Excitation Functions of the (p,pn) and $(p,2p)$ Reactions on Ce^{142} at 60–233 Mev*

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The cross sections of the reactions $Ce^{142}(p,pn)Ce^{141}$ and $Ce^{142}(p,2p)La^{141}$ have been determined for protons of 60, 120, 180, 200, and 233 Mev energy. A comparison is made of the observed yields with those predicted on the basis of Monte Carlo cascade-evaporation calculations at 83, 238, and 368 Mev. For protons of energy between 60 and 240 Mev the agreement is good for the (p,pn) reaction and fair for the $(p,2p)$.

INTRODUCTION

ALTHOUGH considerable effort has been devoted to the elucidation of spallation, fission, and fragmentation reactions at high bombarding energies (>60 Mev), little attention has been given to the study of competition between reactions such as (p,pn) and $(p,2p)$ at these energies. In spallation studies it has been common practice to use targets of monoisotopic elements in order that there be a single primary source of product nuclides, and, while the (p,pn) product is generally amenable to radiochemical analysis, the $(p,2p)$ product is invariably stable.

Although the (p,pn) and $(p,2p)$ reactions are among the most elementary of nuclear reactions, there are several mechanisms whereby the two product nuclides can arise. The contribution from each mechanism depends upon the energy of the incident proton and the nature of its interaction with the target nucleus. For incident protons in the energy range of 60 to 240 Mev, both reactions can result from a direct knock-on process which leaves the resultant nucleus with insufficient residual excitation energy for nucleon evaporation. The (p,pn) product can also arise from two evaporation mechanisms, viz., neutron evaporation from the excited target nucleus and proton evaporation from the excited $(A, Z+1)$ nucleus. The $(p,2p)$ product can, by proton evaporation, also arise from the excited target nucleus. The evaporation mechanisms would be expected to decrease in probability as the incident proton energy increases, while the knock-on mechanism would be expected to increase with increasing proton energy.

Unfortunately, the radiochemical method measures only the total cross section or the sum of the various contributing mechanisms. By measuring the cross sections as a function of incident proton energy information can be obtained regarding the extent to which various mechanisms contribute to the total cross sections, but this is based, to some extent, upon theoretical predictions as to the manner in which the various mechanisms depend upon the bombardment energy. Theoretical predictions based upon Monte Carlo cascade data are very useful since the Monte Carlo method breaks down the total cross section into contributions from various mechanisms, but the difficulty of comparing total cross sections remains. Agreement between experiment and theory can thus be looked upon only as an indication of the importance of the various mechanisms considered in the calculation of the predicted cross section.

Ce^{142} was chosen as the target nucleus for the present study since Ce^{141} and La^{141} are radioactive and have suitable half-lives and decay characteristics. Ce^{142} is the heaviest stable cerium isotope and thus the (p,pn) and $(p,2p)$ products of interest could arise only from Ce^{142}. This was a fortunate choice since Metropolis *et al.*[1] have made a Monte Carlo cascade calculation on Ce^{140}, and sufficient data were also available to permit an evaporation calculation with the cascade data to obtain total cross sections. In addition, these two reactions had been studied by Caretto and Friedlander[2] in the energy range of 380–3000 Mev.

There are relatively few target nuclides suitable for a study of the (p,pn)–$(p,2p)$ competition, and while Ce^{142} is probably one of the most favorable cases, the fact that La^{141} must be determined indirectly limits the accuracy with which the $(p,2p)$ cross sections can be determined for incident protons between 60 and 240 Mev.

EXPERIMENTAL

Spectroscopically pure cerium dioxide was used as the target material. The target contained approximately 100 mg of CeO_2 and was bombarded in the internal proton beam of the Rochester 130-in. synchrocyclotron as described previously.[3] Targets were bombarded for one hour at proton energies of 60–233 Mev.

A direct quantitative determination of 3.7-hr La^{141} by beta proportional counting would have been extremely difficult due to the necessity of separating La from the other rare earth activities in the presence of La carrier. In addition, La^{132} was produced, making accurate decay-curve resolution impossible. These difficulties were overcome by use of an indirect method,

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similar to that used by Caretto,[4] which involved determining La^{141} by counting the daughter Ce^{141}.

