still imposed since the hole created in the Fermi sea of neutrons and the orbital occupied by the newly created proton, together must not produce residual excitation energies so large that particle emission becomes probable.

In this paper are reported the results of cross-section measurements of the (p,n) reaction on Sc^{45}, Ni^{64}, Rh^{103}, Te^{130}, and W^{186} with protons of energy between 130 and 420 MeV.

EXPERIMENTAL

Targets were bombarded in the internal proton beam of the Carnegie Institute of Technology synchro-cyclotron at energies from 130 to 420 MeV obtained by appropriate radial placement of the target. The uncertainty of the proton energy has been estimated as being less than $\pm 5\%$ throughout this energy range. Irradiations were of 10- to 30-min duration depending on the nature of the particular system being studied.

Metallic foil targets were employed when possible. A scandium oxide-impregnated epoxy resin foil was utilized in the case of scandium. Nickel foil was prepared by the electro-deposition from $NiSO_4$ and NH_4Cl solution of nickel onto a chromium-coated aluminum foil, from which the latter was subsequently stripped away. Tellurium was electroplated from an electrolytic

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TABLE I. Details of targets.

Target nuclide	Isotopic abundance (%)	Form bombarded	Purity	Supplier
Al^{27}	100	0.001 in. and 0.003 in. metallic foil	99.99%	a
Sc^{45}	100	0.002 in. oxide impregnated epoxy resin foil	"Specpure"	b
Ni^{64}	1.08	0.001 in. metallic foil	99.8%	c
Rh^{103}	100	0.002 in. metallic foil	99.99%	d
Te^{130}	34.48	0.003 in. electropolated onto 0.001 in. nickel foil	99.999%	e
W^{186}	28.4	0.005 in. metallic foil	spectrographically pure of metals heavier than W	f

[a] Aluminum Company of America.
[b] Johnson, Matthey, Ltd.
[c] Baker and Adamson, General Chemical Division, Allied Chemical Dye Company.
[d] J. Bishop Company.
[e] American Smelting and Refining Company.
[f] General Electric Company.

bath consisting of HF, TeO_2, and H_2SO_4 onto a nickel backing foil. A list of the target materials and their suppliers is given in Table I.

After irradiation the targets were dissolved in the presence of appropriate carriers and chemically purified using adaptations of standard radiochemical procedures.[10] Chemical yields were determined gravimetrically, except in the case of rhenium which was determined spectrophotometrically.

In all cases, cross sections were measured relative to the cross section of the $Al^{27}(p,3pn)Na^{24}$ monitor reaction, using the values suggested by Cumming.[11] The targets therefore always consisted of a 3-mil aluminum foil, sandwiched between two aluminum guard foils, all downstream from the target foil.

Disintegration properties of the product nuclides studied were taken from Strominger *et al.*[12] and from the Landolt-Börnstein compilation.[13] Radiations were detected with end-window methane-flow β-proportional counters, 2-in.×2-in., and 3-in.×3-in. NaI(Tl) crystals connected to a Baird-Atomic single-channel pulse-height analyzer and β-γ coincidence circuitry. Stability of the proportional system was within 3% over the period in which measurements were made. The beta detector and associated electronics was calibrated by determining the absolute disintegration rate of Na^{24} samples produced in aluminum by the standard coincidence method and comparing the counting rates of these samples with those of standard Radium D-E Sources. These two methods were in agreement to within 3%. Counting efficiency factors for Ti^{45}, Cu^{64}, I^{130}, and Re^{186} were estimated by conventional methods. Air, detector window, and source absorption factors were calculated from the empirically determined formula of Gleason *et al.*[14] Saturation backscattering factors were taken from the compilation by Burtt[15] and self-absorption and self-scattering factors were estimated from the curves of Nervik and Stevenson.[16] The disintegration rate of Pd^{103} was determined by measuring the 20-keV x rays by a thin crystal detector. This latter was calibrated by comparison with a standardized Ru^{103} source in which the 495-keV γ radiation of Ru^{103} was measured with a calibrated 3-in.×3-in. NaI(Tl) crystal detector.

A least-squares analysis of all the decay curves was performed with a digital computer, with the exception of Pd^{103} and a few of the Cu^{64} and I^{130} samples, which were resolved graphically.

