Charged-particle emission in reactions of ^{90}Zr with 10–86-MeV protons

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Excitation functions for 12 nuclear reactions of type ^{90}Zr[$(p,(2–4)p, (1–6)n$] have been measured for incident proton energies from 10–86 MeV. The complex excitation functions have been decomposed to yield a component due to emission of one or two α particles, and a component due to the emission of individual nucleons. Combining the α particle portions of the decomposed excitation functions, the excitation energy spectra for those events associated with α particle emission have been derived. Having estimated the excitation functions for a few missing reactions, the total probability of emission of seven combinations of charged particles as a function of incident proton energy has been calculated. The implications of the results on the preequilibrium model of nuclear reactions are discussed.

[NUCLEAR REACTIONS ^{90}Zr$(p, ypxn)$, $y=2–4$, $x=1–6$, $E=10–86$ MeV; measured $\sigma(E)$; deduced α particle yields, charged-particle emission probabilities. Enriched target.]

INTRODUCTION

In a previous paper[1] we reported the results of experiments measuring the excitation functions of $(p, 2pxn)$ reactions for 10–86-MeV protons incident upon ^{202}Hg. Reactions having $x>1$ exhibited two peaks in their excitation functions, the first at, or below, the threshold energy for emission of the requisite nucleons. A procedure was presented for decomposition of the excitation functions into two components; one arising from emission of an α particle, and a second at higher energies from emission of individual nucleons. In addition the α particle component was separated into α particles arising from direct processes and those involving "evaporation" from an excited intermediate nucleus. Finally, the excitation energy spectrum of the excited intermediate nucleus was calculated based on the experimental results.

The target nucleus ^{202}Hg was heavy and highly neutron excessive. It was felt that further information on the process of α particle emission might be gained from a study of a medium weight, neutron deficient nucleus. It was hoped to establish a relationship between the probability of emission of α particles and the mass of the nucleus A, as well as its neutron excess N/Z. Of greater import, however, was extending and testing the procedures described above to the case of $(p, 3pxn)$ and $(p, 4pxn)$ reaction products. In particular, measurable cross sections for $(p, 4pxn)$ products at low incident proton energies might be attributable to processes involving the emission of two α particles in a single interaction. Comparison of the excitation energy distributions in $(p, 2pxn)$, $(p, 3pxn)$, and $(p, 4pxn)$ reactions should also provide comprehensive information on the competition of neutron, proton, and α particle emission.

The target nuclide chosen was the most neutron deficient isotope of zirconium, ^{90}Zr. The products of the three reactions mentioned above are isotopes of Y, Sr, and Rb, most with reasonably well known decay properties and reasonable half-lives. We report the results of experiments performed with protons of 10–86 MeV incident upon ^{90}Zr to measure the excitation functions of the following reactions: ^{90}Zr$(p, 2pxn)$, $x=1–5$; $(p, 3pxn)$, $x=3–6$; and $(p, 4pxn)$, $x=3–6$.

There is little work of a similar nature in the literature. The most extensive is that of Sharp, Diamond, and Wilkinson,[2] who measured excitation functions for up to 100 MeV protons on ^{59}Co: $[p, 2p(5\,\&\,6)n]$, $[p, 3p(1, 3, \&\,5)n]$, and $[p, 4p(5\,\&\,8)n]$. Other than this, Meadows and Holt measured the ^{25}Mg$(p, 4p4n)^{18}$F excitation function to 120 MeV,[3] and Tanaka studied Ni$(p, 4p3n)$ up to 57 MeV.[4] Several other authors have measured some $(p, 3pxn)$ excitation functions in conjunction with $(p, 2pxn)$ studies.[5–8] These have for the most part involved only a few product nuclides; no systematic measurements on series of nuclides have been reported.

The mechanism of emission of α particles together with the more general question of emission of all charged particles in "simple" reactions is a complex one. "On-line" studies measuring the energy spectra and angular distributions of α particles emitted in nuclear reactions provide strong evidence for two modes of production.[9–15] The first, characterized by strong forward peaking and high kinetic energies, is usually attributed to direct processes. The second, characterized by

lower kinetic energies and more nearly isotropic angular distributions, is usually attributed to an "evaporation" process. Until recently, however, calculations of the emission of charged particles and, in particular, of α particles, greatly underestimated the yields of these particles, and could not at all account for the magnitude of the "evaporation" component. This underestimate arose in the calculations from the inability of charged particles to surmount the Coulomb barrier relative to modes of deexcitation involving uncharged species (neutrons, or even γ rays). In turn, this low probability is a direct consequence of the assumption that "evaporation" occurs only after the attainment by the excited nucleus of a state of statistical equilibrium. Recently attempts have been made using various "preequilibrium" models of nuclear reactions to enhance the yield of charge particles by providing for emission from a nucleus which has not yet attained statistical equilibrium. These calculations are discussed extensively in Blann's recent review article.[16]

Unfortunately, the experimental data necessary for testing such calculations has been sparse. "On-line" studies measuring the extent of α particle emission provide no information on the identity or number of nucleons which may also have been emitted in the reaction. Therefore no estimates of the excitation energy of the emitting nucleus may be made, and no calculations of the energy dependence of the competition between charged and uncharged particle emission can be attempted. However, as previously shown,[1] a complete set of excitation functions may be used to define the spectrum for those particular events in which an α particle is emitted. It is apparent that such calculations are improved by taking account of three or four proton, or two α, emission. The excitation energy spectra derived from the excitation functions may then be combined with them and an estimate of the total reaction cross section to calculate the absolute probability of emission of various combinations of charged particles as a function of incident proton energy.

The goals of this work then were twofold: first, to extend our previous study of α particle emission to a medium weight, neutron deficient target nucleus; and second, to continue our study of charged particle emission from highly excited nuclei.

From the first, we have decomposed the experimental excitation functions to derive the cross sections for α particle emission, and have compared the direct interaction and "evaporation" components both to each other and to those from the ^{202}Hg target.

With regard to the second, we have calculated the energy dependence of the emission of charged particles and related the findings to what may be expected from the preequilibrium model of nuclear reactions.

