

Radiochemical study of alpha particle emission in reactions of ^{202}Hg with 10–86 MeV protons

Marti V. Kantelo* and James J. Hogan

Department of Chemistry, McGill University, Montreal, Quebec, Canada

(Received 2 September 1975)

Excitation functions have been measured for $^{202}\text{Hg}(p, 2pxn)$, $x = 0-9$, reactions induced by protons of energy 10–86 MeV. Reactions having $x > 1$ exhibit two peaks in their excitation functions, the first below the energy calculated for emission of the individual nucleons. This peak is attributed to emission of an α particle. A procedure is proposed for decomposing the excitation functions into two components, one arising from α particle emission, the second from emission of individual nucleons. From these, the total yield of α particles as a function of incident energy is determined, and the excitation energy spectra associated with α emission events is deduced. The α particle yields are attributed to more than one mechanism; speculations are advanced on a substantial pre-equilibrium evaporation component to the total α particle production.

[NUCLEAR REACTIONS $^{202}\text{Hg}(p, 2pxn)$, $x = 0-9$, $E = 10-86$ MeV; measured $\sigma(E)$; deduced α particle yields. Enriched target.]

INTRODUCTION

For some time, the existence of clusters of nucleons in the nuclear surface has been of considerable interest. Recent attempts at calculating the probability of α particle substructures have heightened this interest. Wilkinson¹ pointed out 15 years ago that in the surface region of the nucleus where the nucleon density is low, the average spacing between nucleons becomes large compared to the range of the nuclear forces. Thus a clustering process would increase the total binding energy of the nucleus, and the tightly bound α particle would do so most efficiently. The picture which has generally emerged is one of dynamic formation and dissolution of such clusters rather than of the presence of permanent substructures.

Ample experimental evidence exists for such a clustering. In 1958, Hodgson² attributed the existence of high energy α particle tracks in nuclear emulsion studies (45 MeV protons incident upon AgBr) to pre-formed α particle clusters. The many on-line studies in which charged particles are detected have frequently noted the presence of substantial yields of α particles. In particular Igo³, studying the interaction of 915 MeV α particles with various target nuclei measured coincidences of two α particles emitted at the angle required by α - α scattering kinematics. Many energy spectra and angular distributions are available in the literature for " (n, α) " (Refs. 4–6) and " (p, α) " (Refs. 7–13) reactions, although such studies generally measure total α particle emission rather than specifically the (n, α) or (p, α) reactions. Particularly interesting are re-

cent attempts at calculations comparing experimental and theoretical yields and spectra of α particles using various formulations of the pre-equilibrium model. These are discussed more extensively in Blann's recent review article.¹⁴

There seems little doubt that at least two mechanisms, probably difficult to clearly differentiate, are responsible for the emission of α particles in nuclear reactions. The first, characterized by strong forward peaking of high energy α particles is usually attributed to direct, or knock on, interactions. The second, characterized by lower kinetic energies and a more nearly isotropic angular distribution is generally called an evaporation component. However, it is easy to show that the yield of such α particles is too high to be consistent with a conventional equilibrium evaporation process. Such calculations provided some of the impetus for the development of calculations referred to above¹⁴ of evaporationlike processes from nonequilibrium nuclei.

The majority of the research discussed above has concentrated upon some combination of identification, energy spectra, and angular distributions of the emitted α particle itself without reference to the identity of the residual reaction product, and in particular to the possibility or probability of additional emission of nucleons. If one is to understand the emission of α particles, however, knowledge of the residual excitation energy spectrum and the dependency of both this spectrum and the emission rate of α particles as a function of incident bombarding energy is a necessary prerequisite.

To obtain such information, cross sections for the production of many nuclides formed in reac-

tions in which α particles are emitted are needed. Ideally, excitation functions over a range of incident energies encompassing both the predominately compound nucleus region and higher energies where direct interactions predominate must be measured. To determine the residual excitation energy spectra, a related series of $(p, \alpha xn)$ products formed from a given target nucleus would be necessary; the $(p, 2p)$ and $(p, 2pn)$ reactions might then serve as a monitor of multiproton emission without the complicating effects of α particle clustering.

There are two main problems to measuring such a series of excitation functions. The first is the low cross sections for $(p, 2pxn)$ reactions relative to the barn cross sections for the (p, xn) and (p, pxn) reactions. Radiochemical procedures are required for sufficient decontamination to allow accurate measurement of α particle cross sections. Second, and more subtle, is the distinguishing of a $[p, \alpha xn]$ reaction cross section from that of the $[p, 2p(x+2)n]$ which leads to the same product nucleus.

The most extensive previous study of $(p, \alpha xn)$ excitation functions was that of Gauvin¹⁵ where x ranged from 1–5 and the incident energy ranged up to 115 MeV. Unfortunately, the target nucleus ²³²Th is highly fissionable, greatly complicating interpretation of the results. Other studies in the literature have measured only isolated (accessible) product nuclei or used only a single, or limited range of, bombarding energies.^{16–22} All of these data indicate that on-line experiments purporting to measure the (p, α) reaction are in fact usually measuring many different nuclear reactions, emphasizing the need for a systematic study of excitation functions for reactions involving α particle emission.

