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Effective cross sections for the (n, 2pn), (n, 2p), and (n, 3pn) reactions using intermediate-energy neutrons*

John C. Hill and D. G. Shirk
Ames Laboratory-ERDA and Department of Physics, Iowa State University, Ames, Iowa 50010

R. F. Petry
Department of Physics and Astronomy, University of Oklahoma, Norman, Oklahoma 73069

K. H. Wang
Department of Physics, Baylor University, Waco, Texas 76703

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"Effective cross sections" for the $(n,2pn)$, $(n,2p)$, and $(n,3pn)$ reactions on various targets were measured relative to the $^{12}C(n,2n)^{11}C$ reaction. The bombarding particles were the spectrum of neutrons generated in a Cu beamstop by 750-MeV protons. The cross section systematics as a function of target mass number are compared with available 14-MeV neutron cross sections for $(n,{}^3He)$ and $(n,2p)$ reactions and with results from intranuclear-cascade calculations.

[NUCLEAR REACTIONS ^{31}P, ^{45}Sc, ^{51}V, ^{75}As, ^{115}In, ^{127}I, ^{133}Cs, ^{142}Ce, and $^{197}Au(n,2pn)$; ^{45}Sc, ^{89}Y, ^{127}I, ^{133}Cs, and $^{139}La(n,2p)$; ^{31}P, ^{41}K, ^{75}As, ^{115}In, and $^{138}Ba(n,3pn)$; $E_n=0-750$ MeV; measured σ.]

I. INTRODUCTION

Experimental data on the systematics of the $(n,2pn)$, $(n,2p)$, and $(n,3pn)$ reactions are very scarce, and no information exists in the literature on cross sections at neutron energies other than 14 MeV. The above reactions are useful for production of new neutron-rich nuclei in regions not accessible from fission,[1] provided an intense source of high energy neutrons is available and the appropriate cross sections are sufficiently large. Bramlitt *et al.*[2,3] were unable to detect either the $(n,2p)$ or $(n,{}^3He)$ reactions using 14.7-MeV neutrons. Several cases of the $(n,2p)$ reaction were observed by Lulic, Strohal, and Šlaus[4] using 14.6-MeV neutrons. Diksic, Strohal, and Šlaus[5] and Qaim[6] observed several cases of the $(n,{}^3He)$ reaction using 14.6-MeV neutrons, but their results are not in good agreement with each other. All of the above studies employed radiochemical methods. No information on the $(n,3pn)$ reaction is available in the literature.

Study of the $(n,2pn)$, $(n,2p)$, and $(n,3pn)$ reactions was undertaken as part of a program to evaluate the feasibility of using fast neutrons generated by the line A proton beam stop at the Clinton P. Anderson Meson Physics Facility (LAMPF) for production of new neutron-rich nuclides for decay studies. In view of the small amount of data on these reactions, the absence of data above 15 MeV, and the large number of examples observed in this study, it seemed appropriate to report the results, examine their systematics as a function of target mass, and make a comparison with cross section data[4-6] available from 14-MeV neutron studies and results from intranuclear-cascade calculations.[7] This material was presented in preliminary form[8] at the 1974 Pittsburgh Meeting of the American Physical Society.

II. EXPERIMENTAL

In typical irradiations neutrons were generated by 5 μA of 750-MeV protons incident on a cylindrical water-cooled Cu beam stop (17.6 cm in diameter and 43 cm long) in the LAMPF proton line A. Targets varying in mass from 100 mg to 3 g were sealed in polyethylene tubes, placed in a plastic rabbit and irradiated for a suitable period at 90° to the beam axis and a distance of 20.3 cm from the beam axis. The reaction $^{31}P(n,3pn)^{28}Mg$ served as a convenient flux monitor, since it is insensitive to low energy neutrons and $^{28}Mg(\tau_{1/2}=21.1\text{ h})$ could be detected from irradiations as short as 10 min without post-irradiation chemistry. The monitor consisted of 3 g of Li_3PO_4 sealed in a polyethylene tube.

For all targets with $A>31$, intense γ rays from (n,xn) and (n,pxn) completely masked those from the $(n,2pxn)$ or $(n,3pxn)$ reactions of interest. Radiochemical separations following irradiation were thus necessary. Chemical yields were determined by neutron activation analysis of the samples after chemical separation and comparison to a standard. The yield of the residual nucleus from the reaction of interest was measured by counting γ rays using a Ge(Li) detector. Flux normaliza-

TABLE I. Cross sections (all radiochemical) for the $(n, 2pn)$, $(n, 2p)$, $(n, 3pn)$, and (n, α) reactions.