PROCEDURE AND RESULTS

The target material consisted of a uniform mixture of enriched ^{90}Zr as ZrO_2 and spectroscopically pure CuO. The former, obtained from Oak Ridge National Laboratory, contained 97.7% ^{90}Zr and less than 1% of each of the other stable isotopes. The chemical purity of ZrO_2 was 99.8%. No elements of interest to this work were among detectable impurities.

The powdered target was contained in a thin walled aluminum tube and irradiations were performed as in previous experiments at the McGill synchrocyclotron.[17] Irradiations were for periods of about 1 min at a nominal beam intensity of 0.6 μA. The beam was monitored by the known absolute cross sections for the $^{63}Cu(p, n)^{63}Zn$[18] and $^{65}Cu(p, p, pn)^{64}Cu$ reactions.[19]

Some experiments required multiple timed separations to determine the independent yields of nuclides related by β decay. In these experiments, ZrO_2 was irradiated and cross sections measured in this work for the $^{90}Zr(p, 2pn)^{88}Y$ were used as a secondary monitor.

After irradiation, the target material was dissolved in a 10 : 1 mixture of concentrated HF and HNO_3 containing a known amount of Rb^+ and NbF_7^{2-} and ~0.1 mg each of Zn^{2+}, Ni^{2+}, Co^{2+}, and Fe^{3+} as holdback carriers. The solution was heated for several minutes in a hot water bath to effect complete dissolution and oxidation of the target. Carrier amounts of Sr and Y were added to the hot solution which was further heated to ensure isotopic exchange. The solution was then cooled in ice and the mixed fluorides of Sr, Zr, and Y centrifuged. This precipitate was treated to effect separation and purification of Sr and Y isotopes, while the supernate was treated to isolate Nb, Rb, and the Cu monitor. An outline of the chemical procedure follows. Details may be found in Ref. 20.

The precipitate was dissolved in HNO_3 and saturated H_3BO_3. Zirconium was removed by scavenging with Na_2HPO_4, followed by passing of the supernate through a bed of powdered Teflon coated with HDEHP preequilibrated with 10 M HNO_3.[20,21] The Sr^{2+} and Y^{3+} are separated from each other by treatment of the eluent with fuming HNO_3 precipitating $Sr(NO_3)_2$. The Sr was scavenged with $Fe(OH)_3$ and prepared for counting by precipitation with $(NH_4)_2C_2O_4$ as $SrC_2O_4 \cdot H_2O$. The Y was purified by extraction into TBP, back extraction into water, precipitation as the hydroxide, dissolution

in HCl, and finally precipitation with saturated $H_2C_2O_4$ as $Y_2(C_2O_4)_3 \cdot 9H_2O$. Decantation of the $Sr(NO_3)_2$ and $Y(OH)_3$ precipitates was taken as the time of separation of Sr from parent Y and the initial time for growth of Sr in the two samples, respectively. After activity measurements were completed, both the Sr and Y samples were ignited at 800°C and the chemical yield determined by weighing SrO and Y_2O_3, respectively. The mean chemical yields were about 48% for Sr and 46% for Y.

The supernate from the initial fluoride precipitation was scavenged several times with $BaZrF_6$ to ensure complete removal of Zr. The solution was evaporated with concentrated HNO_3 to near dryness at which point Nb_2O_5 precipitates. The remaining solution was treated with HCl to destroy nitrate and evaporated to dryness. After dissolving in 4.5 *N* HCl, Cu^{2+} and Rb^+ are separated from each other on a column of Dowex-1 ion exchange resin. The effluent was evaporated to dryness, dissolved in diluted HCl, and $RbClO_4$ was precipitated in the presence of Sr holdback carrier. After dissolution in water, Rb^+ was precipitated, weighed, and counted as Rb_2PtCl_6. The mean chemical yield of Rb was 40%.

The Cu^{2+} was then eluted from the column with 1.5 *N* HCl, reduced to Cu^+, and precipitated, weighed, and counted as CuSCN in the usual procedure of this laboratory.[22] The average chemical yield was 50%.

In timed experiments for yttrium isotopes growing from zirconium parents, the target of pure ZrO_2 was dissolved as described above in the presence of Nb holdback carrier, and after heating the resultant solution was divided in half. The two solutions were treated identically about 6 h apart. Yttrium carrier was added, precipitating YF_3 and coprecipitating the Zr parent isotopes. Corrections were then made for decay of the parent nuclides by simultaneously measuring the activity of parent and daughter by two different γ rays in each of the two samples. The procedure is easily extended to those cases in which two isomers of the yttrium isotopes exist.

Samples were counted on a 42-cm^3 Ge(Li) detector coupled to a 1600 channel pulse height analyzer. Absolute efficiencies at identical source-to-detector distances were determined from efficiency curves derived using standard IAEA sources. Nuclear properties of the nuclides of interest in this work are listed with references in Table I. Tabulated photon abundances were calculated from transition intensities and conversion

TABLE I. Nuclear properties of products from irradiations of targets containing zirconium.

Nuclide	Photon energy (keV)	Photon abundance [a]	Half-life	Isomeric transition abundance [b]	References
^{90}Nb	1129	92	14.59 h		36
$^{89}Nb^m$	587 [c]	93.1	66 min	0	37
$^{89}Nb^g$	909 [c]	98.8	2.03 h		37
^{88}Y	898	95.7	107 day		38
$^{87}Y^m$	381	77	13.3 h	98.5	20, [d] 39
$^{87}Y^g$	485	91	80.3 h		39
$^{86}Y^m$	208	94	48 min	99.31	40
	1077	0.69			41
$^{86}Y^g$	1077	82.5	14.6 h		40
$^{85}Y^m$	151 [c]	11.6	2.68 h	0	42
	232 [c]	85.0			
$^{85}Y^g$	232	33.6	4.8 h		42
$^{84}Y^g$	793	96	37 min		20, [d] 43
$^{85}Sr^m$	232	85.0	69.5 min	87.9	42
$^{85}Sr^g$	514	99.2	65.19 day		42
^{83}Sr	763	33.1	32.4 h		44, 45
^{82}Sr	777 [c]	13.2	25 day		44, 46
$^{84}Rb^g$	882	75.3	33.0 day		47
^{83}Rb	521	46	83 day		44
$^{82}Rb^m$	777	84	6.2 h	0	46
$^{81}Rb^g$	446	23.5	4.58 h		48
^{64}Cu	511	38.6	12.82 h		35
^{63}Zn	670	8.47	38.8 min		34, 49 [d]

[a] Photons emitted per 100 decays.
[b] Transitions to ground state per 100 decays of metastable state.
[c] Photon emitted in decay of daughter.
[d] Reference for half-life only.