RESULTS

The experimentally determined (p,n) reaction cross sections are presented in Table II. The uncertainties listed are standard deviations from the average for identical determinations. In addition there are systematic uncertainties of about ±10% for the monitor cross section, ±5% for target alignment, ±5% for decay curve resolution, and about ±3% for chemical yield determination. The uncertainty in the counting efficiency factors was about ±10%, except for Pd^{103} which was about ±8%. The overall uncertainty, taken to be the geometric mean of these individual uncertainties, has a value of about ±17%.

The (p,n) reaction should be particulary sensitive to secondary protons since it has very large cross sections at low energies. Koch[17] has measured a contribution to the $Ni^{64}(p,n)Cu^{64}$ cross section of a few tenths of a millibarn per 100 mg/cm² of target. A similar study in this laboratory on the $Zr^{90}(p,n)Nb^{90}$ system

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TABLE II. Experimental (p,n) cross sections in millibarns.

Reaction	Proton bombarding energy (MeV) 130	205	270	298	396	423
$Sc^{45}(p,n)Ti^{45}$	5.05±0.57 (3)[a]	2.32±0.07 (3)		1.44±0.32 (4)	1.19±0.13 (4)	
$Ni^{64}(p,n)Cu^{64}$	6.10±0.9 (6)	3.05±0.5 (5)		2.37±0.4 (5)	1.76±0.2 (6)	
$Rh^{103}(p,n)Pd^{103}$	6.14±0.25 (3)	3.54±0.12 (4)		3.00±0.12 (4)	2.30±0.25 (4)	
$Te^{130}(p,n)I^{130}$	7.68±0.87 (2)	4.07±0.24 (3)	3.26±0.04 (3)		3.34±0.25 (3)	2.32±0.24 (5)
$W^{186}(p,n)Re^{186}$	13.7 ±2.8 (5)	6.17±0.5 (3)		5.10±0.37 (3)	4.35±0.34 (3)	

[a] Numbers in parenthesis indicate the number of independent determinations.

indicated a secondary effect of about 0.5 mb per 100 mg/cm² of target at 400 MeV.[18] The measurement most susceptible to secondary protons is the cross section of the $W^{186}(p,n)Re^{186}$ reaction, which made use of tungsten foils of almost 0.005-in. thickness and had a surface density of slightly less than 200 mg/cm². In this case the secondary correction amounted to about 20% at 400 MeV and about 25% at 130 MeV, assuming an energy dependence for low-energy secondary protons, between 30 and 100 MeV, as predicted by Bernardini *et al.*[19] For the other measurements it is concluded that the secondary correction is about 7% at 400 MeV for the tellurium and rhodium targets, and 10% for these targets at lower energies. The remaining targets at all energies are subject to a correction for the secondaries of about 5%.

DISCUSSION

The (p,n) excitation functions illustrated in Fig. 1 appear to be very similar, apparently independent of target mass, and to have an approximately inverse energy dependence. Since the (p,n) reaction must take place predominately by a charge-exchange scattering process, it seems reasonable to assume that these curves therefore reflect the probability of charge exchange as a function of incident energy. Furthermore, the elementary (p,n) cross section varies from $E^{-0.5}$ to E^{-1} in this same energy region. Thus one might expect the excitation functions of (p,n) reactions in heavy mass targets to be dependent on both the appropriately averaged part of the elementary differential p-n cross section, and on the restriction of low residual excitation energies. Only cases in which a scattering occurs into a narrow band of energies in the nucleus in such a way that the nucleus becomes excited to about 0–10 MeV while the outgoing neutron has kinetic energies up to about the incident energy would lead to the (p,n) product.

Despite the apparent similarity of the excitation functions listed in Fig. 1, certain subtle differences are revealed by inspection of the slopes of these curves. The slopes of the lines were determined by a least-squares fit of the data in the form $\ln\sigma = -s \ln E$, where s is equal to $d\ln\sigma/d\ln E$. The values of the exponent in the energy dependence obtained by this method are listed in Table III. As can be seen from this table, and illustrated in Fig. 2, the energy dependence of these (p,n) reactions becomes weaker the larger the mass number of the target, varying from $E^{-1.31}$ for Sc^{45} to $E^{-0.75}$ for W^{186}. This implies that the attenuation of the incident and outgoing nucleon waves increases with increasing target mass number, thus competing with the dominant E^{-1} dependence of the elementary p-n scattering. This effect has previously been predicted.[20] Thus, for the larger mass number targets suc-

TABLE III. Slopes, $-(d\ln\sigma/d\ln E)$, of (p,n) excitation functions.