Reaction	$\tau_{1/2}$ (residual nucleus)	Q (MeV) [a]	σ (mb)
$^{12}C(n, 2n)^{11}C$	20.3 min	−18.72	18 ± 5
$^{31}P(n, 2pn)^{29}Al$	6.52 min	−20.80	9.8 ± 3.0
$^{45}Sc(n, 2pn)^{43}K$	22.2 h	−19.06	4.4 ± 2.2
$^{51}V(n, 2pn)^{49}Sc$	57.3 min	−20.23	4.2 ± 2.9
$^{75}As(n, 2pn)^{73}Ga$	4.91 h	−17.87	1.8 ± 1.1
$^{115}In(n, 2pn)^{113}Ag$	5.3 h	−17.08	2.1 ± 1.1
$^{127}I(n, 2pn)^{125}Sb$	2.73 yr	−15.29	2.7 ± 1.4
$^{133}Cs(n, 2pn)^{131}I$	8.04 day	−15.30	3.3 ± 2.0
$^{142}Ce(n, 2pn)^{140}Ba$	12.8 day	−15.90	0.25 ± 0.15
$^{197}Au(n, 2pn)^{195}Ir$*	4.2 h	−13.97	0.57 ± 0.40
$^{45}Sc(n, 2p)^{44}K$	22.15 min	−11.36	0.37 ± 0.19
$^{89}Y(n, 2p)^{88}Rb$	17.8 min	−11.54	0.35 ± 0.21
$^{127}I(n, 2p)^{126}Sb$	12.4 day	−9.16	0.22 ± 0.11
$^{133}Cs(n, 2p)^{132}I$	2.28 h	−8.96	0.13 ± 0.08
$^{139}La(n, 2p)^{138}Cs$	33.4 min	−10.28	0.050 ± 0.030
$^{31}P(n, 3pn)^{28}Mg$	21.1 h	−31.29	0.47 ± 0.14
$^{41}K(n, 3pn)^{38}S$	2.87 h	−30.62	0.0066 ± 0.0046
$^{75}As(n, 3pn)^{72}Zn$	46.5 h	−26.76	0.025 ± 0.015
$^{115}In(n, 3pn)^{112}Pd$	20.1 h	−25.14	0.0098 ± 0.0065
$^{138}Ba(n, 3pn)^{135}I$	6.7 h	−26.55	0.0035 ± 0.0025
$^{27}Al(n, \alpha)^{24}Na$	15.0 h	−3.13	47 ± 14
$^{31}P(n, \alpha)^{28}Al$	2.24 min	−1.94	43 ± 13
$^{45}Sc(n, \alpha)^{42}K$	12.4 h	−0.40	14.3 ± 7.2
$^{51}V(n, \alpha)^{48}Sc$	1.83 day	−2.50	8.1 ± 4.0
$^{75}As(n, \alpha)^{72}Ga$	14.1 h	+1.20	6.4 ± 3.2
$^{115}In(n, \alpha)^{112}Ag$	3.13 h	+2.67	5.5 ± 2.7
$^{127}I(n, \alpha)^{124}Sb$	60.2 day	+4.25	9.8 ± 4.9
$^{133}Cs(n, \alpha)^{130}I$	12.4 h	+4.38	11.0 ± 6.6
$^{139}La(n, \alpha)^{136}Cs$	13.0 day	+4.53	3.8 ± 2.3
$^{187}Re(n, \alpha)^{184}Ta$	8.7 h	+7.38	1.7 ± 1.0
$^{197}Au(n, \alpha)^{194}Ir$	19.4 h	+6.95	1.35 ± 0.95

[a] Q values calculated from masses given in Ref. 11.

tion was determined by counting ^{28}Mg γ rays from the Li_3PO_4 monitor. Corrections were made for growth and decay of both the residual nucleus of interest and the flux monitor and for fluctuations in beam intensity during irradiation.

