Nuclear Reactions of Silver and Indium with 200- and 400-MeV Protons*

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(Received 3 March 1967)

The cross sections of a number of $(p,ypxn)$ reactions ($y=0$–4, $x=3$–12) of silver and indium with 200- and 400-MeV protons have been determined. The results are compared with calculations based on the Vegas and Metropolis cascade codes and a Monte Carlo evaporation calculation. Poor agreement is obtained with respect to both the magnitude of the cross sections and the ratios of isobaric yields. Several possible sources of these discrepancies are discussed.

I. INTRODUCTION

THE interaction of high-energy protons with complex nuclei is commonly considered to involve a two-step mechanism. In the first step, an intranuclear cascade is generated by a series of successive nucleon-nucleon collisions. The result of the cascade is the emission of prompt particles and the formation of a variety of residual nuclei having a broad spectrum of excitation energies. These residual nuclei deexcite in a slower second stage by the evaporation of particles and the emission of electromagnetic radiation.

Theoretical analyses of the cascade-evaporation process have usually involved Monte Carlo calculations of each of these steps. The calculation predicts the cross sections and recoil properties of spallation products and these can be compared with experimental determinations of these quantities. A number of comparisons of this type have been reported for a variety of targets and bombarding energies.[1–15]

The present study falls into this category. The cross sections of a number of $(p,ypxn)$ ($y=0$–4, $x=3$–12) reactions of indium and silver with 200- and 400-MeV protons have been measured. The results are compared with cascade-evaporation calculations. The Monte Carlo cascade codes used in this comparison include that by Metropolis *et al.*[16] as well as two versions of the recent Vegas calculation by Chen *et al.*[17] The results of all these computations were combined with a Monte Carlo evaporation calculation based on the code of Dostrovsky *et al.*[18] A comparison of the predicted recoil properties with experimental data for the reactions of present interest is given in the following paper.[19]

The choice of indium and silver as targets was dictated by the fact that the competition from fission is small ($\sigma_F \sim 0.3$ mb)[20] so that this process can be ignored as a competing deexcitation mode in the calculations. In addition, the results can be correlated with experimental data obtained in nuclear emulsion studies.[20] The experimental procedure is described in Sec. II and the results are given in Sec. III. Section IV presents the comparison between experiment and calculation.

II. EXPERIMENTAL PROCEDURE

Targets of metallic silver or indium were irradiated with 200- and 400-MeV protons at the Carnegie Institute of Technology synchrocyclotron. Product nu-

* Supported by the U. S. Atomic Energy Commission.

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TABLE I. Decay properties of observed nuclides.

Nuclide	Mode of decay	L/K[a]	$T_{1/2}$	Method of detection	D/A[b]
Pd^{100}	100% E.C.	0.153	3.63 day	x ray	0.801[c]
Pd^{101}	96.8% E.C.; 3.2% β^+	0.124	8.4 h	x ray	0.885[d]
Ag^{101}			10.1 min	daughter	
Pd^{103}	100% E.C.	0.124	17 day	x ray	1.23[e]
Ag^{103}			1 h	daughter	
Ag^{105}	100% E.C.	0.120	40 day	x ray	1.27[e]
Cd^{105}			55 min	daughter	
Ag^{106m}	100% E.C.	0.120	8.3 day	x ray	1.28[e]
Ag^{111}	100% β^-		7.5 day	β^-	1.00[e]
Ag^{112}	100% β^-		3.2 h	β^-	1.00[e]
Ag^{113}	100% β^-		5.3 h	β^-	1.00[e]

[a] Ratio of L/K electron capture [H. Brysk and M. E. Rose, Rev. Mod. Phys. 30, 1169 (1958)], multiplied by the factor 1.13 [Berol L. Robinson and Richard W. Fink, Rev. Mod. Phys. 32, 117 (1960)].

[b] Disintegration per emitted radiation. Values of nuclides measured via x ray contain corrections for percentage electron capture, fraction of K capture, fraction of K vacancies resulting from γ rays of associated daughter and fluorescence yield of the daughter nuclide.

[c] Based on decay scheme proposed by J. S. Evans and R. A. Naumann, Phys. Rev. B138, 1017 (1965).

[d] Based on decay scheme proposed by J. S. Evans, E. Kashy, R. A. Naumann, and B. F. Petry, Phys. Rev. B138, 9 (1965).

[e] Based on decay schemes as described in *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences—National Research Council, Washington 25, D. C., 1966).

clides of cadmium, silver, and palladium were selected for study on the basis of their convenient radioactive decay properties, as listed in Table I. The half-life of an unreported silver isotope Ag^{101} was determined in the course of this investigation.[21]

The target assembly consisted of 0.002-in.-thick indium or 0.003-in.-thick silver foils of $\geq 99.99\%$ purity, surrounded by 0.001-in. aluminum catcher foils of high purity, followed by 0.001-in. aluminum monitor foils and wrapped with 0.001-in. aluminum.[21] The dimensions of each foil were 1 in.$\times$1 in. Each target foil was cleaned by dipping in dilute nitric acid prior to bombardment. The periods of irradiation varied from 1 to 45 min as described in column 4 of Tables II and III. Solution of the target foil in nitric acid was performed immediately after the irradiation in the determinations of the short-lived nuclides, or two to four days later in the case of the long-lived products. The solution was diluted to 1000-ml and 5-ml aliquots taken for analysis. Duplicate samples were analyzed in the majority of the determinations.