TABLE II. Cross sections (in mb) for ^{90}Zr(p, $2pxn$) reactions.

Proton energy (MeV)	^{88}Y (p, $2pn$)	^{87}Y [a] (p, $2p2n$)	^{86}Y [a] (p, $2p3n$)	^{85}Y [a] (p, $2p4n$)	^{84}Y^{g} [b] (p, $2p5n$)
12		1.0± 0.1			
16		5.1± 0.3			
19		12.1± 0.7			
24		26 ± 2	2.1± 0.1		
30	3.6± 0.1	18 ± 1	41 ± 2		
38	27.6± 1.1	9 ± 3	57 ± 2	3.6± 0.3	
44	40 ± 2	...	28 ± 1	25 ± 1	
47	...	40 ± 10	...	...	
50	46 ± 3	...	14 ± 1	45 ± 3	
56	45 ± 3	108 ± 25	13.3± 0.8	29 ± 2	7.7± 1.0
62	38 ± 4	...	29 ± 2	15.4± 0.9	19.3± 1.8
68	42 ± 3	114 ± 16	82 ± 5	12.9± 0.6	23 ± 2
74	41 ± 3	...	117 ± 7	16.3± 0.5	15.1± 2
80	41 ± 3	...	122 ± 9	34 ± 2	10.2± 1
86	44 ± 3	102 ± 22	118 ± 8	57 ± 4	9.6± 1

[a] Sum of cross sections for individual isomers.
[b] Ground state only.

coefficients. Decay curves were analyzed by the weighted least squares procedure known as ORGLS.[23]

The disintegration rate at the end of irradiation was calculated from the observed count rate by correcting for decay, counter efficiency, absorption, photon abundances, and chemical yield. Absolute cross sections were determined by comparision of the calculated disintegration rate for the product nuclei to that of ^{64}Cu or ^{63}Zn, and thence to the monitor cross sections.[18,19] Uncertainties in the reported cross sections reflect the errors associated with decay curve analysis, detector efficiencies, published uncertainties in decay schemes, and chemical yields. The reported beam energy is the nominal average energy at a given synchrocyclotron radius, estimated to have an uncertainty of ±2 MeV, corrected for energy loss in the thin target.

The measured cross sections are tabulated in Tables II and III and shown in Figs. 1–3. Throughout the following discussion, the following convention will be used. A reaction of form (p, $2pxn$) will represent the overall cross section or excitation function of a given product nuclide, regardless of whether or not α particles are involved. A reaction of form (p, αxn) will be given to mean only that portion of the total cross section (or excitation function) which the authors will argue *does* represent emission of an α particle-cluster.

TABLE III. Cross sections (in mb) for ^{90}Zr(p, $3pxn$) and ^{90}Zr(p, $4pxn$) reactions.

Proton energy (MeV)	^{85}Sr (p, $3p3n$)	^{83}Sr [a] (p, $3p5n$)	^{82}Sr [a] (p, $3p6n$)	^{84}Rb (p, $4p3n$)	^{83}Rb (p, $4p4n$)	^{82}Rb [b] (p, $4p5n$)	
38	7.4± 0.4				0.28± 0.02		
44	18.9± 0.9				0.77± 0.05	0.134± 0.013	
50	27 ± 2			0.024± 0.003	0.51± 0.03	0.97 ± 0.07	
56	25 ± 2			0.38 ± 0.02	0.37± 0.02	2.0 ± 0.1	0.32± 0.06
62	16.5± 0.8	1.2± 0.1		1.00 ± 0.06	0.26± 0.05	1.8 ± 0.1	1.8 ± 0.1
68	12.4± 0.7	13.4± 0.8		2.1 ± 0.1	1.80± 0.11	1.6 ± 0.1	4.0 ± 0.2
74	15.7± 1.1	38 ± 2		2.5 ± 0.2	5.5 ± 0.4	0.95 ± 0.05	4.6 ± 0.3
80	18 ± 1	45 ± 3	4.8± 1.6	2.4 ± 0.2	7.6 ± 0.9	1.9 ± 0.1	3.1 ± 0.2
86	34 ± 2	48 ± 3	19.5± 2.1	2.2 ± 0.1	11.6 ± 0.7	6.5 ± 0.4	2.9 ± 0.2

[a] Cumulative cross sections.
[b] Metastable state only.
[c] Ground state plus unknown fraction of metastable state.

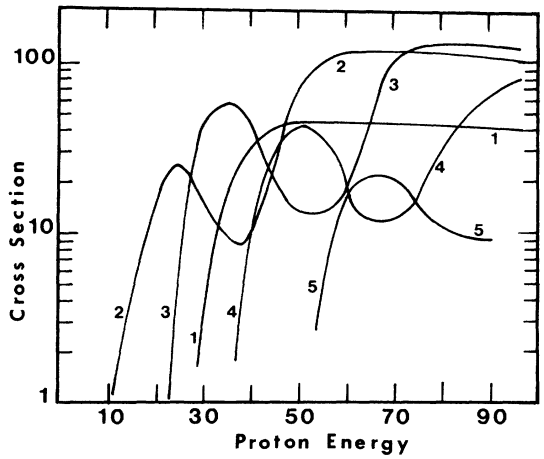

FIG. 1. Excitation functions for $^{90}Zr(p, 2pxn)$ reactions. Each curve is labeled by the value of x. Cross sections are in mb; energy in MeV.