The $^{12}C(n, 2n)^{11}C$ reaction ($Q = -18.72$ MeV) was used as the primary standard. The yield of the above reaction relative to $^{31}P(n, 3pn)^{28}Mg$ was measured by counting annihilation radiation from ^{11}C and γ rays from ^{28}Mg. Using information on the $^{12}C(n, 2n)^{11}C$ cross section up to 400 MeV compiled by Katcoff[9] and the 90° neutron spectrum for the LAMPF beam stop calculated by Perry,[10] an "effective cross section" of 18 ± 5 mb for the ^{12}C-$(n, 2n)^{11}C$ reaction for neutrons above 18.7 MeV was deduced. In the neutron spectrum assumed, for neutrons with energies above 10 MeV, roughly 75, 45, 25, 8, and 2% have energies above 20, 50, 100, 200, and 300 MeV, respectively. In what follows cross section is understood to mean effective cross section for the spectrum of beam-stop neutrons normalized to the $^{12}C(n, 2n)^{11}C$ cross section given above.

III. RESULTS AND DISCUSSION

Cross sections for $(n, 2pn)$, $(n, 2p)$, and $(n, 3pn)$ reactions obtained in this work are tabulated in Table I and plotted as a function of target A in Fig. 1. Results of (n, α) measurements are included for comparison. The errors reflect uncertainties in the $^{12}C(n, 2n)^{11}C$ cross section, chemical yields, and β and γ branching in the decays of the residual nuclei. These values for the cross sections could easily be renormalized if future experiments provide more complete information on either the $^{12}C(n, 2n)^{11}C$ cross section or the neutron spectrum at the beam stop.

A. Systematics and comparison with 14-MeV neutron experiments

1. (n,2pn) cross sections

The $(n, 2pn)$ cross sections measured in this experiment are typically a few mb and generally decrease with increasing A from about 9.8 mb at $A=31$ to about 0.57 mb at $A=197$. They are large enough to be useful in searches for new neutron-rich nuclides through the whole range of the Periodic Table. The ratio $\sigma(n, \alpha)/\sigma(n, 2pn)$ is fairly constant varying from 1.9 to 4.8 and can be used to evaluate the probability of observing the $(n, 2pn)$

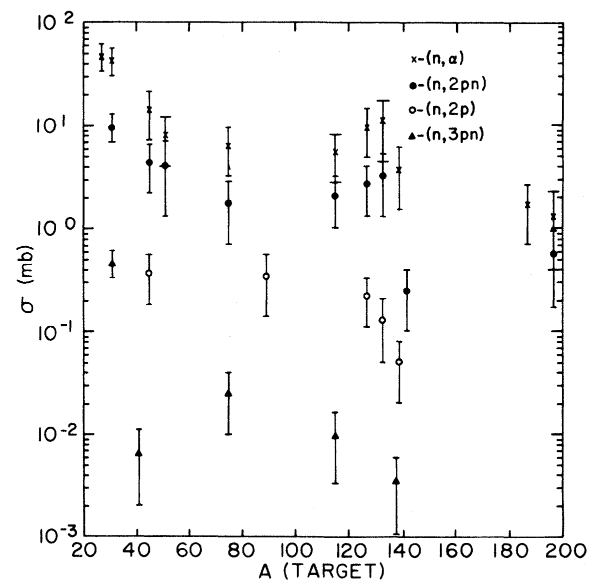

FIG. 1. Cross sections for the $(n, 2pn)$, $(n, 2p)$, $(n, 3pn)$, and (n, α) reactions obtained in this experiment.

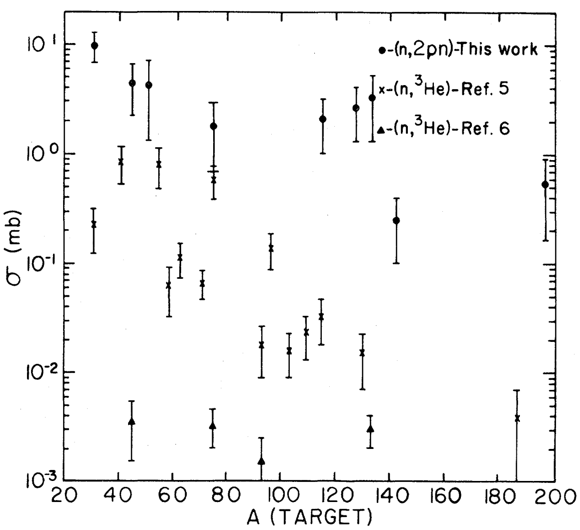

FIG. 2. Comparison of $(n, 2pn)$ cross sections from this work with results obtained using 14.6-MeV neutrons.

product once the corresponding (n, α) product has been identified. The anomalously low value of 0.25 mb for the ^{142}Ce$(n, 2pn)^{140}$Ba reaction cross section may indicate a significant decrease of the $(n, 2pn)$ cross section for targets on the neutron-rich edge of the valley of stability.