Standard radiochemcial procedures for silver and palladium were used to isolate the respective activities.[21] The final precipitates of silver chloride or palladium dimethylglyoxime ($\sim$9 mg/cm^2) were filtered onto tared filter disks, washed, dried, weighed, and mounted on aluminum cards for counting.

The activities of Ag^{101}, Ag^{103}, and Cd^{105} were determined via their respective daughter activities. Removal of the initial daughter activity and isolation of the parent was rapidly carried out for these nuclides following the irradiation. The procedure for silver has been described elsewhere.[21] For Cd^{105}, three successive silver chloride precipitations were followed by the precipitation of cadmium sulfide. The sulfide was allowed to stand for 24 h to permit the complete decay of Cd^{105} into Ag^{105}. The sulfide was then dissolved in the presence of silver carrier and silver radiochemically separated. In the determinations of Pd^{101}, the target foil was dissolved $1\frac{1}{2}$ to 2 h after the irradiation to permit the quantitative decay of Ag^{101} into Pd^{101}. The final separations of silver in the Ag^{112} and Ag^{113} experiments were carried out approximately 6 to 8 h after the irradiation, thereby reducing the contributions from the shorter-lived silver nuclides.

The radioactive decay measurements were performed with the following two instruments. A calibrated NaI crystal spectrometer coupled to a multichannel analyzer was used to determine the x-ray emission rate of the palladium and silver nuclides. A calibrated end-window methane-flow β-proportional counter was used to measure the disintegration rate of the neutron excessive silver products as well as that of Na^{24} in the monitor foils.

The activity of each nuclide at end of bombardment was determined by means of a least-squares analysis of the decay curves. This was performed by a computer program, which also obtained the best half-life fit to the data[21]. The analysis of 33 palladium decay curves gave a half-life of 8.27$\pm$0.09 h for Pd^{101}.

TABLE II. Formation cross sections of silver and palladium nuclides from silver with 200- and 400-MeV protons.

Nuclide	σ(mb) 200 MeV	σ(mb) 400 MeV	T(min) Length of irradiation
$Pd^{100}(c)$	19.1$\pm$0.3	14.8$\pm$0.3	45
	19.5$\pm$0.5	15.8$\pm$0.5	45
	17.8$\pm$0.5	15.3$\pm$0.3	5
Average	18.8$\pm$0.9	15.3$\pm$0.5	
$Pd^{101}(c)$	40.2$\pm$1.2	33.5$\pm$0.5	5
$Ag^{101}(c)$	8.4$\pm$0.3	5.1$\pm$0.1	1
	9.3$\pm$0.2	5.1$\pm$0.1	1
Average	8.9$\pm$0.5	5.1$\pm$0.0	
$Pd^{103}(c)$	68.8$\pm$2.3	47.5$\pm$0.9	45
	71.3$\pm$1.8	51.0$\pm$1.5	45
	62.5$\pm$1.7	49.4$\pm$0.7	5
Average	67.5$\pm$4.6	49.3$\pm$1.8	
$Ag^{103}(c)$	42.7$\pm$1.2	35.1$\pm$6.5	45
	44.0$\pm$1.1	28.8$\pm$1.2	45
	40.8$\pm$1.1	26.0$\pm$0.9	5
Average	42.5$\pm$1.6	28.0$\pm$1.5[a]	
$Ag^{105}(c)$	60.6$\pm$1.3	41.0$\pm$0.8	45
	62.6$\pm$1.0	42.9$\pm$1.3	45
	60.7$\pm$1.7	42.0$\pm$0.6	5
Average	61.2$\pm$1.1	42.0$\pm$1.0	
$Cd^{105}(i)$	10.8$\pm$2.0	4.1$\pm$0.9	1
	12.6$\pm$2.2	4.4$\pm$0.9	1
Average	11.7$\pm$0.9	4.3$\pm$0.2	
$Ag^{106m}(i)$	17.3$\pm$0.5	13.1$\pm$0.5	45
	17.4$\pm$0.6	12.9$\pm$0.4	45
	16.9$\pm$0.3	13.1$\pm$0.3	5
Average	17.2$\pm$0.3	13.1$\pm$0.1	

[a] Weighted average. Weight for each value was taken as inversely proportional to its estimated error.

[21] John A. Panontin and Albert A. Caretto, Jr., J. Inorg. Nucl. Chem. 28, 1795 (1966).

The activities of Pd^{100} and Pd^{103} were determined by programming that section of the decay curve in which Rh^{100} is in equilibrium with Pd^{100}. The activity of Pd^{100} was corrected for the presence of Rh^{100} by an empirically determined factor. A least-squares analysis of three decay curves, in which the Pd^{100}-Rh^{100} activities were less than 0.1% of the total activity, gave a half-life of 16.9±0.1 day for Pd^{103}. An analysis of the residual decay curves of these samples after subtracting the contribution of Pd^{103} from the gross activity gave a half-life of 87.2±2.1 h for Pd^{100}.

The x-ray decay curves of the silver samples from both targets were programmed with 40- and 8.3-day half-periods to yield the corresponding activities of Ag^{105} and Ag^{106m}. The β decay curves of the samples from the silver targets were treated in the same manner. From these data, appropriate conversion factors of x-ray to β activity of Ag^{105} and Ag^{106m} were determined. The β activities of Ag^{111}, Ag^{112}, and Ag^{113} from the indium targets were then obtained by subtracting the β activities of Ag^{105} and Ag^{106m} calculated from their respective x-ray activities from the gross β activity.

The monitor activities were analyzed with 15.0-h and 2.60-year half-periods to yield the respective activities of Na^{24} and Na^{22}.