DISCUSSION

A. Experimental results

1. [p,2p(1–5)n] excitation functions

The behavior of the excitation functions for the reactions shown in Fig. 1 is markedly similar to that observed for the $^{202}Hg(p, 2pxn)$ reactions.[1] The $(p, 2pn)$ reaction has a threshold energy of 20 MeV and is observed at incident energies of 25 MeV and above. It increases sharply, indicative of the rapidly increasing probability of escape of two charged particles with increasing incident energy. Of particular note is that the plateau corresponds to a cross section of 35–40 mb, a factor of 6–8 greater than that for the corresponding $^{202}Hg(p, 2pn)$ reaction. This is due to two factors affecting proton emission from nuclides in the ^{90}Zr region. First, the neutron binding energy for these neutron deficient nuclides is about 3–5 MeV greater than the proton binding energy while they are about the same in ^{202}Hg. Second, and more important, the Coulomb barrier for proton emission from ^{90}Zr is approximately 7 MeV, one-half that for ^{202}Hg. The combination of the two means that the separation energy for a proton is about 15 MeV while that for a neutron is about 12, a difference which while favoring neutron emission does so to a much smaller extent than it does in ^{202}Hg.

The remainder of the excitation functions show the characteristic complex behavior discussed fully in Ref. 1. The excitation function for the $(p, 2p2n)$ reaction peaks at about 21 MeV while the thermodynamic threshold energy for emission of two protons and two neutrons is 29 MeV and the sum of the four separation energies (including

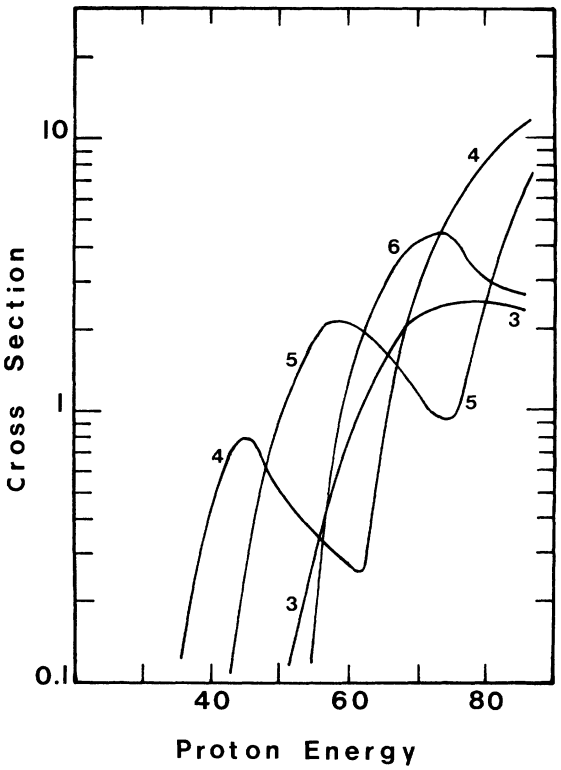

FIG. 3. Excitation functions for $^{90}Zr(p, 4pxn)$ reactions. Each curve is labeled by the value of x. Cross sections are in mb; energy in MeV.

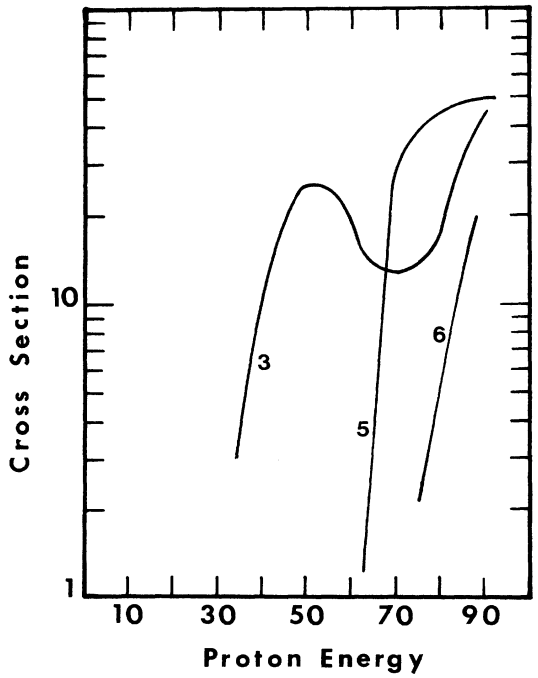

FIG. 2. Excitation functions for $^{90}Zr(p, 3pxn)$ reactions. Each curve is labeled by the value of x. Cross sections are in mb; energy in MeV.

Coulomb barrier) is 45 MeV. Table IV presents a summary of separation energies compared to the incident proton energy of the first peak in the excitation function for each of the reactions under study. As previously discussed,[1] this initial peaking of the excitation function may be related to the emission of α particles wherein the intrinsic binding energy of the α particles lowers the threshold and separation energies by 28–30 MeV. The reason the cross sections do not continue to increase beyond the initial rise is due to the possibility of further emission of nucleons, direct evidence that the mechanisms governing these reactions are not simply those of "clean knockouts", but rather may involve a spectrum of excitation energies.

2. *(p,3pxn) excitation functions*

The three $(p, 3pxn)$ excitation functions measured are shown in Fig. 2. Only in the case of the $(p, 3p3n)$ reaction was there sufficient proton energy available to actually observe the peaking behavior, although it is felt that sufficient evidence exists now to ascribe the initial steeply increasing cross section for the other two reactions to the emission of α particles. For example, the $(p, 3p5n)$ reaction has a cross section of 13.4 mb at 68 MeV, although 66 MeV is the threshold energy for this reaction while the separation energy is about 88 MeV, approximately the incident energy at which the excitation function appears to peak. It is also noteworthy that the cross section for the $(p, 3p5n)$ reaction is a factor of 2 higher at its peak than is the $(p, 2p5n)$. At this point of extreme neutron deficiency the separation energy of protons is actually lower than that of neutrons, even taking account of the Coulomb barrier.