Q values for the $(n, 2pn)$ reaction given in Table I indicate that, with the exception of the ^{197}Au target, the $(n, 2pn)$ channel is closed for 14.6-MeV neutrons; thus only the $(n, ^3\text{He})$ channel is open. The results from this study involve contributions from $(n, 2pn)$ reactions and are compared in Fig. 2 with $(n, ^3\text{He})$ data[5,6] taken using 14.6-MeV neutrons. Our cross sections are typically a factor of 10 higher than those of Diksic *et al.*[5] at low A and a factor of 100 higher at high A. Four points for the $(n, ^3\text{He})$ reaction have been obtained by Qaim[6] and are shown in Fig. 2. They are typically an order of magnitude lower than those of Diksic *et al.*,[5] indicating a normalization problem for one of the data sets.

2. *(n,2p) and (n,3pn) cross sections*

The $(n, 2p)$ cross sections measured in this experiment are typically a few tenths of a mb and are about one order of magnitude less than those for the $(n, 2pn)$ reactions. The $(n, 2p)$ values are compared in Fig. 3 with the 14.6-MeV neutron results of Lulic *et al.*[4] and are roughly an order of magnitude larger. A slight decrease in cross section with increasing A was observed in our results but is not apparent in the 14.6-MeV data. The $(n, 3pn)$ cross section is typically a few μb with the exception of ^{31}P$(3pn)^{28}$Mg, which has a cross section of 470 μb. Both the $(n, 2p)$ and $(n, 3pn)$ reactions could be used to produce many new neutron-rich nuclides. The results of this study suggest that searches below $A = 80$ are feasible.

It has been suggested[12] that for reactions with intermediate energy neutrons such as $(n, 2pn)$, $(n, 2p)$, and $(n, 3pn)$, where proton emission predominates, the cascade phase of the reaction determines the gross properties of the cross section. The large increase in cross sections for the $(n, 2pn)$ and $(n, 2p)$ reactions observed in this work compared to the 14.6-MeV neutron cross sections[4-6] reflects the availability of more energy to drive the cascade. The cross sections plotted in Fig. 1 for the $(n, 2pn)$, $(n, 2p)$, and $(n, 3pn)$ reactions all show a definite decrease with increasing A, but the decrease above $A = 40$ is relatively small. This effect probably reflects the suppression of proton emission at high Z during the evaporation phase of the interaction, due to the Coulomb barrier. Data for the $(n, 2p)$ and $(n, 3pn)$ reactions above $A = 140$ would be helpful in illuminating the role of the Coulomb barrier in these reactions.

B. Comparison with intranuclear cascade calculations

The results obtained from this experiment provide the first test of the validity of the intranuclear cascade approach in describing the relatively rare $(n, 2pn)$, $(n, 2p)$, and $(n, 3pn)$ reactions leading to neutron-rich end products. Such calculations have been made by Bertini,[7] and access to the appropriate radiochemical cross sections, which are on microfilm, is described in Ref. 13. The predicted cross sections are for 25-, 50-, 100-, 150-, 200-, 250-, 300-, 350-, and 400-MeV neutrons incident on targets of ^{27}Al, ^{52}Cr, ^{65}Cu, ^{100}Ru, ^{140}Ce, ^{184}W, ^{207}Pb, and ^{238}U. Unfortunately, none of the targets used in this work were included in the calculations, but a compari-

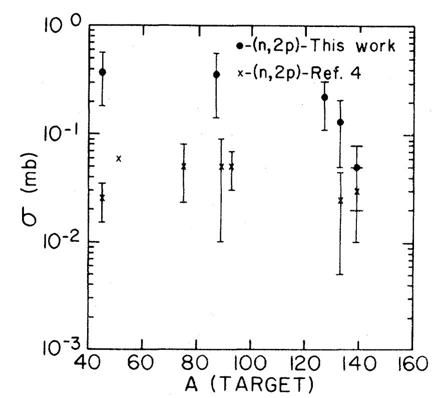

FIG. 3. Comparison of $(n, 2p)$ cross sections from this work with results obtained using 14.6-MeV neutrons.

son with our results is of interest in view of the regular behavior of the experimental cross sections and the trends observed as a function of A.