Target	Slopes, $-(d\ln\sigma/d\ln E)$ Experimental	Calculated
Sc^{45}	1.31±0.12	1.12±0.22
Ni^{64}	1.12±0.18	1.67±0.33
Rh^{103}	0.84±0.17	1.77±0.40
Te^{130}	0.77±0.14	1.22±0.20
W^{186}	0.75±0.15	2.32±0.47

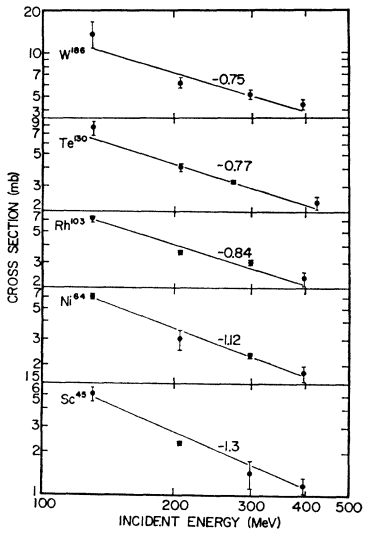

FIG. 1. Experimental excitation functions illustrated on a $\ln\sigma$ versus $\ln E$ plot. Exponents of the incident energy in the relation $\sigma = E^{-s}$, determined from the slopes $(+d\ln\sigma/d\ln E)$, are indicated above the curves.

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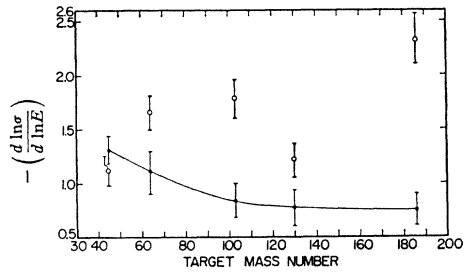

FIG. 2. Exponents of the energy versus target mass number. Solid circles: experimental values of $(-d \ln\sigma/d \ln E)$. Open circles: calculated values of $(-d \ln\sigma/d \ln E)$ from the Monte Carlo results.

cessful (p,n) reactions must be localized to peripheral regions of the nucleus.

Several other (p,n) excitation functions can be found in the literature[21,22] and have energy dependencies which fit with the trend depicted in Fig. 2. However, in a few other cases appreciable scatter is found which can be related to the fairly large experimental uncertainties associated with some of these sets of data. It is generally difficult to compare cross-section data from different laboratories particularly when taken at different times. It is exceedingly difficult to make such comparisons of the slopes of excitation functions, which quantities in general are associated with a larger uncertainty than the individual cross-section measurements. There is, however, a determination by another worker from this laboratory[23] of the $Cd^{111}(p,n)In^{111}$ excitation function which has been observed to have a slope of about -1.31. Variations from the smooth A dependence predicted in Fig. 2 should be anticipated, since if the magnitude of the slope is related to the attenuation of the incident beam in the peripheral region of the nucleus, and the nucleon density in such diffuse regions varies as the neutron number is changed for any given Z, the slope should correspondingly vary.

Despite the questionable validity of treating (p,n) reactions according to the conventional spallation model of intranuclear nucleon-nucleon collisions, a Monte Carlo calculation based on this model was performed specifically to calculate (p,n) cross sections. The Monte Carlo calculations for this study were written for the Control Data G-21 digital computer located at Carnegie Institute of Technology. This calculation was based on the same general assumptions as have been employed by others. As in the Metropolis calculation,[5] a degenerate Fermi gas of noninteracting protons and neutrons bound with a nuclear potential represented by a square well was used. The empirical differential nucleon scattering cross sections of Clements and Winsberg[24] were employed. The program was written in such a way that the computation was immediately terminated whenever any cascade would develop that would lead to a product other than that of the (p,n) reaction. The output information included, therefore, the total number of incident protons N_t, the total number of (p,n) cascades with appropriate residual energy N_n, and the total number of reactions of any type N_R. The (p,n) cross sections were then calculated by multiplying the N_n/N_t ratio by the geometric cross section. Whenever the residual excitation energy was in excess of 8 MeV this case was excluded since this energy is too high for gamma deexcitation to predominate over particle emission.

Generally the calculations were performed with approximately 3000 incident protons. However in some cases as few as 2000 were used and in a few cases as many as 4000 were used.