Within 15 min after termination of the bombardment the CeO_2 target material was quantitatively dissolved in $10M$ HNO_3 and made up to a known volume. Two aliquots were taken, one of which was allowed to stand until all the 4-hr La^{141} had decayed to Ce^{141}. Cerium was immediately separated and precipitated as the oxalate from the other aliquot by the method of Glendenin *et al.*[5] The aliquot which had been allowed to stand until the La^{141} had decayed to Ce^{141} was treated in identical fashion, giving a second oxalate sample for counting which contained Ce^{141} from the (p,pn) reaction plus that from the decay of the $(p,2p)$ product La^{141}. Chemical yields were determined gravimetrically by ignition of the oxalate to CeO_2 after counting was complete.

The samples were counted on an end-window, methane-flow beta proportional counter of the Los Alamos type. The samples were approximately 2.2 cm in diameter and weighed between 15 and 30 mg. No attempt was made to follow the short-lived activities in the samples except to check for radiochemical purity. That the chemical technique produced good radiochemical purity was indicated by the fact that all the activities observed could be accounted for by assuming a pure cerium sample at the time of chemical separation. That good separation was obtained from lanthanum was indicated by the absence of 40-hr La^{140} and by the growth curves obtained in a study of short-lived cerium isotopes.[6] In addition, Ce^{139} and Ce^{141} were the only long-lived isotopes observed to be present.

Each sample was counted for at least six months, and many were followed for over a year. The decay curves could not be resolved graphically because of the excessive time required for Ce^{141} to decay and the relative intensities of Ce^{141} and Ce^{139}. The decay curves were resolved analytically, assuming half-lives of 32.8 days[7–9] and 140 days[10] for Ce^{141} and Ce^{139}, respectively. The data for Ce^{141} were extrapolated to the time corresponding to the end of bombardment. The two inner aluminum foils from each target were counted on the same equipment and the decay curves resolved graphically to obtain the count of Na^{24} at the end of the bombardment.

Conversion of observed counting rates at the end of bombardment into absolute disintegration rates was accomplished with conversion factors taking into account corrections for self-absorption, backscattering, air scattering, chemical yield, counting efficiencies, and counting geometry. The geometrical factor was determined from National Bureau of Standards P^{32} for the geometry used in counting both the cerium and aluminum samples. The air-window absorption was calculated for Ce^{141} and Na^{24} from the coefficient of absorption taken from the graphs of Gleason *et al.*[11] That this was valid for our geometry was demonstrated with P^{32} and Co^{60} by using aluminum absorption techniques. The self-absorption correction for Na^{24} was calculated from the absorption coefficient and was essentially unity. The self-absorption—backscattering—self-scattering correction for Ce^{141} was interpolated from the data of Cuninghame *et al.*[12] The backscattering correction for Na^{24} was determined experimentally. Air scattering was considered negligible. Since the decay schemes of Ce^{141} and Na^{24} are well known, the counting efficiency for each isotope was easily computed. No correction was necessary for the finite resolving time of the counter since all counting was done at rates where this correction was negligible.

Absolute disintegration rates at the end of bombardment were calculated to infinite bombardment time. A half-life of 15.0 hr[13] was used for Na^{24} and of 3.7 hr[14] for La^{141}. Absolute cross sections for the production of Ce^{141} and La^{141} were obtained by comparison with the known cross sections[15] for the $Al^{27}(p,3pn)Na^{24}$ monitor reaction.

It was necessary to correct for the decay of La^{141} to Ce^{141} during the bombardment and between the end of bombardment and the time of chemical separation. The derivation is straightforward.[16]

RESULTS

The (p,pn) cross section and the ratio of the cross sections were calculated first and the $(p,2p)$ cross section calculated from the ratio. The values obtained from the experiments at various energies are presented in Table I. For the (p,pn) cross section the scatter is caused principally by errors in chemical yield, self-absorption corrections, and beam monitoring. It will be noticed that the reproducibility for the (p,pn) cross section is rather good for this type of work while the $(p,2p)$ cross section shows considerable scatter. It is felt that the inaccuracy of the $(p,2p)$ determination is caused principally by the quite low yield of La^{141} as compared to that of Ce^{141}; the enrichment was thus quite small in the sample allowed to stand until the decay of La^{141} had taken place. This tends to magnifiy the errors in the self-absorption correction, chemical yield, and errors in the counting data, since the method required the calculation of a difference that was small. This was the largest factor contributing to the errors in the $(p,2p)$

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TABLE I. Cross sections of the (p,pn) and ($p,2p$) reactions on Ce^{142}.