In order to compare our results with the above calculations, relative neutron intensities of our spectrum were tabulated at the above energies. This information was used to normalize Bertini's calculated cross sections in order to obtain an "effective intranuclear cascade (EINC) cross section" between 25 and 400 MeV. Neglect of neutrons above 400 MeV has little effect on the comparisons, since less than 2% of neutrons above 18.72 MeV have energies above 400 MeV. Also, the cross sections of interest generally decrease above 200 MeV. Our experimental results are of course normalized to the $^{12}C(n, 2n)^{11}C$ reaction having a Q of −18.72 MeV. Normalization problems may arise due to the neglect of neutrons below 25 MeV and will be discussed separately for each case. Comparisons were made only for cases where sufficient "events" were generated by the cascade calculation to make the results meaningful.

1. (n,2pn) cross sections

The EINC cross sections defined above were determined for the $(n, 2pn)$ reactions. Values of 9.3, 3.1, 3.3, 3.0, 1.7, and 0.95 mb were obtained for ^{52}Cr, ^{65}Cu, ^{100}Ru, ^{140}Ce, ^{184}W, and ^{207}Pb targets, respectively. Normalization problems below 25 MeV are not severe, since the calculated cross section with 25-MeV neutrons is negligible except for ^{52}Cr and ^{65}Cu, in which cases it is still small compared to the 50-MeV cross section. Also, $(n, ^3He)$ processes at energies below 18.7 MeV probably do not make significant contributions, as can be seen from the small values for experimental cross sections at 14.6 MeV. The EINC cross sections have the same order of magnitude (a few mb) as our results and exhibit the same slow decrease with increasing A (9.3 mb at $A=52$ to 0.95 mb at $A=207$) observed by us.

2. (n,2p) and (n,3pn) cross sections

The EINC cross sections were also determined for the $(n, 2p)$ reaction. Values of 2.5, 0.61, 1.45, 0.38, and 0.66 mb were obtained for ^{52}Cr, ^{65}Cu, ^{100}Ru, ^{140}Ce, and ^{184}W targets, respectively. Both experimental and EINC cross sections show a slow decrease with increasing A, but a curve through the EINC values averages roughly a factor of 4 higher than one through the experimental values. Normalization problems are not negligible due to the low Q values for the $(n, 2p)$ reaction which result in appreciable cross sections for neutrons with energies below 25 MeV. The difference in the cross section magnitudes does not therefore necessarily imply a deficiency in the $(n, 2p)$ calculations.

The EINC cross sections were determined for the $(n, 3pn)$ reactions. In all cases a zero cross section was assumed for 25-MeV neutrons. Values of 4.1, 0.38, 0.34, and 0.086 mb were obtained for ^{27}Al, ^{52}Cr, ^{65}Cu, and ^{100}Ru targets respectively. Normalization problems at energies below 25 MeV do not enter, since the magnitude of Q for all the reactions considered is greater than 25 MeV. The EINC cross section for $^{100}Ru(n, 3pn)$-^{97}Nb has an additional uncertainty due to the small number of events of this type generated in the intranuclear-cascade calculation. The decrease in the experimental cross section of an order of magnitude in going from a ^{31}P target to targets of higher A is strikingly similar to the decrease in the EINC cross section in going from an ^{27}Al target to targets of higher A, but the EINC cross sections are generally about an order of magnitude higher.

The intranuclear-cascade calculations[7,13] are quite successful, both in describing the change in cross section for the $(n, 2pn)$, $(n, 2p)$, and $(n, 3pn)$ reactions as a function of mass number, and in predicting the magnitude of the $(n, 2pn)$ cross section. The above calculations appear to be less successful in predicting the magnitude of the $(n, 3pn)$ cross section. Because of normalization problems, discussed above, comparisons for the magnitude of the $(n, 2p)$ cross sections are less meaningful.

The uncertainty in the neutron spectrum at the beam stop is the greatest uncertainty which one must confront in the interpretation of the above results. The authors would thus strongly encourage others to measure the beam-stop neutron spectrum. Our results could then be renormalized to the new measurements using the $^{12}C(n, 2n)^{11}C$ reaction.

Note added in proof. Katcoff has recently measured $(n, 2pn)$, $(n, 2p)$, $(n, 3pn)$, and (n, α) cross sections using neutrons generated by a 100 μA beam of 200-MeV protons from the Brookhaven alternating gradient synchrotron (AGS) injector incident on a Cu disk. In cases where the targets were the same, their cross sections were typically 1.5 to 2.0 times larger than ours, and similar trends as a function of target mass number were noted.

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