It was the goal of this work to measure the excitation functions for the ²⁰²Hg($p, 2pxn$) reactions, x varying from 0–9, between 10–86 MeV incident proton energy, and to show how the complex excitation functions may be decomposed into one component associated with emission of α particles and a second arising from emission of two protons and a number of neutrons.

PROCEDURE AND RESULTS

The target consisted of a uniform mixture of enriched ²⁰²Hg as HgO and spectroscopically pure CuO. The former, obtained from Oak Ridge National Laboratory, contained 91.14% ²⁰²Hg, 5.9% ²⁰¹Hg, and less than 1% of each of the remaining four stable isotopes. Proton activation of the HgO showed less than 0.01% chemical impurity of interfering elements.

The target material was contained in a thin walled aluminum tube and irradiations were performed as in many previous experiments at the McGill Synchrocyclotron.²³ Most irradiations were for 60 sec at a nominal beam intensity of approximately 0.6 μ A. The beam was monitored by the absolute cross sections for the ⁶³Cu(p, n)⁶³Zn (Ref. 24) and ⁶⁵Cu(p, pn)⁶⁴Cu reactions.²⁵

After irradiation the target assembly was dissolved in a solution containing 7 ml aqua regia, 1 ml H₂O, 1 ml H₂PtCl₆ (10.7 mg Pt^{IV}), 100.0 μ l ¹⁹⁵Au tracer, 100.0 μ l ²⁰⁴Tl tracer, and 0.1 ml of a holdback carrier solution which contained Zn, Ni, Co, and Fe. Heating the solution for two minutes ensured complete dissolution and oxidation of all elements present.

After cooling, Au³⁺ and Tl³⁺ were extracted into 15 ml ethyl acetate. The Hg²⁺ extracts slightly; Cu, Pt, and the bulk of Hg remain in the aqueous phase. The organic phase was washed three times with 2 M HCl, effectively removing mercury impurities. The gold and thallium were then back extracted with 1 M NH₄OH. The aqueous phase was neutralized and acidified to 1 M HCl and 250 mg of powdered Cu was added. The Cu powder reduces Au³⁺ to elemental gold and adsorbs it while reducing Tl³⁺ to Tl⁺. The suspension was vigorously stirred for 15 sec, filtered, washed with 1 M HCl, suspended in H₂O, refiltered, washed with water and ethyl alcohol, dried and mounted for counting. This chemical procedure is based on the work of Weiss and Reichert²⁶ with some modifications.

Tracer ¹⁹⁵Au serves as a monitor of the chemical yield. The photon detection rate for 99 keV ¹⁹⁵Au tracer was corrected for attenuation in the sample itself by comparison of the observed activity and weight of source to a reference standard. The correction never exceeded 2%. It is estimated that ¹⁹⁵Au produced by nuclear reactions in the target material did not exceed 0.1% of the added tracer. The tracer measurements were made at least 20 days after irradiation to avoid interference from the 97 keV photon of 2.3 day ¹⁹⁸Au^m. The mean chemical yield was 50%.

The aqueous phase from the ethyl acetate extraction containing Cu, Pt, and Hg had 1 ml of 10% CsCl added to precipitate Cs₂PtCl₆. The solution was cooled in ice and filtered. The filtrate was reserved for Cu chemistry. The Cs₂PtCl₆ was washed with ice water and ethyl alcohol, dried, weighed and mounted for counting. The mean chemical yield was 48%.

Copper was separated by the usual procedure of this laboratory,²⁷ eventually being counted as CuSCN. The mean chemical yield was 63%.

Samples were counted on a 42 cm³ Ge(Li) detec-

TABLE I. Nuclear properties of nuclides studied in this work.

Nuclide	Photon energy (keV)	Photon abundance ^a	Half-life	References
²⁰¹ Au	543	2.0	26.4 min	35
²⁰⁰ Au ^m	368	80	18.7 h	36
²⁰⁰ Au ^ε	368	19	48.4 min	37
¹⁹⁹ Au	158	39	3.14 day	38
¹⁹⁸ Au ^m	215	79	2.30 day	39, 40
¹⁹⁸ Au ^ε	412	95.5	2.70 day	41
¹⁹⁶ Au ^m	148	45	9.7 h	42
¹⁹⁶ Au ^ε	356	88	6.18 day	42
¹⁹⁵ Au	98.9	...	183 day	43
¹⁹⁴ Au	328	63	39.5 h	44
¹⁹³ Au	256	6.7	17.65 h	45
¹⁹² Au	316	...	5.03 h	46
⁶³ Zn	670	8.47	38.8 min	47, 48
⁶⁴ Cu	511	38.6	12.82 h	49

^a Photons per 100 decays.

tor 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 coefficients. Decay curves were analyzed by the weighted least squares procedure known as ORGLS.²⁸

The disintegration rate at the end of irradiation was calculated from the observed count rate by