Proton energy (Mev)	(p,pn) (mb)	(p,pn)/($p,2p$)	($p,2p$) (mb)
233	60.7	2.0	30
	62.7	3.3	19
	71.6	7.2	10
	90[a]	...	
	66.9	17[a]	
	av 65.5±3.8	4.2±2	19.6±7
200	65.5	3.4	19.3
	72.3	7.8	9.2
	av 68.9±3.4	5.6±1.5	15.5±6
180	86.0	[b]	[b]
	67.9	9.9	6.9
	67.6	4.7	14.4
	73.4	2.9	25.2
	av 73.7±8	5.8±2.5	15.5±6
120	125	...	
	72.9	4.6	15.9
	105	13.7	7.7
	90	...	
	av 98.2±17	9.1±4.5	11.8±4
60	125	21.7	5.8
	96	7.9	12.2
	121	12.4	9.8
	av 114±12	14.0±5.4	9.2±2

[a] Omitted from the average.
[b] La sample lost.

determination and is inherent in the method. It would be highly desirable to repeat the ($p,2p$) determination using a direct method.

If approximately 5% absolute errors are estimated for all counting corrections except the counting efficiency, and 5% allowed for the uncertainty in the monitor cross section, an estimated 20% absolute error is obtained for the results of the (p,pn) study. This is quite reasonable for this type of work. For the ($p,2p$) reaction, the absolute error is probably much greater.

DISCUSSION

It is of interest to compare the present results with the cross sections to be expected on the basis of the cascade-evaporation model for these reactions. The Monte Carlo calculations on intranuclear cascades of Metropolis *et al.*,[1] fortunately included proton energies of 83 and 238 Mev and Ce^{140} as a target nucleus. The detailed cascade data[17] together with data on the compound nucleus Ce^{141*}, needed for the evaporation calculation and determined by Caretto,[4] permitted calculations to be made of the cross sections to be expected for the (p,pn) and ($p,2p$) reactions with Ce^{142}.

For Ce^{140} the products of interest from the cascade data were Ce^{140*}, Pr^{140*}, Ce^{139*}, and La^{139*}. In addition, it was necessary to know the total number of proton cascades at each energy, the assumed nuclear radius and the number of cases for the various excited product nuclides found falling within given intervals of residual excitation energy. From the data, plots were constructed to give the number of cases per 10-Mev interval of residual excitation energy as a function of residual excitation energy. It was necessary in the case of Ce^{139*} and La^{139*} to determine the number of cases where the residual excitation energy was insufficient for nucleon emission. A cutoff of 9 Mev was used; this was found not to be critical. For Ce^{140*} and Pr^{140*} it was necessary to apply the compound nucleus data for Ce^{142*}. These consisted of the excitation functions for $Ba^{138}(\alpha,n)Ce^{141}$ and $Ba^{138}(\alpha,p)La^{141}$ from 15- to 40-Mev alpha bombarding energy, and the compound nucleus total cross section.[4] It was assumed that these data could be applied to excited Ce^{140} and Pr^{140} and in addition it was assumed that the (p,pn) and ($p,2p$) cross sections calculated from the Monte Carlo cascade data would apply to the experimental results with Ce^{142}. These assumptions are reasonable since these are high energy reactions and in addition no shell effects were considered in the Monte Carlo calculation.

The (α,n) and (α,p) excitation functions plus the total compound nucleus cross section as a function of energy permitted the calculation of the fraction of excited Ce^{140} and Pr^{140} nuclei which lost just a neutron and just a proton, respectively. It was thus a simple matter to calculate the contribution of these two excited nuclides to the Ce^{139} and La^{139} yields. The desired (p,pn) and ($p,2p$) cross sections were then easily obtained from the total number of Ce^{139} and La^{139} cases, respectively. The geometrical cross section was computed from $r=1.3\times10^{-13}A^{\frac{1}{3}}$. The results of the calculations are given in Table II along with the estimated errors. The errors were estimated from the uncertainty in the points on the distribution plots of number of cases per 10-Mev interval vs excitation energy. Also shown are the percentages of the calculated cross section resulting from the various contributing processes.