TABLE IV. Summary of separation energies and the incident proton energy at which excitation functions peak for ^{90}Zr reactions (Ref. 27).

Reaction	Separation energy	Peak energy
^{90}Zr$(p, 2p2n)^{87}$Y	45	21
(p, α)	17.1	
^{90}Zr$(p, 2p3n)^{86}$Y	57.1	36
$(p, \alpha n)$	29.3	
^{90}Zr$(p, 2p4n)^{85}$Y	66.7	50
$(p, \alpha 2n)$	38.8	
^{90}Zr$(p, 2p5n)^{84}$Y	78.8	68
$(p, \alpha 3n)$	50.9	
^{90}Zr$(p, 3p3n)^{85}$Sr	67.2	50
$(p, \alpha pn)$	39.3	
^{90}Zr$(p, 3p5n)^{83}$Sr	87.5	83
$(p, \alpha p3n)$	59.6	
^{90}Zr$(p, 3p6n)^{82}$Sr	96.4	?
$(p, \alpha p4n)$	68.5	
^{90}Zr$(p, 4p3n)^{84}$Rb	87.9	75
$(p, \alpha 2pn)$	60	
^{90}Zr$(p, 4p4n)^{83}$Rb	96.1	44
$(p, \alpha 2p2n)$	68.5	
$(p, 2\alpha)$	41.4	
^{90}Zr$(p, 4p5n)^{82}$Rb	107.2	57
$(p, \alpha 2p3n)$	79.3	
$(p, 2\alpha n)$	52.2	
^{90}Zr$(p, 4p6n)^{81}$Rb	116.2	72
$(p, \alpha 2p4n)$	88.3	
$(p, 2\alpha 2n)$	61.1	

3. *(p,4pxn) excitation functions*

The $(p, 4pxn)$ excitation functions are shown in Fig. 3. The $(p, 4p3n)$ reaction is easily shown from energetic considerations to be most likely $(p, \alpha 2pn)$. The $(p, 4p3n)$ reaction has a separation energy of approximately 88 MeV, about 30 MeV higher than where the experimental cross section was measurable and about 10 MeV higher than the apparent peak.

Of greater importance, however, is that no cross section was measurable below 56 MeV incident proton energy. On the other hand, the $(p, 4p4n)$ reaction *peaked* at a bombarding energy of about 44 MeV, although the threshold for a reaction involving an α particle is 30 MeV and the sum of the separation energies of an α particle, two protons, and two neutrons is about 68 MeV. It must be assumed, therefore, that the reaction involves the emission of two α particles, a reaction with a separation energy of about 41 MeV, and a thermodynamic threshold of only 7 MeV.

The second steep increase is very similar to that of the $(p, 4p3n)$ reaction and is positioned 8–10 MeV higher in bombarding energy. It is still well below the separation energy of 92 MeV for the emission of $4p$ and $4n$ and it must therefore be concluded that this second rise represents the contribution of the $(p, \alpha 2p2n)$ reaction. It is unfortunate that irradiations with sufficient beam intensity at energies higher than 86 MeV are impossible at McGill because there is confidence that a second maximum and a third increase might well have been observed.

The $(p, 4p5n)$ reaction shows identical behavior and the evidence is strong that the $(p, 4p6n)$ reaction as observed is most likely a $(p, 2\alpha 2n)$ component.

4. *Decomposition of excitation functions*

The excitation functions for the $(p, 2pxn)$ and $(p, 3pxn)$ reactions may be decomposed into a com-

ponent representing α particle emission and one representing the emission of individual nucleons. The procedure is discussed in detail in Ref. 1. There are four fundamental assumptions in the process: (a) It is assumed that all of the cross section measurements below the separation energy for individual nucleons result from processes involving α particle emission. (b) It is assumed that the shape of the $[p, 2p(x+2)n]$ component of the $(p, \alpha xn)$ excitation function is similar to that of the $(p, 2pn)$ excitation function. In this work, we extend this assumption to the $[p, 3p(x+2)n]$ component of the $(p, \alpha pxn)$ excitation function. (c) It is assumed that the α emission component decreases at high incident energies in a manner similar to that observed for the ^{90}Zr(p, xn) reactions measured in this laboratory.[20] (d) It is assumed that "missing" excitation functions may be interpolated from the neighboring experimentally determined excitation functions.

The $(p, 4pxn)$ reactions were handled in an identical fashion assuming for those cases with $x>3$ that a two α particle emission component was also present.

With these assumptions, the experimental data may be decomposed to give the two components of each excitation function. With the complete set of α particle emission cross sections, it is then possible to sum the individual cross sections to yield the total α emission cross section for incident protons of 10–86 MeV. This production cross section is given in column 2 of Table V.

This total production cross section for α particle emission may be further divided into a "direct interaction" component and an isotropic "evaporation" component with the help of angular distribution studies. The fraction of α particles emitted with an isotropic distribution has been estimated to be negligible for 15 MeV protons incident upon ^{93}Nb and ^{89}Y,[24] but 70% and 66% for 23 MeV protons on Cu and Rh, respectively,[25] and 84% and 87% for 56 MeV protons in Cu and Ru, respectively.[26] Interpolating between these points, the cross sections for direct interaction (forward peaked) and "evaporation" (isotropic) α particles may be calculated. Results of these calculations are given in columns 3 and 4 of Table V. The last three columns of this table repeat the data of Ref. 1 for purposes of comparison. These results are consistent with the reported cross sections for direct emission from other targets in on-line experiments. With 56 MeV protons, the direct cross sections of Muto *et al.* are 25 mb for Cu, 21 for Rh, and 22 for Ag.[9]

There are two principal conclusions to be drawn from this comparison: First, the direct interaction components of the total production are quite similar for the two target nuclei over the entire incident energy range; and second, the increase of a factor of about 4 in the cross sections for Zr relative to Hg is accounted for by an increase in the probability of "evaporation" of α particles.

