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ABSTRACT

Cross sections for neutron-evaporation reactions from compound nuclei produced by bombardment of gold with carbon ions and of platinum with nitrogen ions have been determined. The magnitudes of the cross sections are considerably lower than would be predicted on the assumption that neutron emission is the only important mode of decay of the intermediate nuclei. This observation is explained on the basis of fission competition with neutron emission. To a much lesser extent, charged-particle evaporation is also a competing mode of decay. The arguments presented indicate that fission occurs either with comparable magnitudes in several nuclei in the neutron-evaporation chain, or preferentially in one or two nuclei near the end of the chain, rather than predominantly in the initial compound nucleus. Problems arising from the possible existence of isomers in the odd-odd astatine nuclides are discussed.

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I. INTRODUCTION

Many features of heavy-ion-induced reactions in the heavy-element region have been investigated. Britt and Quinton,[1] Goldberg, Reynolds, and Kerlee,[2] Gilmore,[3] Gordon et al.,[4] and Sikkeland and co-workers[5] have measured fission cross sections in heavy-ion reactions. Britt and Quinton have measured the cross sections, angular distributions, and energy spectra of alpha particles and protons produced in these reactions.[6] Angular distributions of fragments from heavy-ion-induced fission have been studied by Viola, Thomas, and Seaborg,[7] Britt and Quinton,[1] Goldberg, Reynolds, and Kerlee,[2] Gordon et al.,[4] and Sikkeland et al.[5]

We have measured the cross sections for production of various astatine nuclides by neutron evaporation from At^{212}, At^{210}, and At^{209} compound nuclei formed by C^{12} and N^{14} bombardments of gold and platinum, respectively. This work was done in order to account for another part of the total interaction cross section of heavy ions with gold and platinum nuclei and to provide some rather specific pieces of information that must be fitted by any theory that attempts to describe the heavy-ion reactions. The specific nature of the information furnished by measurements on the products formed by neutron evaporation arises from the fact that production of such an astatine nucleus means that fission and (or) charged-particle emission has not occurred in that

particular evaporation chain, whereas observation of a fission fragment or charged particle does not alone indicate in which nucleus the event originated, nor what other events preceded or followed the observed event.

II. EXPERIMENTAL WORK

A. General

In C^{12} bombardments of Au^{197}, two types of experiments were done. In one series of experiments, a thin gold target was bombarded with C^{12} ions of various energies, and the production of one or two prominent astatine activities was quantitatively measured at each energy. In other experiments, hereinafter designated "ratio experiments", stacked foils (consisting of gold targets and aluminum foils) were bombarded and the ratios of production of low-activity astatine nuclides to that of the prominent activities were measured. For measurements of $Pt(N^{14},xn)$ cross sections, in addition to those two types of experiments, it was necessary to do ratio experiments on a target of platinum enriched in Pt^{198}.

Bombardments were done at the Berkeley heavy-ion linear accelerator (Hilac), which accelerates heavy ions to 10.4 Mev per nucleon. Lower-energy particles were obtained by placing aluminum degrading foils in the beam path. Corrections for energy loss in the absorber foils, targets, and recoil catchers were made by use of the range-energy curves of Walton[8] and others, as summarized by Hubbard.[9]

B. Bombardment Procedures

1. Quantitative Experiments

The production of prominent astatine activities was determined quantitatively by use of a target chamber designed to allow rapid removal of the foil containing the reaction products. A collimator and absorbers, mounted on probes which could be inserted into the beam path, were placed in another chamber

preceding that containing the target. Thin, uniform targets were made by vaporizing $\leqslant 100\ \mu g/cm^2$ of gold or platinum metal onto a known area of previously weighed 0.25-mil Al foils. Target thicknesses were determined by weighing the foils before and after vaporization. Targets were placed in the holder with the target material down beam (i.e. on the side facing away from the impinging beam). Because the neutron-evaporation products are given large momenta along the beam direction,[10] they were knocked out of the target and collected on a 0.25-mil Al recoil catching foil placed immediately behind the target in the evacuated assembly. By direct counting of gross alpha activity in the target and catcher, the recoil collection efficiency was found to be approximately 99% for targets of thicknesses up to $100\ \mu g/cm^2$.