The excitation functions for the (p,pn) and ($p,2p$) reactions are plotted in Fig. 1, along with the values obtained from the cascade-evaporation calculations. The values reported[2] for 380 Mev are also shown. For proton energy between 60 and 240 Mev the agreement with the calculations is good for the (p,pn) reaction and fair for the ($p,2p$) reaction. The cross sections in this energy interval will be considered first.

The low ($p,2p$) cross section compared to the (p,pn) cross section at 60 Mev may be considered as due to the following factors: (a) Ce^{141} can be produced by three processes whereas only two contribute to the production of the ($p,2p$) product. (b) Neutron evaporation from excited Ce^{142} in the range of excitation energies from 15 to 40 Mev is three to four times more probable than proton evaporation. This is indicated by the (α,n) and (α,p) excitation functions of Caretto.[4]

As can be seen from Table II, the Monte Carlo calculation predicts that at 84 Mev the contribution from the direct knock-on process will be relatively small, but that there is a higher probability of knocking out a neutron than a proton. This is no doubt in part

[17] Kindly supplied by Dr. Anthony Turkevich.

TABLE II. Cross sections predicted by cascade evaporation calculations.

	83 Mev		238 Mev		360 Mev	
	No. cases	% σ	No. cases	% σ	No. cases	σ
Incident protons	1264	...	974	...	886	...
$Ce^{140}(p,pn)Ce^{139}$	12	14	12	29	14	45
$Ce^{140}(p,n)Pr^{140} \rightarrow Ce^{139}+p$	20	23	7.3	18	1.8	5
$Ce^{140}(p,p)Ce^{140} \rightarrow Ce^{139}+n$	54	63	21.8	53	15.3	49
$Ce^{140}(p,2p)La^{139}$	3.7	16	2.5	22	6.5	52
$Ce^{140}(p,p)Ce^{140} \rightarrow La^{139}+p$	19	84	8.7	70	6.0	48
$\sigma(p,pn)$, mb	97±10		60±9		50±10	
$\sigma(p,2p)$, mb	25±6		16.5±5		20±6	
$\sigma(p,pn)/\sigma(p,2p)$	4±1		3.6±0.5		2.5±0.8	

due to the presence of more neutrons than protons in the target nucleus. Thus at 60 Mev the ratio of the two cross sections is probably determined by potential barrier considerations and the fact that charge exchange and deuteron pick-up reactions contribute to the (p,pn) cross section.

As the proton energy is increased, the experimental value of the cross section of the $(p,2p)$ reaction (Table I) appears to increase slightly, whereas the $(p,2p)$ cross section is about one-fourth that of the (p,pn). The (p,pn) excitation function can be interpreted in the following manner. As the proton energy is increased the charge exchange reaction followed by proton emission would be expected to decrease, as well as the reaction involving neutron evaporation from excited Ce^{142}. Larger amounts of energy are being deposited in the target nucleus favoring the loss of more than one nucleon. However, the probability of forming Ce^{142} with an excitation energy of 10 to 30 Mev will decrease rapidly as the proton energy is increased, since the average energy deposited in the target nucleus is a slowly increasing function of energy. Thus the contribution from Ce^{142} through excitation followed by neutron evaporation is expected to decrease slowly in going from 60 to 240 Mev. The Monte Carlo calculation predicts a slow decrease in the contribution from charge exchange as the proton energy is increased.

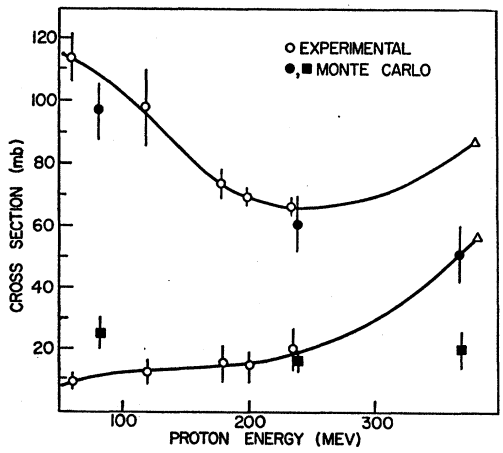

FIG. 1. Experimental and calculated cross sections of the (p,pn) and $(p,2p)$ reactions on Ce^{142} as a function of energy.