The activities of Ag^{101}, Ag^{103}, and Cd^{105} at the time of separation from the corresponding daughter activities were obtained by means of the standard relationship. The activities of these nuclides at the end of bombardment were obtained by use of the half-lives given in Table I. The resulting values were then increased by an estimated 3% to correct for scavenging losses in the milking experiments.

TABLE III. Formation cross sections of silver and palladium nuclides from indium with 200- and 400-MeV protons.

Nuclide	σ(mb) 200 MeV	σ(mb) 400 MeV	T(min) Length of irradiation
$Pd^{100}(c)$	2.58 ±0.05	8.79 ±0.07	45
	2.81 ±0.06	8.65 ±0.15	45
	3.04 ±0.09	9.01 ±0.15	5
Average	2.81 ±0.22	8.82 ±0.17	
$Pd^{101}(c)$	10.2 ±0.4	20.2 ±0.4	5
$Ag^{101}(c)$	1.5 ±0.0	3.8 ±0.1	1
	1.7 ±0.0	3.5 ±0.1	1
Average	1.6 ±0.1	3.7 ±0.2	
$Pd^{103}(c)$	24.0 ±0.4	36.8 ±0.1	45
	22.8 ±0.4	35.8 ±0.5	45
	24.4 ±0.7	37.5 ±0.7	5
Average	23.7 ±0.9	36.7 ±0.9	
$Ag^{103}(c)$	20.2 ±0.2	25.4 ±0.1	45
	20.8 ±0.4	25.8 ±0.7	45
	16.4 ±0.4	31.8 ±0.5	5
Average	19.1 ±2.4	27.7 ±3.6	
$Ag^{105}(c)$	38.6 ±0.6	40.1 ±0.6	45
	37.1 ±0.6	38.9 ±0.5	45
	39.0 ±1.3	41.3 ±0.8	5
Average	38.2 ±1.0	40.1 ±1.3	
$Cd^{105}(c)$	15.3 ±0.4	13.9 ±0.5	5
	16.0 ±0.4	13.0 ±0.4	5
Average	15.7 ±0.4	13.5 ±0.5	
$Ag^{106m}(i)$	14.6 ±0.6	16.9 ±0.6	45
	14.2 ±0.4	16.1 ±0.5	45
	14.3 ±0.3	17.0 ±0.5	5
Average	14.4 ±0.2	16.7 ±0.5	
$Ag^{111}(c)$	2.37 ±0.07	3.77 ±0.14	45
	2.34 ±0.04	3.88 ±0.05	45
	2.31 ±0.02	3.76 ±0.01	5
	2.25 ±0.05	3.54 ±0.07	1
	2.38 ±0.04	3.58 ±0.06	1
Average	2.33 ±0.02	3.71 ±0.06	
$Ag^{112}(i)$	1.20 ±0.07	1.82 ±0.09	1
	1.14 ±0.05	1.80 ±0.09	1
Average	1.17 ±0.03	1.81 ±0.01	
$Ag^{113}(i)$	0.042±0.039	0.299±0.048	1
	0.099±0.026	0.272±0.048	1
Average	0.076±0.031[a]	0.286±0.014	

[a] Weighted average. Weight of each value taken as inversely proportional to its estimated error.

TABLE IV. Comparison of measured cross sections of products from silver bombarded with 400-MeV protons.

Nuclide	σ(mb) Kurchatov *et al.*[a]	σ(mb) Present work
$Pd^{100}(c)$	13.4 (12.7)[b]	15.3±0.5
$Pd^{101}(c)$	20.4 (19.4)	28.4±0.5
Pd^{103}	10.7 (10.0)	22.4±2.6
$Ag^{103}(c)$	6.0 (5.4)	26.9±1.6
Ag^{105}	29.0 (26.2)	37.6±1.2
$Ag^{106m}(i)$	10.7 (10.0)	13.1±0.7
$Cd^{105}(i)$	1.7 (1.3)	4.3±0.2

[a] Reference 23.
[b] The values in parentheses are the values reported by the authors for 480-MeV protons.

III. RESULTS

The activities of each product were corrected for chemical yield, detector efficiency, recoil loss, aliquot, and disintegration conversion factors, and the cross sections calculated by the standard relationships. The measured cross sections are relative to that of the Al^{27} $(p,3pn)$ monitor reaction. The reported[22] values of 9.3 mb at 200 MeV and 10.5 mb at 400 MeV were used. The results are presented in Tables II and III, where column 1 gives the product nuclide and type of formation, i.e., (i) independent or (c) cumulative, columns 2 and 3 the cross section for 200- and 400-MeV protons, respectively, and column 4 the length of the irradiation in minutes. In those cases where more than one determination was made, the average cross section as well as the individual values is given. The uncertainties quoted for the average cross sections are the standard deviations in the mean. Those listed for the individual determination are estimates of the random errors associated with decay curve analysis, chemical yield determinations, and foil thickness measurements. The over-all uncertainty of the measurements is estimated

[22] J. B. Cumming, Ann. Rev. Nucl. Sci. 13, 261 (1963).

