

## Excitation Functions for Nuclear Reactions between Complex Nuclei. II. Charged Particle Emission

T. J. KLINGEN\* AND G. R. CHOPPIN

Department of Chemistry, Florida State University, Tallahassee, Florida

(Received 5 July 1962; revised manuscript received 14 February 1963)

The production of  $\text{La}^{135}$ ,  $\text{Ba}^{135m}$ , and  $\text{Ba}^{133m}$  from the nuclear reactions of  $\text{Te}^{128} + \text{C}^{12}$ ,  $\text{Te}^{130} + \text{C}^{12}$ , and  $\text{Sn}^{124} + \text{O}^{16}$  has been measured radiochemically. The function of  $\text{La}^{135}$  appears to occur by nucleon evaporation following compound nucleus formation. The maximum cross section is consistent with this picture when compared to the maximum cross section for  $\text{Ce}^{135}$  formation. The maximum cross section for the formation of the barium isotopes is much greater than for formation of the lanthanum and cerium isobars. This would seem to suggest a direct-interaction mechanism for alpha particle emission.

### INTRODUCTION

IN our first paper<sup>1</sup> the effects of angular momentum on the excitation functions of heavy-ion-induced nuclear reactions has been considered for the emission of neutrons. The excitation functions for the emission of light charged particles (protons and alphas) were also determined. The reactions studied produced  $\text{La}^{135}$ ,  $\text{Ba}^{133m}$ , and  $\text{Ba}^{135m}$  by  $\text{C}^{12}$  bombardment of  $\text{Te}^{128}$  and  $\text{Te}^{130}$  and by  $\text{O}^{16}$  bombardment of  $\text{Sn}^{124}$ .

In their investigations of fission induced by heavy-ion bombardment of gold and platinum, Blann<sup>2</sup> and Gordon *et al.*<sup>3</sup> found that their results could be best interpreted

on the basis of increasing probability for charged particle emission with increasing excitation energy of the compound nucleus. However, because of the competition of fission, these workers were unable to determine directly the amount of competition between neutron and charged particle evaporation. Gordon<sup>4</sup> has estimated that, for  $\text{Au}^{197} + \text{C}^{12}$ ,  $\sigma$  (charged particle)/ $\sigma$  (compound nucleus formation) has a value of 0.5 to 0.6. Our purpose was to obtain a more direct comparison between neutron evaporation and charged particle emission in the absence of fission.

### EXPERIMENTAL

The experimental method used in this investigation was similar to that used for the determination of the excitation functions for neutron emission in these same reactions.<sup>1</sup> The lanthanum and barium nuclides produced in the bombardments were separated chemically, simultaneously with the cerium isotopes, using a procedure basically similar to that given by Hicks.<sup>5</sup> The final lanthanum counting samples were obtained as lanthanum oxalate nonahydrate, and the final barium counting samples were obtained as barium sulfate.

The only lanthanum isotope formed which was radioactive was  $\text{La}^{135}$ , which decays by electron capture. The activity of the lanthanum counting samples was determined from the intensity of their x-ray spectra. Although the method of calculation of the absolute decay and, then, of the cross section for the production of  $\text{La}^{135}$  as a function of bombarding energy is basically the same as that described previously (see reference 1 for details), it was necessary to correct the absolute counting rate for the amount of  $\text{La}^{135}$  produced by the decay of  $\text{Ce}^{135}$ . This correction was accomplished by using the measured amount of  $\text{Ce}^{135}$  formed and the time from end of bombardment to separation of La and Ce (26 to 30 h).

In the case of the barium isotopes,  $\text{Ba}^{133m}$  and  $\text{Ba}^{135m}$ , the counting and determination of the absolute cross sections was accomplished by measurements of the

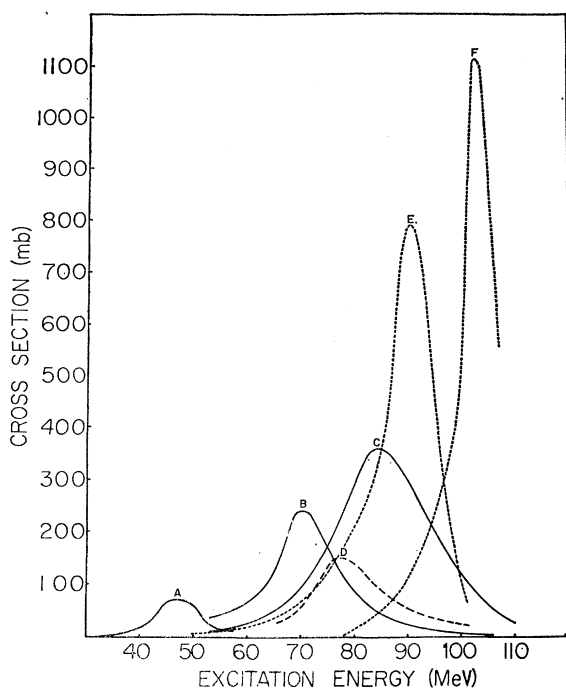


FIG. 1. Excitation functions for  $\text{Te}^{128} + \text{C}^{12}$ ; A =  $\text{Ce}^{137m}$ , B =  $\text{Ce}^{135}$ , C =  $\text{Ce}^{134}$ , D =  $\text{La}^{135}$ , E =  $\text{Ba}^{135m}$ , F =  $\text{Ba}^{133m}$ .