The target and its holder, and the recoil catcher and its holder (whose back plate was thick enough to completely stop the beam particles) were electronically isolated from the surroundings and served as the Faraday cup for measuring beam currents. Before striking the target assembly, the beam particles were always passed through stripping or absorber foils and could thus be considered fully ionized at all energies at which the reactions were studied.[11] The accuracy of the Faraday cup readings was investigated by substituting for the recoil catcher a similarly shaped holder containing a calorimeter. The latter was used to compare the power actually deposited by a beam of 124-Mev C^{12} ions with that calculated from a simultaneous measurement of the charge collected on the Faraday cup. The calorimetric and electronic measurements agreed to within 5%.

2. "Ratio Experiments"

Platinum. Targets for the "ratio experiments" were made by electroplating 200 to $300 \mu g/cm^2$ of metallic platinum onto 0.1-mil Ag foil. Targets of natural platinum and platinum enriched in Pt^{198} (60.95% Pt^{198}, 26.47% Pt^{196},

8.97% Pt^{195}, 3.57% Pt^{194}, 0.042% Pt^{192}, and 0.012% Pt^{190}) were used.[12] Target thicknesses were determined by weighing the foils before and after plating. The target foils were attached to stainless steel holders which were mounted (with target material down beam) in an assembly which had provision for circulating helium gas to cool the foils. The 0.25-mil Al catcher foils were also mounted on stainless steel holders and placed behind the targets. Spacers placed between the foils allowed the helium to circulate between them. The recoil method was used in order to preserve the targets, particularly those enriched in Pt^{198}. As the targets were considerably thicker than those used in the quantitative experiments, the recoil efficiency in the ratio experiments may have been several percent lower than that obtained in the quantitative experiments. However, the ratio of recoil collection efficiencies for various reaction products could be appreciably different from unity only if there were large differences in straggling in the recoil-range distributions. The data of Leachman and Atterling indicate that this is not a serious effect.[10]

Gold. Targets for the "ratio experiments" were of gold leaf (approx 2mg/cm^2). Pieces of gold leaf, backed by 0.25-mil Al foils, were stacked in the assembly.

C. Chemical Separations

1. Quantitative Experiments

As no At^{211} was produced in the Hilac bombardments, it was possible to determine chemical yields by using At^{211} as a tracer. The At^{211} was produced free of other detectable alpha activity by bombarding Bi^{209} with 28-Mev He^4 ions at the Crocker 60-in. cyclotron.[13] In order to have the astatine tracer in the same chemical environment as the Hilac-produced activity, the At^{211} atoms were also caught in an aluminum recoil catcher. Prior to the

heavy-ion bombardment, the recoil catcher was cut up and the alpha particles being emitted from each sample were counted to obtain the desired amounts of tracer activity.

Following the heavy-ion bombardment, the recoil catcher containing the heavy-ion-produced activities and a plate containing a known amount of tracer activity were simultaneously dissolved in 8M HCl. The astatine was separated from other alpha-emitting activities (mainly polonium) by extraction into di-isopropyl ether (DIPE). The DIPE fraction was transferred to a platinum counting disk and evaporated to dryness under a heat lamp.

2. "Ratio Experiments"

When it was necessary to measure only the ratios of the various activities produced, the double-vaporization method described in reference 14 was used. The samples produced by this method, being essentially mass-free gave well-resolved alpha-particle energy spectra (full width at half-maximum of the peaks approx 30 to 35 kev). The samples obtained from the chemical separation procedures which employed DIPE were rather thick, giving much more poorly resolved spectra. This was one of the reasons for doing the ratio experiments to determine yields of the less prominent activities. Also, the vaporization method was faster, allowing one to obtain better counting statistics for some of the very short-lived astatine nuclides. During the periods of counting (up to 8 hr) no loss of astatine from the counting plates was observed.