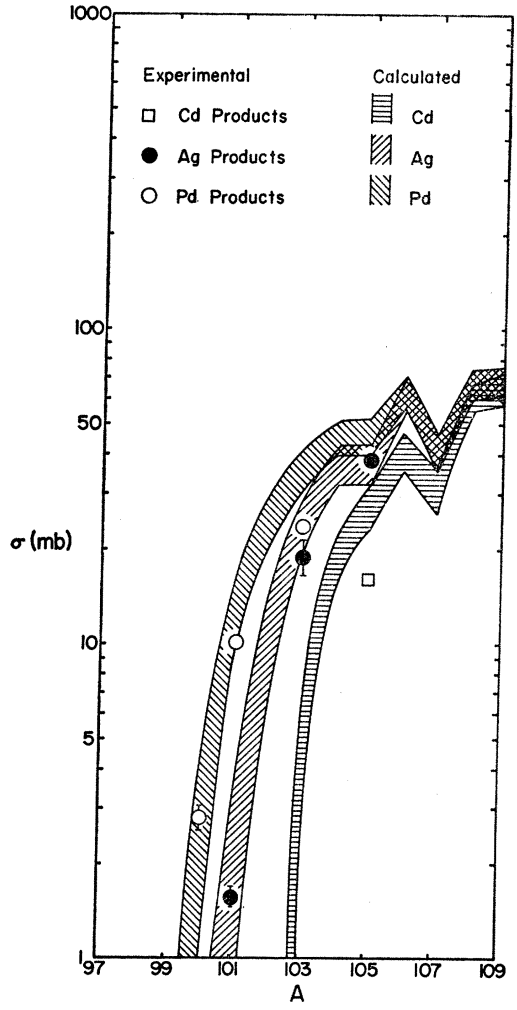

FIG. 1. Comparison of calculated cumulative cross sections with experimental values of observed Cd, Ag, and Pd nuclides from 200-MeV proton bombardment of natural indium. (Calculated curves based on Vegas cascade program with refraction.)

to be about 20% for the electron capture products and approximately 12% for the β emitters. This is mainly comprised of the systematic error associated with the efficiencies of the detectors (5%), the uncertainty in the cross section of the monitor reaction (6.5%), and the uncertainty in the conversion of x-ray activities into disintegration rates (10%).

The present results may be compared with the work of Kurchatov *et al.*[23] These authors studied the spallation of silver with high-energy protons. The most pertinent comparison may be made with their results at 480 MeV. Their data are compared with the present work in Table IV, a correction having been made for the difference in bombarding energy by means of the energy dependence found in the present work. The results of the two studies show rather poor agreement. The present cross sections thus are greater by factors of 1.2 to 4.5. Kurchatov *et al.*[23] report a reaction cross section of 430 mb from a summation of all yields, both measured and interpolated, and note a discrepancy between their reported value and that obtained from emulsion studies[24] for the same bombarding energy, 753±150 mb. The latter is in good agreement with the value 856±85 mb, estimated for silver bombarded with 400-MeV protons from an analysis of the present and other radiochemical data.[25] The cumulative cross sections are plotted as a function of mass number in Figs. 1–3. The same general trend is observable for both targets at the two bombarding energies, namely a decrease in cumulative cross

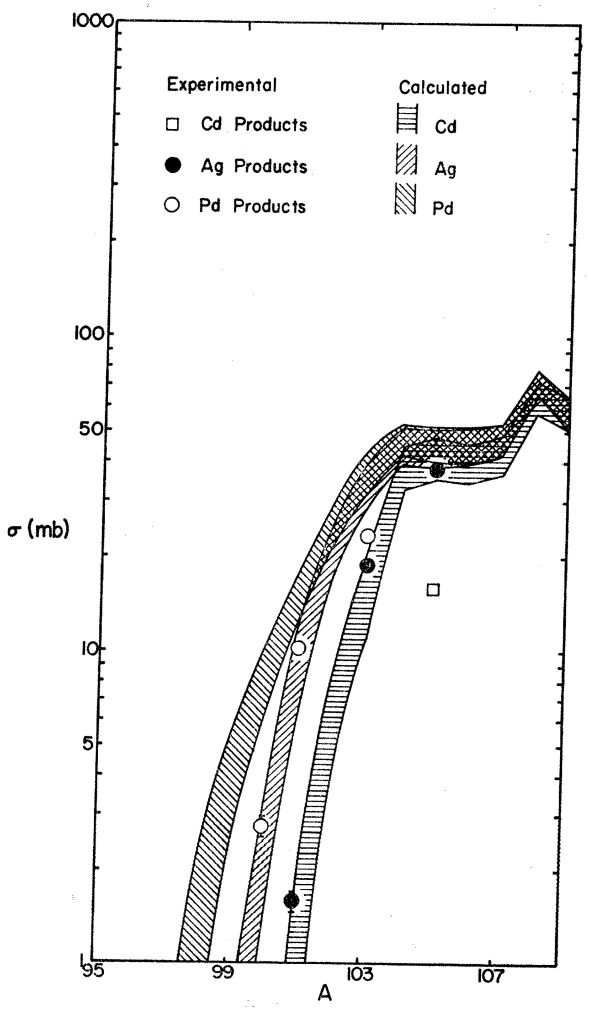

FIG. 2. Comparison of calculated cumulative cross sections with experimental values of observed Cd, Ag, and Pd nuclides from 200-MeV proton bombardment of natural indium. (Calculated curves based on Vegas cascade program without refraction.)

[23] B. V. Kurchatov, V. N. Mekhedov, N. I. Borisova, M. Ya. Kuznetsova, L. N. Kurchatova, and L. V. Chistyakov, in Peaceful Uses Atomic Energy, Moscow, 1955 (unpublished); U. S. Atomic Energy Comission Translation 2435, 1955, Part 2, p. 111, (unpublished).

[24] G. Bernardini, E. T. Booth, and S. J. Lindenbaum, Phys. Rev. 88, 1017 (1952).