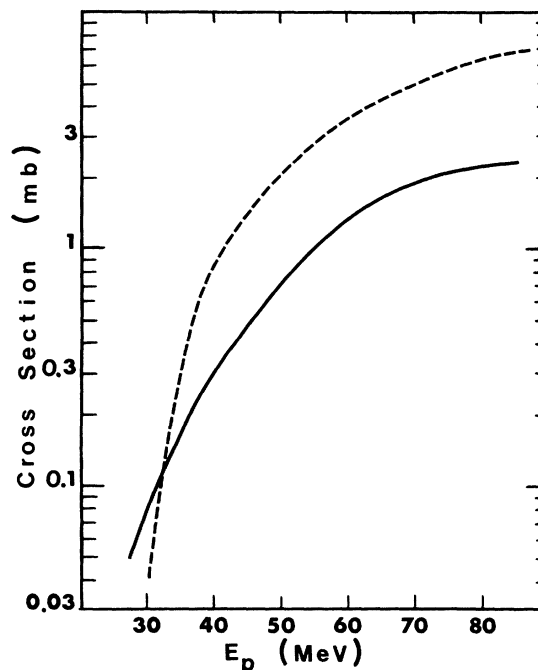


FIG. 1. Excitation functions of $^{202}\text{Hg}(p, 2p)^{201}\text{Au}$ and $^{202}\text{Hg}(p, 2pn)^{200}\text{Au}$ reactions: —, ^{201}Au ; ---, ^{200}Au .

correcting for decay, counter efficiency, absorption, photon abundances, and chemical yield. Absolute cross sections were determined by comparison of the calculated disintegration rate for the product nuclei to that of ^{64}Cu , and thence to the monitor cross sections.^{24,25} Uncertainties in the reported cross sections reflect the errors associated with decay curve analysis, detector

TABLE II. Experimental cross sections for $^{202}\text{Hg}(p, 2pxn)$ reactions in mb.

Proton energy (MeV)	²⁰¹ Au (p, 2p)	²⁰⁰ Au (p, 2pn)	¹⁹⁹ Au (p, 2p2n)	¹⁹⁸ Au (p, 2p3n)	¹⁹⁶ Au (p, 2p5n)	¹⁹⁴ Au (p, 2p7n)	¹⁹³ Au (p, 2p8n)	¹⁹² Au ^a (p, 2p9n)
12			0.064 ± 0.011					
15			0.35 ± 0.07	0.015 ± 0.05				
19			3.5 ± 0.5	0.29 ± 0.05				
24			6.0 ± 0.8	0.99 ± 0.13				
30		0.045 ± 0.009	4.7 ± 0.5	4.2 ± 0.4				
34	0.157 ± 0.028	0.194 ± 0.027	3.8 ± 0.5	5.7 ± 0.9	0.23 ± 0.02			
38		0.56 ± 0.09	2.2 ± 0.4	4.6 ± 1.5	0.87 ± 0.11			
44		1.51 ± 0.21	2.3 ± 0.3	4.0 ± 1.3	5.9 ± 0.7			
50	0.68 ± 0.17	1.85 ± 0.24	2.5 ± 0.3	3.8 ± 1.3	8.2 ± 0.8			
56	1.22 ± 0.26	2.60 ± 0.26	3.2 ± 0.4	3.3 ± 0.8	9.7 ± 1.0	0.78 ± 0.10		
62	1.20 ± 0.25	4.0 ± 0.5	5.2 ± 0.7	4.1 ± 0.8	9.9 ± 1.0	3.6 ± 0.5		
68	1.74 ± 0.43	4.6 ± 0.6	6.2 ± 0.9	5.0 ± 1.2	7.8 ± 0.8	10.0 ± 1.3	1.53 ± 0.31	
74	2.00 ± 0.48	5.6 ± 0.7	8.1 ± 1.2	6.8 ± 0.9	6.9 ± 0.7	15.2 ± 1.9	4.8 ± 0.7	0.54 ± 0.07
80	1.85 ± 0.45	6.4 ± 1.1	9.8 ± 1.7	8.8 ± 1.7	6.7 ± 0.9	15.1 ± 2.6	12.0 ± 2.3	1.92 ± 0.34
86	2.3 ± 1.0	6.6 ± 0.8	11.4 ± 1.5	10.4 ± 2.3	7.1 ± 0.7	12.6 ± 1.6	14.2 ± 2.0	5.5 ± 0.6

^a Assuming 316 keV photon to be 100% abundant.

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 Table II 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 over-all cross section or excitation function of a given product nuclide, regardless of whether or not α particles are involved. A reaction of form $(p, \alpha xn)$ 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.

DISCUSSION

A. $(p, 2p)$ and $(p, 2pn)$ excitation functions

The threshold energy for these reactions is 8 and 15 MeV, respectively, although as shown in Fig. 1, there was no measurable cross section below 30–35 MeV. The lack of a peak occurring 5–10 MeV above threshold as is commonly observed in (p, xn) reactions is indicative of the small probability of two proton evaporation from a compound nucleus. Rather, both excitation functions reflect the rapidly increasing probability of a nonequilibrium emission of two protons by a direct interaction process with increasing incident proton energy. The rapidity of the increase derives from two points: first, the increasing probability of direct reaction processes and, second, the increasing probability of escape of collision partners with increasing energy. The Coulomb barrier for proton evaporation from ^{202}Hg is 14 MeV; its separation energy is 22 MeV compared with the neutron binding energy of 8 MeV. Neglecting angular momentum effects, the ratio of the probability for proton evaporation to that for neutron evaporation increases with increasing energy from about 10^{-5} at 30 MeV to 10^{-2} at 80 MeV.²⁹ Thus in the discussion of the decomposition of the complex excitation functions of Figs. 2 and 3, a conventional compound nucleus formation followed by evaporation from a statistically equilibrated nucleus plays no role. This lack of a substantial compound nucleus component as evidenced by the $(p, 2p)$ and $(p, 2pn)$ reactions will play an important role in the determination of α particle emission.