D. Counting Procedures

The only absolute counting was that of the plates containing At^{211} tracer. The alpha particles from At^{211} (and EC daughter, Po^{211}) were counted in an ionization chamber having 50% geometry.

For determining the ratio of prominent astatine alpha activities to that of the At^{211}, alpha particles emitted from the samples produced by the method involving extraction into DIPE were pulse analyzed in an alpha grid chamber connected to a 50- or 100-channel pulse-height analyzer. Decay properties of the astatine and polonium nuclides of interest are listed in Table I. Note that the 5.89-Mev alpha group from At^{211} falls in the same energy region as the alpha particles from At^{204} and At^{205}. For this reason, it was necessary to determine the tracer yield by observing the 7.44-Mev alpha particles from Po^{211}. Using the number of Po^{211} alpha particles counted and the alpha branching ratio of At^{211}, the contribution of tracer activity to the alpha-article group near 5.9 Mev was determined and subtracted. Frequently (as with At^{204}-At^{205} and At^{206}-At^{207}, for example) the energies of the alpha groups from two or more isotopes were so close together that their corresponding peaks could not be cleanly separated. In these cases, the counting rate corresponding to the combined peaks was plotted as a function of time and the decay curve was analyzed into its components.

E. Calculation of Cross Sections

Cross sections were calculated in the usual manner, using the branching ratios and half lives listed in Table I. For gold bombardments, the information from the ratio experiments was simply combined with that from the quantitative experiments to obtain the cross-section curves shown in Fig. 1. For the natural platinum targets, the prominent activity, At^{203} or At^{205}, (or both) was usually produced by more than one reaction because of the presence of the many platinum isotopes; thus, these quantitative experiments yielded the sum of the weighted cross sections of the several reactions producing the activity. Ratio experiments were done using both natural platinum and

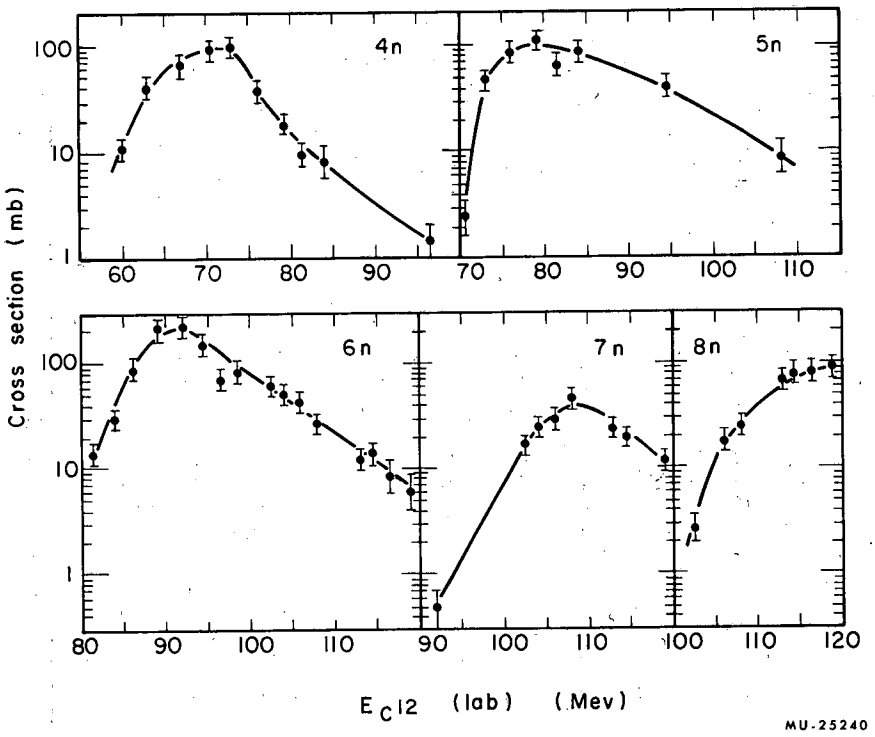

Fig. 1. Excitation functions for C^{12},xn reactions on Au^{197}.