[25] J. A. Panontin (to be published).

section with decreasing product mass. This trend is in the main a consequence of the shape of the deposition energy spectrum of the residual nuclei formed in the cascade. It is, of course, expected that the various curves will go through a maximum as the mass number of the product approaches that of the target.

The energy dependence of the measured cross sections is summarized in Table V. It is seen that the cumulative yields of the (p,xn), (p,pxn), and $(p,2pxn)$ products exhibit an inverse dependence on bombarding energy, whereas the cumulative yields of $(p,3pxn)$ and $(p,4pxn)$ products exhibit a direct dependence. These observations undoubtedly reflect a relative decrease with increasing proton energy in low-deposition energy events and a concomitant increase in high-deposition energy events. Further, the (p,xn) (p,pxn) and $(p,2pxn)$ reaction cross sections show an energy dependence that parallels that of the elementary nucleon-nucleon cross sections. This behavior is indicative of the importance of the cascade process in determining the yields of the simple nuclear reactions.

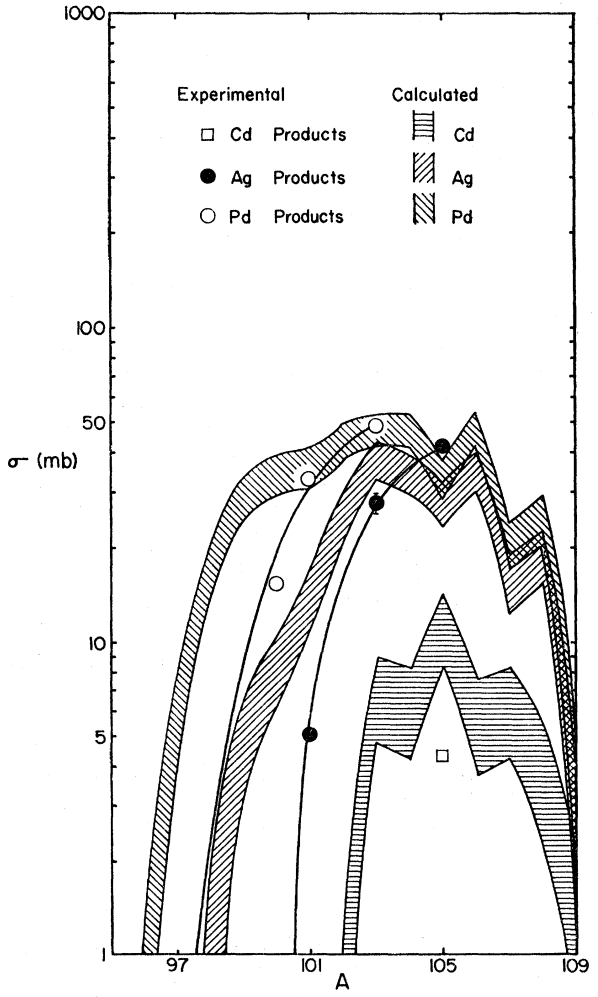

FIG. 3. Comparison of calculated cumulative cross sections with experimental values of observed Cd, Ag, and Pd nuclides from 400-MeV proton bombardment of natural silver. (Calculated curves based on Vegas cascade program with refraction.)

TABLE V. Calculated[a] and experimental ratios of the cumulative cross section of a product from 400-MeV protons bombardments to the value from 200-MeV proton bombardments of silver and indium.

Target	Product	Calculated[b]	Experimental
Ag	Pd^{100}	0.69± 0.11	0.82±0.05
	Pd^{101}	0.69± 0.11	0.83±0.03
	Pd^{103}	0.65± 0.09	0.73±0.06
	Ag^{101}	0.48± 0.12	0.57±0.03
	Ag^{103}	0.61± 0.16	0.66±0.16
	Ag^{105}	0.55± 0.09	0.69±0.02
	Cd^{105}	0.45± 0.13	0.36±0.03
In	Pd^{100}	17.2 ±10.5	3.14±0.26
	Pd^{101}	3.1 ± 0.9	1.98±0.09
	Pd^{103}	1.25± 0.24	1.55±0.07
	Ag^{101}	9.2 ± 5.8	2.30±0.13
	Ag^{103}	1.81± 0.41	1.44±0.25
	Ag^{105}	1.30± 0.25	1.05±0.04
	Cd^{105}	1.37± 0.31	0.86±0.04
	$Ag^{111-2-3}$	7.6 ± 8.7	1.62±0.04

[a] The calculated cross sections have been normalized by the appropriate factors in Table IV.
[b] The calculated cross sections used to determine the ratios in this column are based on the Vegas program with refraction, STEP.

The measured cross sections may be used to infer a number of independent yields. The independent cross sections for the formation of Pd^{103} and Pd^{101} may thus be obtained directly from the respective measured values by subtracting the cumulative cross sections of Ag^{103} and Ag^{101}. The independent cross sections for the formation of the latter may in turn be obtained with the aid of estimated[25] values of the cumulative cross sections of the corresponding cadmium nuclides. Since the contribution of these very neutron-deficient products is small, reasonably accurate independent yields may be obtained. The independent cross sections obtained in this fashion are summarized in Table VI and the corresponding isobaric yield ratios are given in Table VII.