It is also possible that triton pickup may enhance the nonisotropic component of the reaction at low incident energies. At energies of 15 MeV (on ^{93}Nb and ^{89}Y)[24] and 20–22 MeV (on ^{90}Zr and ^{89}Y),[28] Vergennes *et al.* and Fulmer and Cohen, respectively, have reported angular distributions which are consistent with triton pickup, but not necessarily inconsistent with direct knockout of an α particle. Either of the two suggestions above, and probably both, may be responsible for the steep increase in the direct interaction cross section at low bombarding energies.

On the basis of this work, it is impossible to draw conclusions about the probability of α clustering with changing mass of the target nucleus. The experimental evidence of equal cross sections for the direct interaction component may be interpreted in three mutually contradictory fashions: (a) since the cross sections are the same, the probability of existence of α clusters is the same; (b) since the cross sections are the same but Zr is a smaller nucleus with a smaller surface where clustering is expected to be most important, the degree of clustering in Zr must be

TABLE V. Cross sections (in mb) for α particle emission.

	Target: ^{90}Zr			Target: ^{202}Hg (Ref. 1)		
Proton energy	Total α production	Direct interactions	Evaporation	Total α production	Direct interactions	Evaporation
20	15	9	6	4	4	0
30	55	16	39	9	7.5	1.5
40	80	19	61	15	11	4
50	100	20	80	24	16	8
60	130	22	108	33	21	11
70	150	25	125	41	26	15
80	170	29	141	49	29	20

greater; and (c) since the cross sections are the same but Hg has a larger barrier restraining the emission of α particles, the degree of clustering in Hg must be greater. Each of these three possible conclusions has been previously supported by the experimental data of Detraz *et al.*[29] [conclusion (a)], Bertrand and Peelle[14] (b), and Bachelier *et al.*[30] (c). Finally, Milazzo-Colli and Braga-Marcazzan report no consistent variation.[15,31]

B. Pre-equilibrium model implications

The experimental data presented in this work may be used to derive information of value to calculations of prequilibrium emission of charged particles, particularly of α particles. There are two basic types of information: the excitation energy spectra of nuclei which emit α particles, and the absolute probability of emission of various combinations of protons and α particles.

1. Excitation energy spectra for α emission events

The set of excitation functions for α emission events derived by decomposing the experimental excitation functions as previously described yield for any given incident energy the relative probabilities that, in addition to the α particle, one or more other particles are emitted. In Ref. 1 we made the assumption that on the average 10 MeV is required for emission of a neutron and on that basis derived the excitation energy spectra shown in Fig. 5 of that work. A further assumption is made in this work that 15 MeV is on the average required for evaporation of a proton.

In addition to the experimental data of Figs. 1–3, interpolations were made of the excitation functions of the $(p, 3p2n)$ and $(p, 3p4n)$ reactions based on the $(p, 3p3n)$ and $[p, 3p(5\,\&\,6)n]$ results, and of the $[p, 2p(6\,\&\,7)n]$ based on the $[p, 2p(2-5)n]$ results. The spectra of excitation energies associated only with those events in which an α particle is emitted for various incident proton energies is shown in Fig. 4.

The spectra are similar to those of the previously reported ^{202}Hg results in that they show a peak at a substantial fraction of the incident proton energy. Except at low bombarding energies, there is no evidence for a large fraction of very low (0–10 MeV) deposition events, such as might be expected for clean knockout processes. This is consistent with the results of the decomposition of the excitation functions discussed above in that while at low incident energies most events leave little or no excitation energy, such is not the case at higher energies. A more subtle effect is also noticeable. At incident energies of 50–80 MeV where on line studies suggest 15–20% of events are "direct interactions", i.e., forward peaked, the spectra show only 2–5% of events, leaving insufficient excitation energy to emit another particle or particles. It is felt to be likely, therefore, that previous ideas of clean knockout of preformed α clusters may well be too simplistic and that even the forward peaking of angular distributions may be insufficient to define clearly the mechanism of reaction. Apparently many events producing forward peaked α particles also leave sufficient excitation energy to evaporate further nucleons.

It is also noteworthy that the peaks in the spectra fall at energies more removed from the incident proton energy than the consistently observed value of 20 MeV in the Hg experiments. In this work, the peak excitation energy differs from the incident energy by ~ 25 MeV at 50 MeV of proton energy, increasing to ~ 40 MeV at 80 MeV of incident energy. The consistency in the Hg results strongly suggested a compound nucleus component; these results imply a decreased probability of compound nucleus production, probably related to an overall reduction in the probability of proton

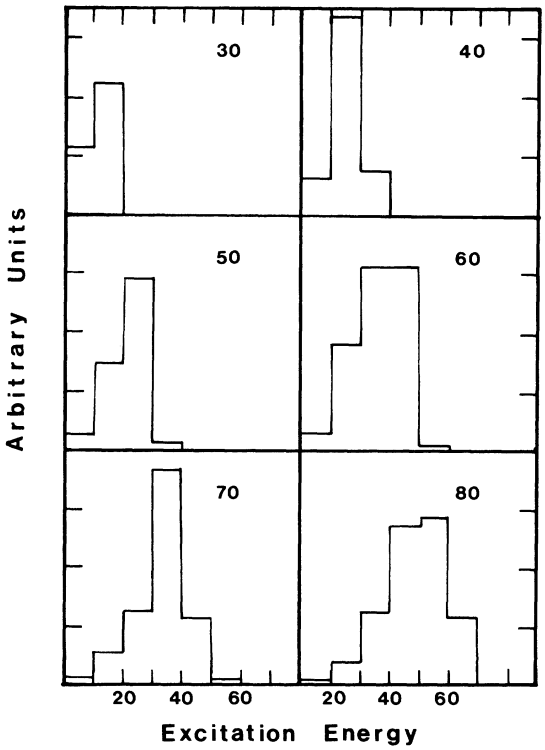

FIG. 4. Excitation energy spectra for those events in which an α particle is emitted. The incident proton energies in MeV are indicated by the number in the upper right of each spectrum.

capture by the smaller, highly neutron deficient ^{90}Zr target nucleus.