Table. I. Decay properties of astatine and polonium isotopes

Isotopes	Half life	Reference	Alpha-particle energy (Mev)	Reference	Alpha decay Total disint.	Reference
At^{201}	1.5±0.1 min	15	6.348	15		
At^{202}	3.0±0.2 min	15,a	6.231(36%) 6.133(64%)	15	0.120±0.008	a
At^{203}	7.4±0.3 min	15,a	6.086	15	0.138±0.006	a
At^{204}	9.3±0.3 min	15,a	5.950	15	0.045±0.004	a
At^{205}	26.2±0.5 min	15,a	5.899	15	0.184±0.016	a
At^{206}	29.5±0.6 min	a	5.699	15	0.0088±0.0008	a
At^{207}	107.8±2.7 min	a	5.750	15	0.100	16
At^{208}	1.6±0.2 hr	17	5.65	18	0.0055±0.0005	18
At^{211}	7.17±0.09 hr	a	5.89	20	0.409	b
Po^{208}	2.93±0.03 yr	b	5.109	b	1.00	b
Po^{211}	0.52 sec	b	7.44	b	1.00	b

a. Determined in this work or Ref. 14

b. See compilation of Ref. 19

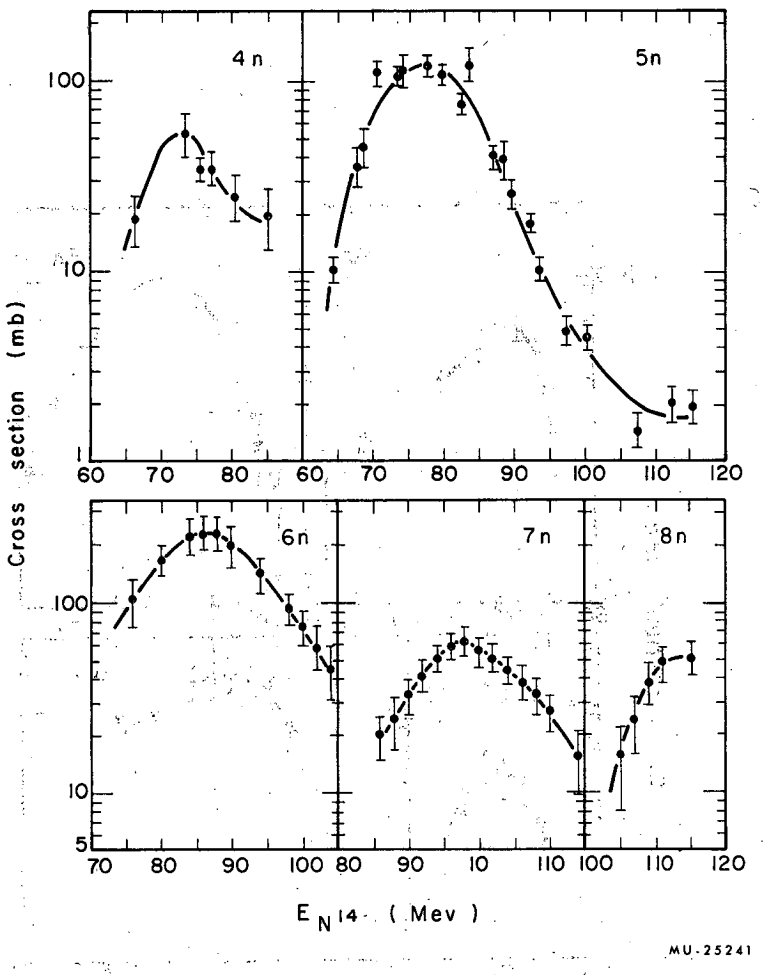

Fig. 2. Excitation functions for N^{14},xn reactions on Pt^{198}.