The experimentally determined isobaric yield ratios exhibit an unusual dependence on the neutron-to-proton ratio (N/Z) of the target. In all but one instance the more neutron-deficient member of an isobaric pair, e.g., Ag^{103}, is preferentially formed from the more neutron-rich target, i.e., In^{115}. This result is in sharp contrast with the observation by Porile and Church[10] that, other factors being equal, the more neutron-deficient isobaric products will preferentially be formed from the more neutron-deficient target and the more neutron-excessive products from the more neutron-rich target. This conclusion was based on a study of isobaric yields of products with $A=72$ formed in reactions of 1.8-GeV protons with Zr^{96}, Mo^{96}, and Ru^{96}. The data of Kaufman[26] on the formation of the $A=72$ isobars from heavier elements support the conclusions of Porile and Church.[10] These conclusions can be regarded as a modification of the earlier observation by

[26] Sheldon Kaufman, Phys. Rev. 128, 1866 (1963).

TABLE VI. Comparison of experimentally determined independent formation cross sections with Monte Carlo calculations.

Bombarding energy	Target	Product	σ(expt)	σ(calc)[a]	σ(calc)[b]	σ(calc)[c]
200 MeV	Silver	Cd^{105}	11.7±0.9	24.9±4.2		
		Ag^{105}	49.4±1.5	25.6±4.3		
		Ag^{103}	40.5±2.1	48.4±5.9		
		Pd^{103}	25.2±3.1	11.9±2.8		
		Ag^{101}	8.8±0.5	27.1±4.4		
		Pd^{101}	31.1±1.7	24.5±4.2		
		Pd^{100}	16.3±1.6	34.0±4.9		
	Indium	Cd^{105} [d]	15.7±0.4	27.6±4.6	41.7±6.0	
		Ag^{105}	22.5±1.4	9.7±2.7	4.3±2.0	
		Ag^{103}	19.0±2.3	20.9±4.0	20.0±4.2	
		Pd^{103}	3.7±1.0	13.4±3.2	4.3±2.0	
		Ag^{101}	1.6±0.1	2.2±1.3	8.7±2.7	
		Pd^{101}	8.6±0.5	10.4±2.8	6.1±2.3	
		Pd^{100}	2.9±0.3	2.2±1.3	9.6±2.9	
400 MeV	Silver	Cd^{105}	4.3±0.2	11.3±2.9		5.6±1.9
		Ag^{105}	37.6±1.2	16.5±3.2		20.7±3.6
		Ag^{103}	27.3±1.7	30.9±4.4		26.4±4.1
		Pd^{103}	21.6±2.6	10.1±2.5		13.2±2.9
		Ag^{101}	5.1±0.0	13.3±2.9		15.7±3.2
		Pd^{101}	28.2±0.5	22.8±3.8		23.8±3.9
		Pd^{100}	14.0±0.9	27.2±4.2		28.2±4.2
	Indium	Cd^{105} [d]	13.5±0.5	37.9±5.7		43.1±7.5
		Ag^{105}	26.6±1.8	10.6±3.0		14.4±4.3
		Ag^{103}	27.4±3.5	28.2±5.0		13.1±4.2
		Pd^{103}	9.5±1.2	3.6±1.8		9.2±3.4
		Ag^{101}	3.7±0.1	17.7±4.0		14.4±4.3
		Pd^{101}	16.5±0.5	18.5±4.0		17.0±4.7
		Pd^{100}	9.1±0.4	27.3±4.9		22.2±5.4

[a] Vegas with refraction. [b] Vegas without refraction. [c] Metropolis *et al.* [d] Cumulative yields.

Miller and Hudis[27] that the isobaric yield ratios are independent of target provided the products are at least a few mass and atomic numbers removed from the target. The latter conclusion is still expected to be valid, provided the various targets have nearly the same N/Z.

IV. COMPARISON WITH MONTE CARLO CALCULATIONS

The experimental results may be compared with the Monte Carlo cascade calculations by Chen *et al.*[17] and by Metropolis *et al.*[16] The Vegas program,[17] in its STEP version, takes into account the radial variation of the nuclear density by dividing the nucleus into seven concentric zones of different constant density. Refraction and reflection of the cascade particles at the interfaces of these zones are included in the calculation. On the other hand, the Metropolis code[16] assumes a nucleus of constant density and does not consider reflection and refraction. The output of each of these calculations consisted of about 500 cascades each of which is characterized by the atomic number Z, mass number A, and excitation energy U, of the product nucleus.

The Metropolis calculation was compared with the 400-MeV data. The calculation for Ru^{100} bombarded by 460-MeV protons was used for this purpose. The Z and A values of the cascade products were appropriately shifted to correspond to Ag^{107}, Ag^{109}, and In^{115} targets. The slight difference in energy between experiment and calculation primarily manifests itself in a small difference in the number of predicted particles emitted in either the cascade or evaporation phase, this number being smaller at 400 MeV. On the other hand, the difference in target mass between experiment and calculation leads to a small effect in the opposite direction because of the increase in the average excitation energy with target mass. It was assumed that these two small effects balanced each other so that the only allowance made for the differences between experiment and calculation was the above-mentioned shift in the Z and A values.

The Vegas calculation[17] was programmed for 200- and 400-MeV protons incident on Ag^{108} and In^{115}. The (Z,A) distribution for Ag^{108} was shifted to correspond to Ag^{107} and Ag^{109}. The computation at 200 MeV was also performed with the STEPNO version of the program in which refraction and reflection effects are not considered.

The cascade products were used as the starting nuclei for an evaporation calculation based on the Monte Carlo code of Dostrovsky *et al.*[18] The calculation used a level density parameter $a=A/10$, pairing energies listed by Cameron,[28] and the inverse reaction cross-section

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TABLE VII. Experimental and calculated isobaric yield ratios.