2. *Emission of charged particles*

Using as a basis all of the experimental data discussed above, it is possible to estimate the probability of emission of various combinations of charged particles from the interaction of 10–86-MeV protons with ^{90}Zr. Additional missing excitation functions are the $(p,2p)$ evaluated from considering the ^{90}Zr$(p,2pn)$ and the ^{202}Hg$(p,2p)$ and $(p,2pn)$ results of Ref. 1, and the $(p,3p)$ and $(p,3pn)$ excitation functions thought to be very small. Decomposition of all of the experimental data into their α particle and individual nucleon components allows calculation of the probability of emission of, for example, a single α particle and any number of neutrons at any given incident energy. Similarly, one may calculate the probability of emission of two protons, or a proton and an α particle, or any combination of protons and α particles up to a total of four charges. Both the lack of data and the questionable nature of the mechanism in which only a single charge (which may or may not be the incident proton) is emitted prevent an estimate of the probability of emission of a single proton.

Because the absolute values of the total cross section for emission of a given combination of charged particles is valid, strictly speaking, only for the ^{90}Zr target nucleus, or possibly a few other neutron deficient, medium weight target nuclei, the results of this analysis are presented as the fractional probability of emission of the given combination of charged particles. These were prepared using the total reaction cross sections calculated by the optical model code known as ABACUS-2 of Auerbach[32] using the parameters of Becchetti and Greenlees.[33] It is felt that the *relative* probabilities of emitting the various combinations of charged particles are likely to be more universally valid than the specific cross sections for a ^{90}Zr target. The results of this analysis are shown in Fig. 5.

This represents the first attempt at such calculations based almost solely on experimental data. Specifically, each line on the graph represents the fraction of the total reaction cross section as a function of incident proton energy in which a certain combination of charged particles is emitted. Thus, the lines marked $(\alpha+p)$ and $(3p)$ represent the excitation functions for the sum of all $(p,\alpha pxn)$ and $(p,3pxn)$ excitation functions respectively. The sum of the two (not shown) gives the total probability of production of nuclides having three fewer charges than the compound nucleus.

It is felt that these results provide fertile ground for the testing of prequilibrium model calculations of nuclear reactions.

It is interesting that the probability of emission for a single α particle rises sharply, goes through a broad maximum and then decreases, reminiscent of simple (p,xn) reactions and for the same reason apparently. The sharp increase is representative of a rapidly increasing probability of emission of the doubly charged α particles at energies above its separation energy. The broad peak suggests that over a range of energies further emission of charged particles is unlikely, the nucleus preferring to dissipate energy by emission of neutrons. However, at some higher energy, emission of a proton starts to occur [rising $(\alpha+p)$ line], successfully competing with neutron emission in the deexcitation of those nuclides from which an α particle has already been emitted. The result is a decrease in the fractional probability of emitting only an α particle. Similar understanding of the other probabilities underlines the similarities to other, simpler, (p,xn) reactions.

SUMMARY AND CONCLUSIONS

In Ref. 1, the authors suggest that substantial "evaporation" of α particles from highly excited

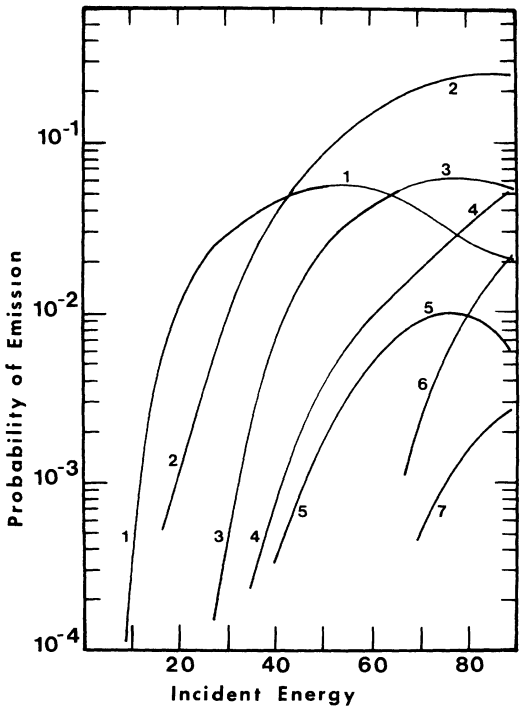

FIG. 5. Fractional probability of emission of charged particles from ^{90}Zr reactions. Line 1, 1α; line 2, $2p$; line 3, $\alpha+p$; line 4, $3p$; line 5, 2α; line 6, $\alpha+2p$; line 7, $4p$ (estimated).

nuclei is necessary to explain the yields of ^{202}Hg(p,2pxn) reaction products. In this work, the yield of α particles is nearly four times as great, while angular distribution and particle spectra studies[24-26] suggest that nearly the entire increase is isotropic "evaporation" α particles. Results show this increase to vary from a factor of about 20 at low proton energies to a factor of about 7 at 85 MeV. This is completely consistent with the enhancement of α particle yields by emission from a rapidly rotating, highly excited nucleus. The general increase in the yield of α particles (and protons) arises from the lower separation energies for charged particles discussed earlier. The Coulomb barrier is lower in the lighter ^{90}Zr target; the binding energies for charged particle emission are smaller relative to that of the neutron in the more neutron-deficient ^{90}Zr target. It is also to be expected that these differences would be more pronounced at lower energies where the probability of charged particle emission is critically dependent upon barrier heights than at higher energies.

In summary, having measured a large number of excitation functions for ^{90}Zr[p,(2-4)pxn] reactions, we have separated the component derived from emission of α particles from that involving emission of individual nucleons, derived the excitation energy spectra for those events yielding an α particle, and calculated the probability of emission of various combinations of charged particles, all at incident proton energies varying from 10-85 MeV. It is hoped that the results of these experiments will stimulate further testing and improvement of preequilibrium models of nuclear reactions, particularly in the field of α particle emission.