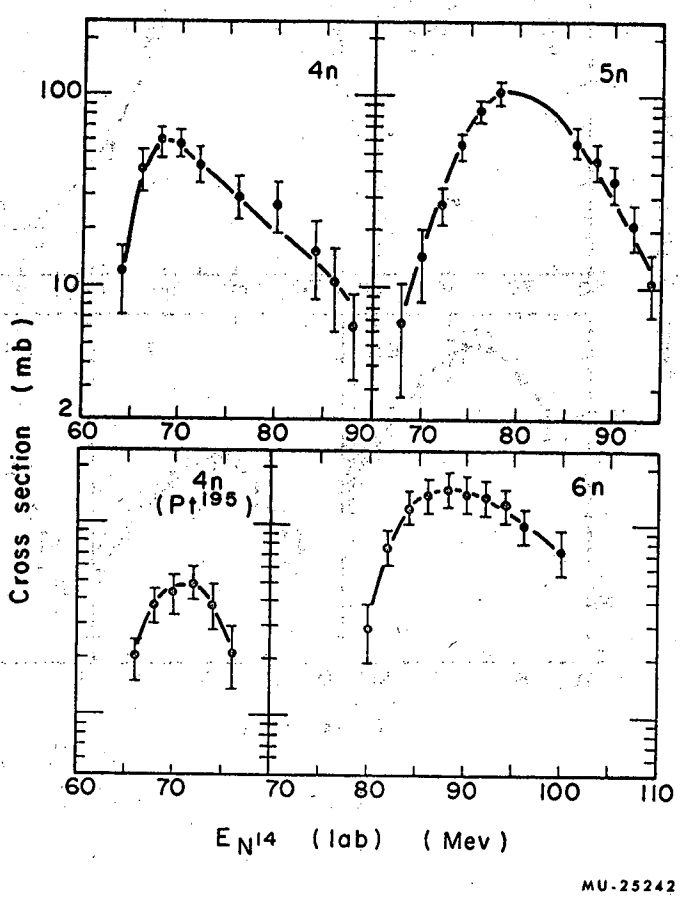

Fig. 3. Excitation functions for N^{14},xn reactions on Pt^{196} and for the N^{14},4n reaction on Pt^{195}.

enriched-Pt^{198} targets. These results were put into the appropriate simultaneous equations, and solutions of these equations were combined with the data from the quantitative experiments in order to obtain the cross-section curves shown in Fig. 2 and 3. Because of the complexity resulting from the presence of the many platinum isotopes, simplifying assumptions were required to obtain the individual cross sections. It is assumed that for platinum isotopes 196 or below, the N^{14},xn cross sections for $x \leqslant 3$ are negligible. Although it was not possible to check this assumption directly, careful observation of the alpha spectra of products from low-energy bombardments revealed no evidence for production of At^{209}, the product of the $Pt^{198}(N^{14},3n)$ reaction. This result, along with the small size of the observed N^{14},4n reaction cross sections, is taken as evidence for validity of the assumption. At the highest energies (about 100 Mev), it is assumed that most of the At^{205} resulted from the $Pt^{198}(N^{14},7n)$ reaction. A small correction was made for the amount from the $Pt^{196}(N^{14},5n)$ reaction by extrapolation of the high-energy tail of the excitation function. Since the data from the three types of experiments were usually not obtained at exactly the same bombarding energies, smooth curves were drawn through the experimental points corresponding to the absolute yields and ratios of isotopes produced as functions of energy. Thus, for reactions whose cross sections are given at regularly spaced energy intervals, this is not meant to imply that cross sections were determined at exactly those energies, but that the simultaneous equations were solved at those particular energies. Scatter in the experimental points was necessarily removed by the smoothing process. The limits of error given for these cross sections represent estimated uncertainties in the positions of the smoothed curves. These estimates, as well as the limits of error given for the other cross sections, include uncertainties in the counting rates of the radiations

from the product nuclei, target thickness, and decay-during-bombardment corrections. Uncertainty in the values used for the α/EC branching ratios have not been included.