Bombarding energy	Target	Ratio	R(expt)	R(calc)[a]	R(calc)[b]	R(calc)[c]
200 MeV	Silver	Cd^{105}/Ag^{105}	0.24±0.02	0.97±0.23		
		Ag^{103}/Pd^{103}	1.6 ±0.2	4.1 ±1.1		
		Ag^{101}/Pd^{101}	0.28±0.02	1.1 ±0.3		
	Indium	Cd^{105}/Ag^{105}	0.70±0.05	2.8 ±0.9	9.7±4.8	
		Ag^{103}/Pd^{103}	5.1 ±1.5	1.6 ±0.5	4.7±2.4	
		Ag^{101}/Pd^{101}	0.19±0.02	0.21±0.14	1.4±0.7	
400 MeV	Silver	Cd^{105}/Ag^{105}	0.11±0.01	0.68±0.22		0.27±0.10
		Ag^{103}/Pd^{103}	1.3 ±0.2	3.1 ±0.9		2.0 ±0.5
		Ag^{101}/Pd^{101}	0.18±0.00	0.58±0.16		0.66±0.17
	Indium	Cd^{105}/Ag^{105}	0.51±0.04	3.6 ±1.2		3.0 ±1.0
		Ag^{103}/Pd^{103}	2.9 ±0.5	7.8 ±4.1		1.4 ±0.7
		Ag^{101}/Pd^{101}	0.22±0.01	0.96±0.31		0.85±0.35

[a] Vegas with refraction. [b] Vegas without refraction. [c] Metropolis.

constants given by Dostrovsky *et al.*[18] The calculation was performed with the Purdue-7094 computer. In order to improve the over-all statistics of the calculation, three or four evaporation iterations were performed for each cascade product. In the case of the silver target two or these iterations were performed for cascade products from Ag^{107} and two for the corresponding products from Ag^{109}. The resulting yield distributions were combined for comparison with experiment.

A direct comparison of the experimental data with the calculated cross sections showed that the latter were, in general, substantially larger than the former. Consequently, the calculated values were normalized so that the total calculated reaction cross sections were equal to their respective experimental values. The experimental reaction cross sections used are 820 and 865 mb for silver with 200- and 400-MeV protons, respectively, and 855 and 905 mb for indium at the corresponding energies. These values were obtained from an analysis of the present work and previously reported data and are described elsewhere.[25] The quoted values for indium are considerably smaller than the reported[29] reaction cross section for 180-MeV protons incident on indium, 1165±34 mb. In view of this discrepancy it seems more reasonable to consider the normalization as an arbitrary adjustment designed to bring the over-all calculated yields into approximate agreement with experiment. The origin of this normalizing factor can either be the above-mentioned overestimate of the calculated reaction cross section or a gross distortion of the calculated mass yield curve resulting in an overestimation of the cross sections of the products of interest. The normalization factors are listed in Table VIII. These values may be compared to the value 0.60±0.01 found necessary by Porile and Church[10] to normalize the yields predicted by the Metropolis calculation[16] for the $A=72$ isobars to the experimental values from 1.8-GeV proton bombardment of several targets with $A=96$. The second of the above possibilities for such a discrepancy was invoked in that work.

Some of the results from the 200-MeV proton bombardments are presented graphically in Figs. 1 and 2, in which the cumulative cross section of a product is plotted against its mass number, as well as in Table VI. The shaded areas represent the calculated values plus or minus their standard deviation. The calculated cumulative cross sections of the silver and palladium products from silver are in good agreement near the experimentally observed maximum values for these products (Pd^{103} and Ag^{105}) but are in rather poor agreement as the product mass decreases. The calculated cross section of the (p,xn) product, Cd^{105}, is also in poor agreement. Good (Fig. 1) to rather poor (Fig. 2) agreement is found for the calculated cumulative cross sections of the observed silver and palladium products from indium. The curves of Fig. 1 represent the results obtained from the Vegas STEP program, whereas those of Fig. 2 represent the results obtained from the Vegas STEPNO program.

A more detailed comparison is presented in Table VII, in which the experimental isobaric yield ratios are compared with the calculated values. It is seen that in the case of silver the calculated values are consistently larger than the experimental ones. The same trend is observable for indium, albeit less consistently. These observations suggest that the cascade-evaporation calculation underestimates the emission of charged particles.

The results at 400 MeV are presented in Table VI

TABLE VIII. Factors used in normalizing results from Monte Carlo calculations.

Target	Proton energy (MeV) 200	400	460
Ag	0.585[a]	0.701[a]	0.790[b]
In	0.596[a]	0.709[a]	0.790[b]
	0.698[c]		

[a] Vegas program with refraction.
[b] Metropolis program.
[c] Vegas program without refraction.

[29] A. Johansson, U. Svanberg, and O. Sundberg, Arkiv Fysik **19**, 527 (1961).

and the comparison for the cumulative yields from silver is shown in Fig. 3. The comparison reveals substantially the same features as at 200 MeV. The calculated cumulative cross sections of palladium and silver products thus are in good agreement near the experimentally observed maximum values for these products. On the other hand, the calculations overestimate the yields of the most neutron-deficient palladium and silver products.