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[1]M. V. Kantelo and J. J. Hogan, Phys. Rev. C 13, 1095 (1976).
[2]R. A. Sharp, R. M. Diamond, and G. Wilkinson, Phys. Rev. 101, 1493 (1956).
[3]J. W. Meadows and R. B. Holt, Phys. Rev. 83, 47 (1951).
[4]S. Tanaka, M. Furukawa and M. Chiba, J. Inorg. Nucl. Chem. 34, 2419 (1972).
[5]N. T. Porile, S. Tanaka, H. Amano, M. Furukawa, S, Iwata, and M. Yagi, Nucl. Phys. 43, 500 (1963).
[6]S. Hontzeas and L. Yaffe, Can. J. Chem. 41, 2194 (1963).
[7]C. L. Rao and L. Yaffe, Can. J. Chem. 41, 2156 (1963).
[8]R. Bimbot and M. Lefort, J. Phys. (Paris) 27, 385 (1966).
[9]J. Muto, H. Itoh, K. Okano, N. Shiomi, K. Fukuda, Y. Omori, and M. Kihara, Nucl. Phys. 47, 19 (1963).
[10]H. Dubost, M. Lefort, J. Peter, and X. Terrago, Phys. Rev. 136, B1618 (1964).
[11]M. Lefort, J. P. Cohen, H. Dubost, and X. Tarrago, Phys. Rev. 139, B1500 (1965).
[12]H. Dubost, B. Gatty, M. Lefort, J. Peter, and X. Tarrago, J. Phys. (Paris) 28, 257 (1967).
[13]E. Gadioli, I. Iori, N. Molho, and L. Zetta, Phys. Rev. C 4, 1412 (1971).
[14]F. E. Bertrand and R. W. Peelle, Phys. Rev. C 8, 1045 (1973).
[15]L. Milazzo-Colli, G. M. Braga-Marcazzan, and M. Milazzo, Nucl. Phys. A218, 274 (1974).
[16]M. Blann, Annu. Rev. Nucl. Sci. 25, 123 (1975).
[17]J. S. Foster, J. W. Hilborn, and L. Yaffe, Can. J. Phys. 36, 555 (1958).
[18]R. Colle, R. Kishore, and J. B. Cumming, Phys. Rev. C 9, 1819 (1974).
[19]D. A. Newton, S. Sarkar, L. Yaffe, and R. B. Moore, J. Inorg. Nucl. Chem. 35, 361 (1973).
[20]M. V. Kantelo, Ph.D. thesis, McGill University, 1975 (unpublished).
[21]R. Denig, N. Trautmann, and G. Herrmann, J. Radioanal. Chem. 5, 223 (1970).
[22]K. A. Kraus and G. E. Moore, J. Amer. Chem. Soc. 75, 1460 (1953).
[23]W. R. Busing and H. A. Levy, Oak Ridge National Laboratory Report No. ORNL-TM-271, 1962 (unpublished).
[24]M. Vergennes, G. Rotbard, J. Kalifa, and G. Berrier-Ronsin, Phys. Rev. C 10, 1156 (1974).
[25]C. B. Fulmer and B. L. Cohen, Phys. Rev. 112, 1672 (1958).
[26]J. Muto, H. Itoh, K. Okano, N. Shiomi, K. Fukuda, Y. Omori, and M. Kihara, Nucl. Phys. 47, 19 (1963).
[27]W. D. Myers and W. J. Swiatecki, Report No. UCRL-11980, 1965 (unpublished).
[28]C. B. Fulmer and J. B. Ball, Phys. Rev. 140, B330 (1965).
[29]C. Detraz, D. D. Zafiratos, C. E. Moss, and C. S. Zaidins, Nucl. Phys. A177, 258 (1971).
[30]D. Bachelier, M. Bernas, O. M. Bilaniuk, J. L. Boyard, J. C. Jourdain, and P. Radvanyi, Phys. Rev. C 7, 165 (1973).
[31]L. Milazzo-Colli and G. M. Braga-Marcazzan, Nucl. Phys. A210, 297 (1973).
[32]E. H. Auerbach, Brookhaven National Laboratory Report No. BNL-6562, 1962 (unpublished).
[33]F. D. Becchetti, Jr., and G. W. Greenlees, Phys. Rev. 182, 1190 (1969).
[34]I. Borchert, Z. Phys. 223, 473 (1969).

[35]H. Verheul, Nucl. Data B2, 64 (1967).
[36]J. B. Ball, M. W. Johns, and K. Way, Nucl. Data A8, 407 (1970).
[37]M. W. Johns, J. Y. Park, S. M. Shafroth, D. M. Van Patter, and K. Way, Nucl. Data A8, 373 (1970).
[38]H. Houtermans, International Atomic Energy Agency (private communication).
[39]H. Verheul, Nucl. Data B5, 457 (1971).
[40]R. L. Auble, Nucl. Data B5, 151 (1971).
[41]M. L. Simpson, J. E. Kitching, and S. K. Mark, Nucl. Phys. A186, 171 (1972).
[42]D. J. Horen, Nucl. Data B5, 131 (1971).
[43]N. G. Zaitseva, B. Kratsik, M. G. Loshchilov, G. Muziol, C. T. Min', and H. S. Strusny, Izv. Akad. Nauk SSSR Ser. Fiz. 33, 1283 (1969) [Bull. Acad. Sci. USSR Phys. Ser. 33, 1186 (1969)].
[44]C. M. Lederer, J. M. Hollander, and I. Perlman, *Table of Isotopes* (Wiley, New York, 1967).
[45]R. C. Etherton, L. M. Beyer, W. H. Kelly, and D. J. Horen, Phys. Rev. 168, 1249 (1968).
[46]J. Vrzal, B. S. Dzhelepov, A. G. Dmitriev, N. N. Zhukovskii, J. Liptak, L. N. Moskvin, J. Urbanets, and L. G. Tsaritsyna, Bull. Acad. Sci. USSR Phys. Ser. 31, 1701 (1967).
[47]R. L. Auble, Nucl. Data B5, 109 (1971).
[48]A. H. Wapstra and N. B. Gove, Nucl. Data A9, 265 (1970).
[49]H. Verheul, Nucl. Data B2, 31 (1967).