III. DISCUSSION

Specific mention of the details of certain of the reaction products should be made.

At^{201}. The alpha branching ratio is not known for At^{201}. From the trends in the branching ratios of the other astatine isotopes, we have arbitrarily used 0.1 for this quantity, with the result that the absolute values of the $Au^{197}(C^{12},8n)$ cross sections may be substantially in error.

At^{204}-At^{206}. In a previous paper, we noted the possible existence of isomers for At^{204} and At^{206}.[14] Barton et al.[16] and Stoner[17] reported half lives of 25 min and ~2.8 hr for At^{204} and At^{206}, respectively, formed by high-energy-He^4 bombardment of Bi^{209}. We have observed half lives of 9.4 min and 29.5 min for these isotopes formed by heavy-ion bombardment. We have set upper limits of 0.04 and 0.07 on the ratio of the amount of a long-lived isomer to that of the short-lived isomers produced. The decay properties of the astatine isotopes as reported by Hoff, Asaro, and Perlman,[15] who formed the astatine isotopes by heavy-ion bombardment, are in good agreement with those we have measured.

At^{208}. The existence of two isomers of At^{208} is well known. Both the 1.7-hr and 6.3-hr isomers have been produced by high-energy-He^4 ions on Bi^{209}.[16,17] The 1.7-hr isomer is also formed in the alpha decay of Fr^{212}.[18] In this work, the 1.7-hr isomer was frequently observed, but no evidence for production of the 6.3-hr activity was found. Unfortunately, the alpha activity of the daughter, Po^{208}, was too weak and the alpha branching ratio of the 1.7-hr At^{208} too uncertain to permit us to set an upper limit on the ratio of production of

the 6.3-hr isomer to that of the 1.7-hr one. Thus it would appear that heavy-ion reactions produce short-lived odd-odd astatine isomers, whereas helium-ion bombardments yield the longer-lived isomers. In searching for an explanation of this result, one's first inclination might be to suggest that the isomers, having very different spins, are produced in a ratio that is strongly dependent upon the angular momentum deposited in the compound nucleus in the reaction. This explanation is, however, unattractive for two reasons:

(a) The work of Pik-Pichak,[20] Hiskes,[21] and Huizenga and Vandenbosch[22] suggests that fissionability increases with increasing angular momentum. Calculations based on the Huizenga-Vandenbosch formulation indicate that high-spin states are largely removed by fission.[23] This effect would tend to "wash out" differences in the spin spectra of nuclei resulting from He^4 or heavy-ion bombardments.

(b) It seems unlikely that an angular momentum effect would be this exclusive, i.e., that one of the isomers is formed nearly exclusively in one type of bombardment and almost not at all in the other type of reaction.

Clearly this problem of odd-odd astatine isomers is unresolved, and more work should be done on it.

It should be noted that the peak heights of the various excitation functions for the $Au^{197}(C^{12},xn)$ reactions do not show a smooth behavior with increasing number of neutrons emitted. Some of the curves may be low because of failure to account for the formation of isomers that do not decay by alpha emission or that have very short half lives for alpha emission. The effect might also result from errors in the alpha branching ratios of the polonium isotopes upon which the alpha branching ratios of the astatine isotopes were based.[14]

In Fig. 4 we have plotted the sum of the "reduced cross sections" for the production of nuclides resulting from neutron-evaporation reactions as a