It will be noticed that the (p,pn) excitation function levels off as the proton energy approaches 240 Mev. This is undoubtedly due to the increase in the relative proportion of the direct knock-on production of the (p,pn) product as the energy increases. This relative increase in the direct knock-on reaction with energy is predicted by the Monte Carlo results and can be explained in the following manner. At 60 Mev the direct knock-on reaction may be thought of as occurring near the diffuse rim of the nucleus. This is suggested by the work of Elton and Gomes[18] who found that to explain certain inelastic scattering results it was necessary to assume that the knock-on reaction with 31-Mev protons on medium to heavy nuclei was taking place in the diffuse rim. This would also be the case to some extent at 60 Mev. With 31-Mev protons they found theoretically a very low probability for an interaction in the central core of the nucleus that would result simply in the loss of the proton and one neutron without additional interaction.

As the proton energy is increased it is reasonable to expect that a larger portion of the outer shell of the nucleus would participate in the knock-on reaction. The nucleus becomes more transparent as the energy increases, and as a consequence there is a higher probability of protons passing through the nucleus and interacting at or near the back surface to knock out a nucleon. Since the energy of the struck nucleon will tend to increase with increasing proton energy, the struck nucleon will have a higher probability of escape without further interaction. Thus instead of the knock-on reaction merely involving the diffuse rim, at higher energies an appreciable portion of the surface shell on the back side (relative to the direction of the incident particle) of the nucleus might be expected to participate, and the probability would increase. Thus as the excitation function levels off, the decreasing probability of the evaporation mechanism is being presumably counterbalanced by an increase in the knock-on mechanism.

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The increasing trend in the $(p,2p)$ excitation function may also be attributed to an increase in the knock-on process and a decrease in the mechanism involving proton evaporation from excited Ce^{142}. It will be noticed (Table II) that the cascade-evaporation calculations perhaps predict a slight decrease in the $(p,2p)$ cross section with increasing energy. However, they also predict that the direct knock-on reaction will increase while the evaporation mechanism decreases in probability. It appears that not enough weight is given to the knock-on process.

At 240 Mev the observation that the (p,pn) reaction has approximately four times as great a probability as the $(p,2p)$ process can be interpreted as due in part to the contribution of neutron evaporation from excited Ce^{142}, which will still be two to three times more probable than proton evaporation, since the same range of nuclear excitation energies is involved. In addition it is reasonable to assume that the knock-on process would still favor neutrons over protons owing to their relative abundances. There are other factors that influence the knock-on probability, such as the exclusion principle for all interactions taking place within the Fermi volume, the nucleon-nucleon inelastic cross sections, the probability for nucleons to escape once struck, etc. The Monte Carlo calculation of course utilizes such considerations.

In making a comparison with the results of Caretto[2] at 380 Mev it is necessary to consider the absolute errors in the measurements. Since more or less the same method was used in the two investigations, it does not seem reasonable that an error of 30 to 40% would exist. In fact, a real rise in $\sigma_{p,pn}$ is confirmed by recent data of Strohal and Caretto.[19] In the $(p,2p)$ case, the increase is too large to be accounted for by absolute errors. A plot of the ratio $\sigma(p,pn)/\sigma(p,2p)$ as a function of energy gives a very smooth curve from 60 to 380 Mev, but this is to some extent to be expected since many counting corrections cancel in the ratio. However, this smooth curve may be fortuitous, as may the cascade-evaporation calculation prediction of the observed cross section ratios at 240 and 380 Mev.

Thus it would appear that the Monte Carlo cascade data and evaporation calculation, although based on a model which leaves a great deal to be desired, are somewhat realistic. In addition, reasonable explanations can be given for the observed excitation functions. However, the limitations of the radiochemical method discussed at the beginning of this paper must be kept in mind.

ACKNOWLEDGMENTS

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