Between 30 and 40 MeV, the two excitation functions cross indicating that the probability of deposition of sufficient excitation energy to evaporate a neutron is increasing sharply as is to be ex-

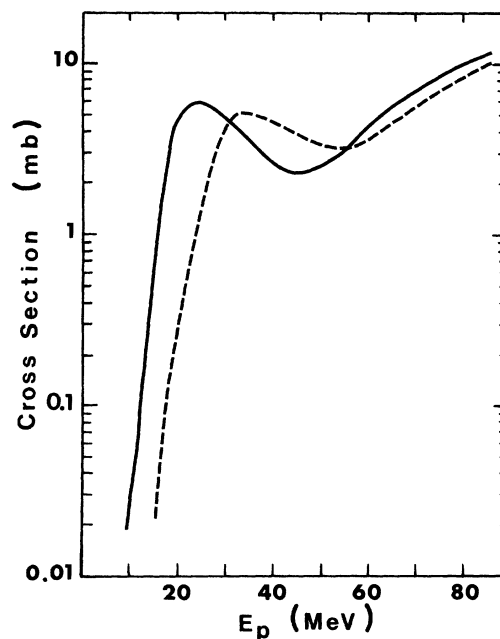


FIG. 2. Excitation functions of $^{202}\text{Hg}(p, 2p2n)^{199}\text{Au}$ and $^{202}\text{Hg}(p, 2p3n)^{198}\text{Au}$ reactions: —, ^{199}Au ; ---, ^{198}Au .

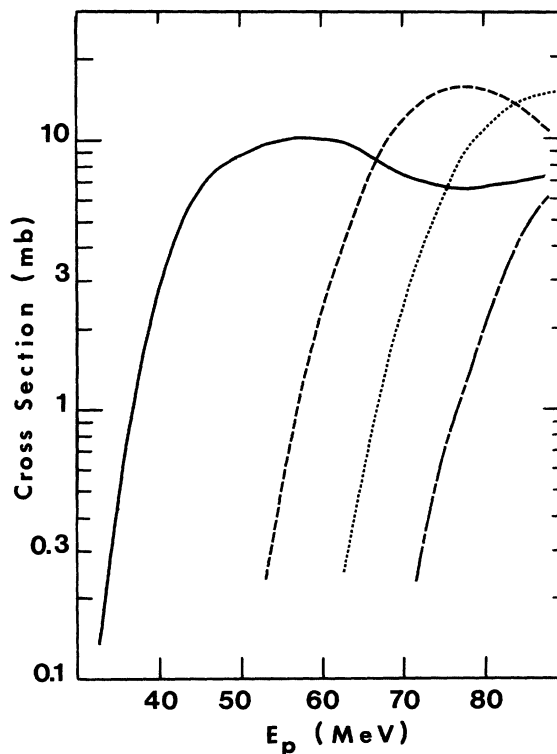


FIG. 3. Excitation functions of $^{202}\text{Hg}(p, 2p5n)^{196}\text{Au}$, $^{202}\text{Hg}(p, 2p7n)^{194}\text{Au}$, $^{202}\text{Hg}(p, 2p8n)^{193}\text{Au}$, and $^{202}\text{Hg}(p, 2p9n)^{192}\text{Au}$ reactions: —, ^{196}Au ; ---, ^{194}Au ; ···, ^{193}Au ; - · - ·, ^{192}Au .

pected. Of greater note to this work is that the excitation functions remain parallel from 40 to 86 MeV. This suggests that the relative probabilities of depositing two different excitation energies is approximately independent of energy. The increase in the excitation functions then may be attributed to the increased probability of two-proton-out direct interactions with little change in the residual excitation energy spectrum. This point has been noted previously³⁰ in (p, xn) reactions and is of some importance in determining the α particle yields.

B. $[p, 2p(2-9)n]$ excitation functions

The following general features apply to all of these excitation functions. With increasing bombarding energy, there is a steep increase in the cross section to a peak *below* the sum of the separation energies for a nuclear reaction emitting the requisite number of protons and neutrons; indeed the peak is below the separation energy for the reactions shown in Fig. 2. For example, while the $(p, 2p2n)$ excitation function clearly peaks at about 24 MeV, the separation energy required is more than 30 MeV, and the thermodynamic threshold is 22 MeV. Some clustering of the outgoing particles must be invoked to account for these reactions; due to the large binding energy of α particles it is reasonable to assume that in this bombarding energy range, the $(p, 2p2n)$ reaction is more properly written (p, α) . A similar argument holds for other $(p, 2pxn)$ reactions having $x > 2$. A summary of thermodynamic threshold energies, separation energies (including the Coulomb barrier), and approximate bombarding energies at which the excitation functions peak is given in Table III.