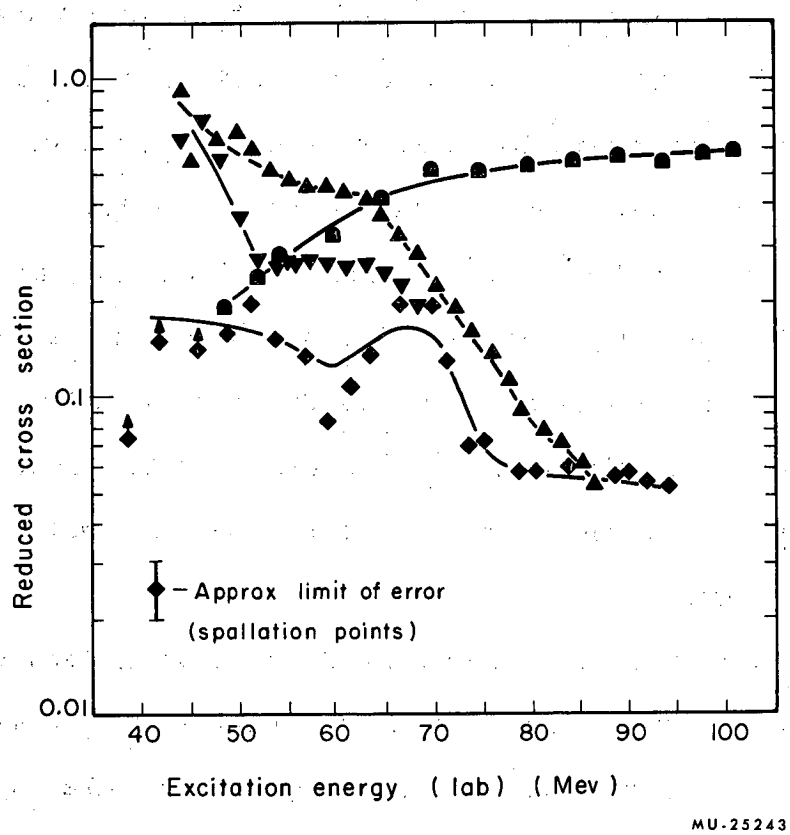

Fig. 4. Reduced cross sections (see text) for neutron-evaporation and fission reactions of astatine compound nuclei.

○ - Reduced fission cross section for $Au^{197} + C^{12}$ (compound nucleus, at^{209}). Data from Reference 4.

□ - Sum of the reduced neutron-evaporation cross sections for $Au^{197} + C^{12}$ (compound nucleus, At^{209}).

▽ - Sum of the reduced neutron-evaporation cross sections for $Pt^{196} + N^{14}$ (compound nucleus, At^{210}).

△ - Sum of the reduced neutron-evaporation cross sections for $Pt^{198} + N^{14}$ (compound nucleus, At^{212}).

function of excitation energy of the initial compound nucleus for the various systems studied. (The "reduced cross section" is the actual cross section divided by the cross section for compound-nucleus formation, which is here taken to be that calculated by Thomas[24]). For Au + C^{12}, this sum decreases from 0.20 at low energies to ~0.05 at the highest energies, and for Pt + N^{14}, from ~0.7 to about 0.06. This is in contrast to the observations by Bell and Skarsgard, who found, for protons on bismuth to produce excitation energies in the range studied here, that the sum of the p,xn cross section was nearly equal to the calculated reaction cross section over the entire energy range.[25] Subsequent work by Kavanagh and Bell indicates that most of the reaction cross section not accounted for by p,xn reactions is taken up by p,pxn reactions.[26] Thus, in the heavy-ion reactions, many of the neutron-evaporation products are removed by competing reactions. From the data of Gordon et al.[4] (plotted in Fig. 4 as the reduced fission cross section), it is clear that the principal competing reaction is fission, as it accounts for a major portion of the reaction cross section. The data of Britt and Quinton, who studied alpha-particle and proton emission in heavy-ion-induced reactions, demonstrate that there is also competition from these reactions.[6] However, competition from charged-particle evaporation is much less important than that from fission, since for Au^{197} + 126-Mev C^{12}, Britt and Quinton found a cross section of only 120 mb for alpha-particle evaporation, and for Bi^{209} + 126-Mev C^{12}, 110 mb for proton evaporation (representing reduced cross sections of about 0.05). The cross sections for charged-particle evaporation decrease rapidly with decreasing excitation energy.[6]

Let us consider the following three different assumptions regarding the stage in the evaporation chain at which fission occurs: (a) that it occurs almost exclusively in the original compound nucleus; (b) that it

occurs mainly in one nucleus near the end of the evaporation chain (i.e. at some low excitation energy); (c) that it occurs with comparable probability in several nuclides in the chain.