The comparison of calculated and experimental isobaric yield ratios again reveals that the calculation underestimates the emission of charged particles. Similar conclusions have previously been drawn for both the Vegas and Metropolis calculations from a number of studies in the heavy-element region[1,7,9,30] at comparable bombarding energies. This discrepancy can be attributed to either the cascade or the evaporation calculation. The validity of the latter has been checked by comparison of the calculated cross sections for the reactions of 20–40-MeV He^4 ions with Ag^{107} with the experimental excitation functions of Fukushima *et al.*[31] Using a value of $a=A/20$, which is the appropriate value for intermediate-energy compound nuclear reactions and the particular formulation of Dostrovsky *et al.*,[18] reasonably good agreement with experiment was obtained. For instance the calculated value of $\sigma(\alpha,pn)/\sigma(\alpha,2n)$ at an energy corresponding to the maximum of the $(\alpha,2n)$ excitation function is 0.05; the corresponding experimental value is 0.06. Similar agreement has been obtained between experiment and the same calculation for the reactions of Cd^{106} with He^4 ions,[32] albeit with a smaller value of a. It would thus appear that while the evaporation calculation may somewhat underestimate charged-particle emission in the mass region of interest, it probably cannot account for the factor of 4 that is required by many of the present isobaric ratios.

An important part of the discrepancy in isobaric yield ratios can thus be attributed to the cascade calculations. One common feature of the Metropolis and Vegas calculations is that the possible emission of complex particles, such as deuterons and α's, is not considered. In recent years ample evidence concerning the emission of prompt complex particles has been obtained. The emission of α particles in high-energy reactions has been studied using emulsion,[33–37] radiochemical,[38–40] and counter[41] techniques. The energy spectra, angular distributions, and emission probabilities of these particles have been analyzed[33–35,41,42] and explained in terms of emission during both the evaporation and cascade steps. Although the former mechanism appears to be the more important one, the contribution from the cascade process is far from negligible and, in fact, becomes comparable in magnitude at higher bombarding energies. The inclusion of this process in the cascade calculations would have the effect of reducing the discrepancy between experiment and calculation found in the present study.

The energy dependence of the cumulative cross sections is compared with the calculation based on the Vegas program with refraction in Table V. It is seen that the calculation can qualitatively reproduce the observed energy dependence. Both the calculated and experimental cross sections of the (p,xn) (p,pxn), and $(p,2pxn)$ reactions thus decrease with increasing energy, whereas those of the $(p,3pxn)$ and $(p,4pxn)$ reactions show the opposite trend. Evidently the factors responsible for the discrepancy in the isobaric yield ratios do not have a strong dependence on bombarding energy.

V. CONCLUSIONS

The cross sections of a number of $(p,ypxn)$ reactions of indium and silver with 200- and 400-MeV protons have been measured and compared with cascade-evaporation calculations. Two significant discrepancies have been noted. First, the calculation predicts substantially larger cross sections than are obtained experimentally. It is tempting to attribute this discrepancy to an overestimation of the calculated reaction cross section. Unfortunately, the lack of agreement between the radiochemical[25] and counter[29] experiments as to the value of the reaction cross section precludes a definite conclusion.

Second, the calculation underestimates the relative number of emitted charged particles as evident by the consistently larger values of the isobaric yield ratios. Although this discrepancy may in part be due to a shortcoming of the evaporation code, it also appears to be associated with the cascade calculation. It is suggested that the emission of complex particles during the intranuclear cascade may account for this effect.

The experimental results have been compared with three different cascade calculations. The comparison at 200 MeV indicates that the Vegas program with refraction, STEP, gives better agreement than that in which

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refraction is not included, STEPNO. This is true for both the actual cross sections and the isobaric ratios. On the other hand, the program without refraction is in better agreement with the estimated total reaction cross section, in the sense that the normalization factor for this calculation is closer to unity. The comparison at 400 MeV indicates that the Metropolis calculation is in somewhat better agreement with both the measured cross sections and the isobaric yield ratios than the Vegas program with refraction. This reflects the fact that the former computation predicts an average of 1.5 protons emitted per cascade compared with a value of 1.0 for Vegas.

ACKNOWLEDGMENTS

The authors wish to thank the operating staff of the Carnegie Institute of Technology Nuclear Research Center for their cooperation in carrying out the irradiations and C. Hombach for her laboratory assistance. The Vegas cascade calculations were kindly supplied by Dr. G. Friedlander.

PHYSICAL REVIEW VOLUME 165, NUMBER 4 20 JANUARY 1968

Recoil Properties of Products from the Spallation of Silver and Indium*

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(Received 7 June 1967)

Average projected recoil ranges in the forward, backward, and perpendicular direction to the beam have been measured for a number of palladium and silver products formed in the interaction of silver and indium with 200- and 400-MeV protons. The values of the forward and perpendicular ranges increase with the mass difference between target and product and the dependence becomes exponential for $\Delta A > 7$. The results have been compared with a Monte Carlo cascade-evaporation calculation based on the Vegas cascade code. The calculation included a correction for scattering effects. It is found that the calculated values are in moderately good agreement with the measured values of the forward and perpendicular ranges, but underestimate the values of the backward range.

I. INTRODUCTION

THE formation of products from the interaction of high-energy particles with complex nuclei is considered to occur by one or more of the following mechanisms: spallation, fission, or fragmentation. Of these three mechanisms, only the spallation process has thus far been quantitatively formulated to predict the yields and recoil properties of final products. This process is usually described in terms of a prompt intranuclear cascade initiated by the incident particle and a slower evaporation stage. The analysis of each of these stages is usually performed by means of Monte Carlo calculations programmed for computer iteration.[1–3] The results can be compared with experimental data, and a number of such comparisons have been reported for both cross sections[4,5] and recoil properties.[6–10]

The simplest recoil experiment, and that which has been most extensively used, employs a thick target and 2π geometry for the catcher foils.[11,12] This type of experiment provides only limited recoil information, namely, the average projected range of a product nuclide along a particular axis. The determination of

* Supported by the U. S. Atomic Energy Commission.
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