For the $(p, 2p2n)$ and $(p, 2p3n)$ reactions shown in Fig. 2, the cross section decreases after the peak, proceeds through a minimum, and then increases a second time. Of particular note is that the shape of this second increase parallels very closely the shape of the increase in the $(p, 2pn)$ reaction. The implication is strong that the second increase may be attributed to processes similar to those responsible for the reactions shown in Fig. 1 with the proviso that sufficient excitation energy may be deposited for the emission of the required number of neutrons. There is also further evidence for the insensitivity of the excitation energy spectrum to incident bombarding energy in the parallel rise of the second peak in the two reactions of Fig. 2.

The reactions in which $x \geq 5$ (Fig. 3) show evidence of the same behavior although the second maximum is not attained in the accessible range of bombarding energies. However, each of the excitation functions increase with a slope similar to the initial increase in the $(p, 2p2n)$ reaction but dissimilar to that of the $(p, 2p)$ reaction. Even the excitation function for $(p, 2p7n)$ still exhibits a clearly defined maximum. Furthermore, this peak at about 75 MeV lies 9 MeV below the sum of the separation energies of the nine emitted particles and there is a cross section measurement of ~ 1 mb which is below the thermodynamic threshold of 58.5 MeV.

Taken together, this evidence suggests that although the two peaks are not observed, it is reasonable to assume that the behavior is as measured for the $(p, 2p2n)$ and $(p, 2p3n)$ reactions; the initial rise to a peak observed in those excitation functions shown in Fig. 3 has been attributed to reactions in which an α particle is emitted.

TABLE III. Summary of threshold energies, separation energies, and the incident proton energy at which excitation functions peak for ^{202}Hg reactions (Ref. 50).

Reaction	Thermodynamic threshold energy	Sum of separation energies	Peak in excitation function
$^{202}\text{Hg}(p, 2p2n)^{198}\text{Au}$	21.5	47.3	24
(p, α)	-6.8	17.0	
$^{202}\text{Hg}(p, 2p3n)^{198}\text{Au}$	29.3	55.1	34
$(p, \alpha n)$	1.0	24.8	
$^{202}\text{Hg}(p, 2p5n)^{196}\text{Au}$	43.7	69.2	60
$(p, \alpha 3n)$	15.4	39.2	
$^{202}\text{Hg}(p, 2p7n)^{194}\text{Au}$	58.5	84.3	77
$(p, \alpha 5n)$	30.2	54.0	
$^{202}\text{Hg}(p, 2p8n)^{193}\text{Au}$	65.3	91.1	85
$(p, \alpha 6n)$	37.0	60.8	
$^{202}\text{Hg}(p, 2p9n)^{192}\text{Au}$	73.8	99.6	85
$(p, \alpha 7n)$	45.5	69.5	

C. Decomposition of the excitation functions

The evidence above suggests that the emission of α particles as opposed to multinucleon emissions may be largely evaluated by studying the initial peak in the excitation function for each of the reactions. Therefore it may be possible to separate partial cross sections for $(p, \alpha xn)$ reactions from the curves for the over-all $[p, 2p(x+2)n]$ reactions. The following procedure has been adopted for the decomposition of the experimental excitation functions into α emission and multinucleon emission components.

(a) We assume that all of the cross section below the separation energy necessary for emission of the requisite nucleons corresponds to α emission. Thus we ignore any processes involving clusters other than α particles. This assumption is probably valid in that other clusters lead to separation energies only trivially different from that of the multinucleon process.

(b) We assume that the shape of the $[p, 2p(x+2)n]$ component is similar to that of the $(p, 2p)$ and $(p, 2pn)$ reactions. This is equivalent to assuming the slope of the rising portion toward the second maximum in $[p, 2p(x+2)n]$ reactions is the same as that observed in the $(p, 2p)$ and $(p, 2pn)$ reaction. More fundamentally, this extends the point previously made that the shape of the excitation energy spectrum is relatively independent of incident energy.

(c) Finally, we took the shape of the α component to have a high energy tail similar to the high energy tail of (p, xn) reactions. If the (p, α) component is to be considered at all like a direct knockout reaction as suggested by the particle spectra and angular distribution work of several authors,^{7,10,11} it should be reasonable to assume that it would vary in a manner like the direct knockout of neutrons in high energy (p, n) reactions. Similarly the tail of the $(p, \alpha n)$ component might be expected to vary as the $(p, 2n)$ tail etc. In other experiments³¹ the excitation functions of the $^{202}\text{Hg}(p, xn)$ reactions have been measured in order to determine the slope of the high energy tails.

(d) The two missing excitation functions $(p, 2p4n)$ and $(p, 2p6n)$ leading to stable ^{197}Au and 183 day ^{195}Au were interpolated from the seven other curves. As there are no large variations in the peak heights, the shapes for all the $(p, 2pxn)$ reactions are similar, and both missing excitation functions are bracketed by measured ones, it is felt the interpolation introduces no serious error to the analysis.