There is some evidence to indicate that assumption (a) is probably not correct. If it were correct, the reduced fission cross sections at one energy would be negligibly small compared with the reduced cross section at an energy higher by slightly more than the separation energy of a neutron (see the arguments presented by Fairhall et al. concerning this interpretation of fission cross-section data[27]). Otherwise, one could expect appreciable contributions to the fission cross section from other than the initial compound nucleus. That the reduced fission cross section does not drop this rapidly with decreasing energy may be seen in Fig. 4.

It might be argued that because of direct interactions, the cross section for true compound-nucleus formation is less than the calculated reaction cross section, particularly at the higher energies. Hence, the reduced fission cross sections may actually rise more steeply than indicated in Fig. 4. However, even if we assume that the reduced cross section for fission has reached unity at the highest energy shown, there is a change of only a factor of 5 in this quantity for a change in excitation energy of 50 Mev. This corresponds to a change by a factor of about 1.4 for each neutron emitted (assuming 10 Mev per neutron).

Furthermore, measurements of angular distributions of fragments from fission of gold with carbon ions, and other heavy-ion-induced fission reactions in this region, have been interpreted as indicating that, on the average, several neutrons are emitted prior to fission.[4,7] Because of the number of assumptions that must be made in arriving at these interpretations, this conclusion is not completely convincing, but it does lend weight to the

argument that assumption (a) is not correct.

The data currently available are not sufficient to make a decision between assumptions (b) and (c). An argument in favor of (b) has been presented by Thomas.[28] If (b) is indeed correct, then Γ_f/Γ (ratio of level width for fission to the total level width) is effectively zero during the early part of the evaporation chain, rising sharply at the end.

If, on the other hand, assumption (c) is correct, only a rather small value of Γ_f/Γ is needed to account for the data. The sum of the reduced neutron-evaporation cross sections in the region where the $Au^{197}(C^{12},7n)$ reaction predominates is approximately equal to $\langle\Gamma_n/\Gamma\rangle$,[7] where $\langle\Gamma_n/\Gamma\rangle$ is an average value of Γ_n/Γ over the evaporation chain. From the sum of the reduced cross sections, 0.05, we can conclude $\overline{\Gamma_n/\Gamma} \approx 0.65$. Assuming $\Gamma = \Gamma_n + \Gamma_f$ (i.e., ignoring the competition from charged particle evaporation), we conclude that $\langle\Gamma_f/\Gamma\rangle$ is 0.35. Thus, although the reduced fission cross section is 10 or 12 times the reduced cross section for neutron emission, the probability for neutron emission at any step during the de-excitation is about twice that for fission.

At lower excitation energies, the magnitude of fission competition appears to decrease with increasing mass number in the series of compound systems At^{209}, At^{210}, At^{212}. This decrease may be due to higher fission barriers in the more neutron-rich nuclei because of their lower values of the usual fission parameter, Z^2/A. Also, in the Swiatecki formulation of fission barriers, the positive corrections to the liquid-drop barriers for the ground-state masses are greater for the more neutron-rich compound system.[29] Because of the apparent convergence of the reduced neutron-evaporation cross sections for the various systems at higher excitation energies, it appears that the differences in fission competition in the various systems are washed out at the higher excitation energies.

IV. SUMMARY

We have found that the reduced cross sections for the formation of products resulting from neutron evaporation in heavy-ion-induced reactions in the astatine region are substantially less than the cross sections for formation of such products in proton-induced reactions. The principal competing reaction in the heavy-ion-induced reactions is fission.

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FOOTNOTES AND REFERENCES

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