In Fig. 3 the calculated excitation functions are illustrated on a log-log plot. These should be compared

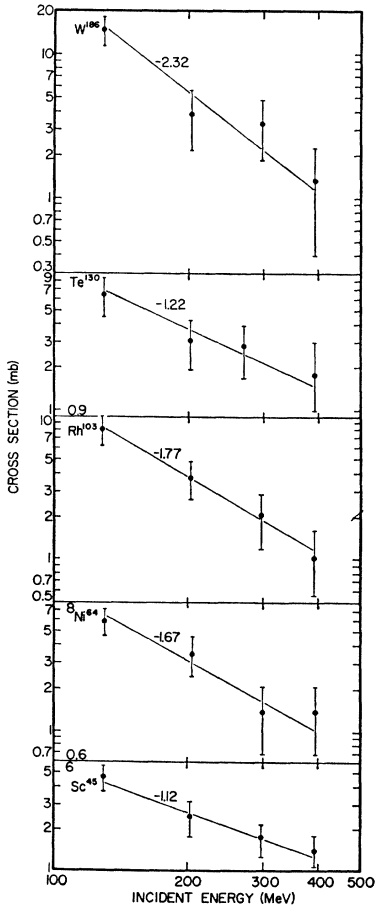

FIG. 3. Functions calculated from Monte Carlo computations based on simple nucleonic cascades of (p,n) events.

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with the experimental results illustrated in Fig. 1. The slopes of these calculated curves are also listed in Table III and in general increase with increasing target mass number as shown in Fig. 2. There is thus an inconsistency between the energy dependency predicted by this calculation and the experimental results, as well as with the qualitative interpretation previously suggested for these results. In view of the fact that this calculation does not take into account the refraction of the incident beam, and makes use of a square potential well rather than one with a diffuse edge, the lack of agreement is not too surprising. However, simple nuclear reactions of this type should properly be described by the appropriate quantal calculations.[20] Recently Read and Miller[22] have shown that they can predict the (p,n) cross-section variation with target mass number by making use of a model devised by Benioff.[25] Also, a quantitative theory has recently been worked out by Grover[26] in which the details of the shell-model configurations of the targets of (p,n) reactions are taken into account.

[25] P. A. Benioff, Phys. Rev. 119, 324 (1960).

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Neutron Total Cross Section of Pu^{242}†

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The neutron total cross section of Pu^{242} has been studied from 0.02 to 400 eV using the time-of-flight facility at the Livermore linear electron accelerator. Resonance parameters were determined for fourteen resonances, and the s-wave neutron strength function was found to be $(0.95\pm0.40)\times10^{-4}$. The total cross section at 0.025 eV was found to be 38.9 ± 1.6 b.

INTRODUCTION

STRENGTH functions for s-wave neutrons have been measured for a fairly large number of heavy nuclei.[1] A peak in these values is apparent in the neighborhood of $A=239$. However, the position and shape of this peak have not been well determined, and accurate information on the strength function at $A=242$ would be helpful. The neutron cross sections of Pu^{242} are also of interest, since this isotope is a possible target for heavy-element production by multiple neutron capture in a nuclear explosion.[2] We have, therefore, measured the neutron total cross section of Pu^{242} from 0.02 to 400 eV by the time-of-flight method using the 30-MeV electron linear accelerator of the Lawrence Radiation Laboratory as the pulsed neutron source.

† Work performed under the auspices of the U. S. Atomic Energy Commission.
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[2] D. Dorn (private communication).

EXPERIMENTAL DETAILS

The neutron detector for this experiment was situated at the end of an evacuated flight tube 16.7 m long, arranged at 90° with respect to the electron beam direction. The experimental apparatus in the vicinity of the detector is shown in Fig. 1. The dimensions of the neutron beam were reduced from 3 in. in diameter near the neutron source to $\frac{3}{4}$ in. at the sample by two collimators. The primary collimator located at the end of the flight tube consisted of 2 ft of borated polyethylene (about 10% by weight natural boron) lined with a $\frac{3}{8}$-in.-thick iron sleeve. This collimator reduced the beam diameter to $1\frac{1}{2}$ in. The purpose of the sleeve was to prevent the detector from looking directly at any hydrogenous material. The 4-in.-square iron collimator which formed part of the remotely controlled sample changer further reduced the beam diameter to $\frac{3}{4}$ in., about $\frac{1}{8}$ in. less than that of the samples which were placed in the middle of this collimator.

The neutron detector was located 29 in. from the samples. It was shielded by a $\frac{1}{2}$-in.-thick lead sleeve