With these four assumptions, the excitation functions of Figs. 2 and 3 may be decomposed to give the two components of each curve. An ex-

ample of this analysis is shown in Fig. 4. With the complete set of α reaction products at each bombarding energy, it then becomes possible to evaluate the total α emission cross section for incident protons of 10–86 MeV. This total cross section for α particle emission is given in column 2 of Table IV for various incident proton energies.

[It is to be noted that the excitation function of the $(p, 3p3n)$ reaction peaks at <1 mb.³¹ It is therefore assumed the $(p, 3pxn)$ and $(p, 4pxn)$ reactions contribute little to the total cross section for α emission.]

There is obviously a substantial error in the summing of seven decomposed excitation functions. However, while absolute numbers may be suspect, the variation is quite likely reliable. In addition, most of the total cross section for any given energy comes from only one or two reaction products near the peak of their excitation functions—the single region of the curves which is best defined since only α particle emission is energetically feasible. Calculations using different slopes for the high energy tails and several different interpolations for the two missing excitation functions show a surprising insensitivity to the assumptions made. Absolute numbers varied less than 10%; the over-all trend with increasing energy varied not at all. The uncertainty in the cross sections of Table IV is expected to be about 30%. The results of Table IV are in good agreement with on-line studies of neighboring nuclei

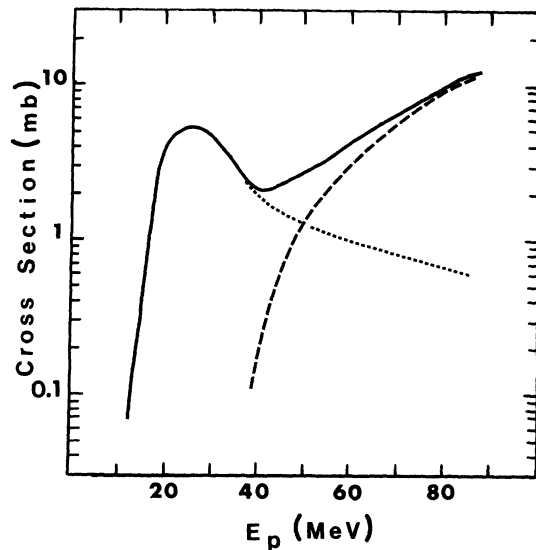


FIG. 4. Decomposition of the $^{202}\text{Hg}(p, 2p2n)^{199}\text{Au}$ excitation function into an α particle component and a four particle emission component.

which lends confidence to the procedure adopted to separate the excitation functions into two parts. For example, Bertrand and Peelle¹² measured 32 mb for 62 MeV protons on ¹⁹⁷Au compared with our estimate of 33, while Muto *et al.*⁷ give a value of 27 mb for 56 MeV protons on ²⁰⁹Bi compared with our 30. Our value is, however, 25% less than Muto's value of 45 mb for a gold target at the same energy. The increase in cross section with increasing bombarding energy is consistent with the increase in the results of Muto *et al.*⁷ and Dubost *et al.*³²

D. Excitation energy spectrum for α emission events

Making the assumption that 10 MeV is required to emit a neutron from an excited nucleus, the set of excitation functions derived for α particle events may be used to construct the spectrum of excitation energies at any given bombarding energy for those events in which an α particle is emitted. Such distributions are shown in Fig. 5 for bombarding energies of 50, 60, 70, and 80 MeV.

It is noteworthy that there are strong peaks in the excitation energy spectra invariably about 20 MeV below the incident proton energy. Including the proton binding energy, this is about 27 MeV below the excitation energy of a compound nucleus. The separation energy of an α particle from ²⁰²Hg (including the Coulomb barrier) is 24 MeV. This result suggests that a substantial fraction of α emission events may well originate in compound nucleus production. This point is discussed further in the next section. If α emission events were exclusively derived from direct knockout interactions, a broad and relatively flat spectrum such as calculated for (p, xn) reactions by intranuclear cascade methods would have been expected.

E. Mechanism of α particle emission

Evidence for a peak in the α emission cross sections coming presumably from a knockout process, coupled with the excitation energy spectra of Fig. 5 which imply a substantial yield of α particles at higher energies associated with compound nucleus events, required the postulation of two mechanisms for these reactions. The first, a fast knock on component, yields strongly forward peaked α particles. The second, associated with compound nucleus production, would be expected to involve an "evaporation" process, slow at least on the time scale of nuclear rotations although not implying conventional evaporation from a nucleus in a state of statistical equilibrium. Three on-line

TABLE IV. Cross sections for production of α particles.

Proton energy	Total alpha production (mb)	Direct interactions (mb)	"Evaporation" (mb)
20	4	4	0
30	9	7.5	1.5
40	15	11	4
50	24	16	8
60	33	21	11
70	41	26	15
80	49	29	20

studies have measured angular distributions of α particles emitted from the interaction of ¹⁹⁷Au with 24–41 MeV protons,¹¹ 56 MeV protons,⁷ and 157 MeV protons.¹⁰ Decomposition of these angular distributions into an isotropic and a forward peaked component yields as the percentage of α particles which are isotropic 10% at 24 MeV, rising to 25% at 41 MeV, 33% at 56 MeV, and 66% at 157 MeV. Interpolating between these points, and using the total α particle cross sections as derived in Sec. III, the cross sections for direct interaction and isotropic α particles may each be calculated. The results are tabulated in columns 3 and 4 of Table IV.

The cross section for α particle production by direct mechanisms increases sharply over this energy region as is to be expected. This is primarily due to an increased probability of escape of a struck α particle. For kinematic reasons, a maximum of 64% of the proton's energy may be transferred to the struck α particle. With a 28 MeV Coulomb barrier to overcome, it is hardly surprising that such a process would be highly energy dependent.

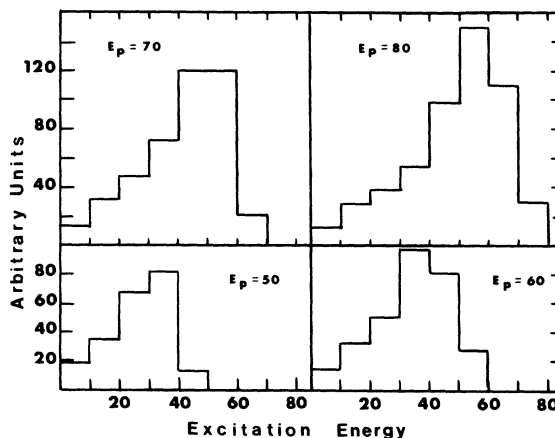


FIG. 5. The distribution of excitation energy in those nuclei associated with α emission events.

Equally interesting is the substantial isotropic component shown by a combination of the on-line studies and the excitation energy spectra of Fig. 5 to be most likely associated with the deexcitation of compound nuclei.

Conventional evaporation from an equilibrium configuration is easily shown to be many orders of magnitude less likely to occur than has been observed in the experiments.²⁹ However, two factors undoubtedly play a large role in enhancing the α production.

Dudey and Sugihara³³ have pointed out that α particle emission probabilities are enhanced from a high angular momentum system because the mass of the α particle allows for efficient reduction of the high angular momentum. Grover and Gilat have developed this point³⁴ based on the principle that more final states exist following α emission from a high angular momentum state than following emission of neutrons or γ rays. However, the Dudey and Sugihara work concerned itself with much lower excitation energies than would be encountered in compound nucleus production in this work, while Grover and Gilat dealt with much higher angular momentum states such as derive from heavy ion bombardments. Nevertheless, there is little question that the results of this work are being strongly affected by enhanced emission of α particles from high angular momentum states of the compound nuclei.

The second factor, which is not entirely separable from the first, is that isotropic emission requires only that lifetimes be longer than rotational periods and not necessarily the attainment of the statistical equilibrium configuration of conventional evaporation theory. An evaporative process from an incompletely equilibrated nucleus

may well produce the yield of isotropic α particles these experiments demand.

The state of the art of pre-equilibrium decay calculations is not yet ready to handle such a problem in that for the most part the yields of α particles are used as a normalizing factor to determine the parameter related to the preformation of clusters. Even calculations of particle spectra and angular distributions in general ignore the question of yields. The Blann review article¹⁴ concludes a brief section on cluster emission with the words "The work which has been done with respect to complex particle emission must be regarded as bold explorations in an extremely difficult and evasive area."

Both of these factors combine then to allow a substantial production of α particles by an "evaporation" type process from a pre-equilibrium nucleus having high angular momentum. Such a process is still a small fraction of all interactions of protons with ²⁰²Hg; α particle emission accounts for only 2% of the total inelastic cross section at 60–80 MeV. Because of the interest in the mechanisms responsible for this 2% of the total, however, it is hoped that the results of this work will further stimulate theoretical interpretations of α emission in nuclear reactions.

ACKNOWLEDGMENT

The authors wish to thank Mr. R. H. Mills and Mr. M. Della Neve of the McGill Synchrocyclotron for aid in performing the irradiations. The work would not have been possible without grants from the National Research Council of Canada and a special grant from the McGill University Faculty of Graduate Studies.

*Present address: McClellan Central Laboratory, McClellan Air Force Base, California 95652.

¹D. H. Wilkinson, in *Proceedings of the Rutherford Jubilee International Conference, Manchester, 1961*, edited by J. B. Birks (Academic, New York, 1961), p. 339.

²P. E. Hodgson, *Nucl. Phys.* **8**, 1 (1958).

³G. Igo, L. F. Hanson, and T. J. Gooding, *Phys. Rev.* **131**, 337 (1963).

⁴P. Kulisic, N. Cindro, and P. Strohal, *Nucl. Phys.* **73**, 548 (1965).

⁵N. Cindro, *Rev. Mod. Phys.* **38**, 391 (1966).

⁶L. Milazzo-Colli and G. M. Braga-Marcazzan, *Nucl. Phys.* **A210**, 297 (1973).

⁷J. Muto, H. Itoh, K. Okano, N. Shiomi, K. Fukuda, Y. Omori, and M. Kihara, *Nucl. Phys.* **47**, 19 (1963).

⁸H. Dubost, M. Lefort, J. Peter, and X. Tarrago, *Phys. Rev.* **136**, B1618 (1964).

⁹M. Lefort, J. P. Cohen, H. Dubost, and X. Tarrago, *Phys. Rev.* **139**, B1500 (1965).

¹⁰H. Dubost, B. Gatty, M. Lefort, J. Peter, and X. Tarrago, *J. Phys. (Paris)* **28**, 257 (1967).

¹¹E. Gadioli, I. Iori, N. Molho, and L. Zetta, *Phys. Rev. C* **4**, 1412 (1971).

¹²F. E. Bertrand and R. W. Peelle, *Phys. Rev. C* **8**, 1045 (1973).

¹³L. Milazzo-Colli, G. M. Braga-Marcazzan, and M. Milazzo, *Nucl. Phys.* **A218**, 274 (1974).

¹⁴M. Blann, *Annu. Rev. Nucl. Sci.* **25**, 123 (1975).

¹⁵H. Gauvin, *J. Phys. (Paris)* **24**, 836 (1963).

¹⁶R. A. Sharp, R. M. Diamond, and G. Wilkinson, *Phys. Rev.* **101**, 1493 (1956).

¹⁷N. T. Porile, S. Tanaka, H. Amano, M. Furukawa, S. Iwata, and M. Yagi, *Nucl. Phys.* **43**, 500 (1963).

¹⁸S. Hontzeas and L. Yaffe, *Can. J. Chem.* **41**, 2194 (1963).

- ¹⁹C. L. Rao and L. Yaffe, *Can. J. Chem.* 41, 2156 (1963).
- ²⁰R. Bimbot and M. Lefort, *J. Phys. (Paris)* 27, 385 (1966).
- ²¹D. R. Sachdev, N. T. Porile, and L. Yaffe, *Can. J. Chem.* 45, 1149 (1967).
- ²²S. Tanaka, M. Furukawa, and M. Chiba, *J. Inorg. Nucl. Chem.* 34, 2419 (1972).
- ²³J. S. Foster, J. W. Hilborn, and L. Yaffe, *Can. J. Phys.* 36, 555 (1958).
- ²⁴R. Collé, R. Kishore, and J. B. Cumming, *Phys. Rev. C* 9, 1819 (1974).
- ²⁵D. A. Newton, S. Sarkar, L. Yaffe, and R. B. Moore, *J. Inorg. Nucl. Chem.* 35, 361 (1973).
- ²⁶H. V. Weiss and W. L. Reichert, *Anal. Chim. Acta* 34, 119 (1966).
- ²⁷K. A. Kraus and G. E. Moore, *J. Chem. Soc.* 75, 1460 (1953).
- ²⁸W. R. Busing and H. A. Levy, Oak Ridge National Laboratory Report No. ORNL-TM-271, 1962 (unpublished).
- ²⁹K. J. LeCouteur, *Proc. Phys. Soc.* A63, 259 (1950).
- ³⁰J. J. Hogan, *Phys. Rev. C* 6, 810 (1972).
- ³¹M. V. Kantelo, Ph.D. thesis, McGill University, 1975 (unpublished).
- ³²H. Dubost, B. Gatty, M. Lefort, J. Peter, and X. Tarago, *J. Phys. (Paris)* 28, 257 (1967).
- ³³N. D. Dudev and T. T. Sugihara, *Phys. Rev.* 139, B896 (1965).
- ³⁴J. R. Grover and J. M. Gilat, *Phys. Rev.* 157, 183 (1967).
- ³⁵A. Pakkanen, H. Helppi, T. Komppa, and P. Puumalainen, *Z. Phys.* 254, 98 (1972).
- ³⁶J. C. Cunnane, R. Hockel, S. W. Yates, and P. J. Daly, *Nucl. Phys.* A196, 593 (1972).
- ³⁷H. Helppi and A. Pakkanen, *Z. Phys.* 255, 385 (1972).
- ³⁸M. B. Lewis, *Nucl. Data* B6, 355 (1971).
- ³⁹J. C. Cunnane and P. J. Daly, *Phys. Rev. C* 6, 1407 (1972).
- ⁴⁰A. Pakkanen, P. Puumalainen, H. Helppi, and T. Komppa, *Nucl. Phys.* A206, 164 (1973).
- ⁴¹R. L. Auble, *Nucl. Data* B6, 319 (1971).
- ⁴²M. R. Schmorak, *Nucl. Data* B7, 395 (1972).
- ⁴³M. J. Martin, *Nucl. Data* B8, 431 (1972).
- ⁴⁴R. L. Auble, *Nucl. Data* B7, 95 (1972).
- ⁴⁵M. B. Lewis, *Nucl. Data* B8, 389 (1972).
- ⁴⁶M. R. Schmorak, *Nucl. Data* B9, 195 (1973).
- ⁴⁷H. Verheul, *Nucl. Data* B2, 31 (1967).
- ⁴⁸I. Borchert, *Z. Phys.* 223, 473 (1969).
- ⁴⁹H. Verheul, *Nucl. Data* B2, 64 (1967).
- ⁵⁰A. H. Wapstra and N. B. Gove, *Nucl. Data* A9, 265 (1971).