\* Based on the Ph.D. dissertation submitted to Florida State University, 1962, by T. J. K.

<sup>1</sup> G. R. Choppin and T. J. Klingen, *Phys. Rev.* (to be published).

<sup>2</sup> H. M. Blann, *Phys. Rev.* **123**, 1356 (1961).

<sup>3</sup> G. E. Gordon, A. E. Larsh, T. Sikkeland, and G. T. Seaborg, *Phys. Rev.* **120**, 1341 (1960).

<sup>4</sup> G. E. Gordon, Lawrence Radiation Laboratory Report, UCRL-9083, 1960 (unpublished).

<sup>5</sup> H. G. Hicks, in "The Radiochemistry of the Rare Earths," edited by P. C. Stevenson and W. E. Nervi, NAS-NS-3020 (1961). Available through the OTS.

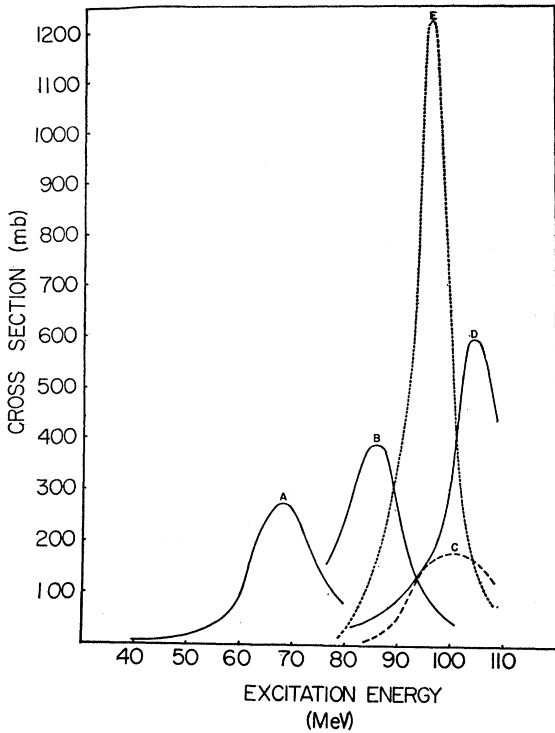


FIG. 2. Excitation function for  $Te^{130} + C^{12}$ ; A =  $Ce^{137m}$ , B =  $Ce^{135}$ , C =  $La^{135}$ , D =  $Ce^{134}$ , E =  $Ba^{135m}$ .

internal transition gamma spectra of the counting samples. Both isotopes have internal transition gamma rays of the order of 270 keV. In addition to this, the

TABLE I. Charged-particle excitation function data for  $Te^{130} + C^{12}$ .

$E$ (MeV)	$p^4n$ $\sigma$ (mb)	$\pm \epsilon_\sigma$ (mb)	$E$ (MeV)	$\alpha n$ $\sigma$ (mb)	$\pm \epsilon_\sigma$ (mb)
83	8	3	79	14	6
86	17		80	36	
88	32		81	50	
90	49		83	71	
91	95	21	84	104	
93	124		85	128	40
95	153		86	157	
97	171		87	207	
101	180	27	89	289	
105	159		90	342	84
108	140	24	91	442	
			93	570	
			94	698	
			94	870	
			95	998	
			96	1169	
			97	1219	164
			97	1197	
			98	1112	
			99	855	
			100	627	92
			101	428	
			101	406	
			105	171	
			106	114	
			108	80	14

TABLE II. Charged particle excitation function data for  $Te^{128} + C^{12}$ .

$E$ (MeV)	$p^4n$ $\sigma$ (mb)	$\pm \epsilon_\sigma$ (mb)	$E$ (MeV)	$\alpha n$ $\sigma$ (mb)	$\pm \epsilon_\sigma$ (mb)	$E$ (MeV)	$\alpha 3n$ $\sigma$ (mb)	$\pm \epsilon_\sigma$ (mb)
66	34	6	50	7	1	78	3	
67	31		55	11		80	17	[5] <sup>a</sup>
68	36		60	5		83	31	
69	42		61	25		85	51	
71	75		64	32		87	77	
72	89	14	66	20		89	111	[23] <sup>a</sup>
73	99		70	97	11	92	154	
74	109		71	59		94	223	
74	124	19	73	123		97	334	
75	139		75	128		99	462	[70] <sup>a</sup>
76	148		75	148		100	616	
78	149	21	77	123		101	770	
80	137		79	185	31	103	1113	122
82	135		80	210		105	830	
84	121		82	259		106	599	72
85	115	19	83	296				
88	76		84	358				
90	49		86	469	113			
92	32		87	555				
96	27		88	617				
101	24	5	90	765				
			91	789				
			92	703				
			93	641				
			94	493				
			95	407				
			96	296	[96] <sup>a</sup>			
			98	222				
			99	160				
			101	62	[23] <sup>a</sup>			

<sup>a</sup> Assumes validity of curve resolution method.

half-lives of the barium isotopes are 39 h ( $Ba^{138m}$ ) and 29 h ( $Ba^{135m}$ ). Since the barium isotopes could not be individually identified from their decay characteristics, a plot of the total barium activity vs bombardment energy was obtained. These plots showed two maxima for the  $Te^{128}$  and  $Sn^{124}$  systems, one for  $Ce^{133m}$  and one for  $Ce^{135m}$ . These maxima are reasonably separated in

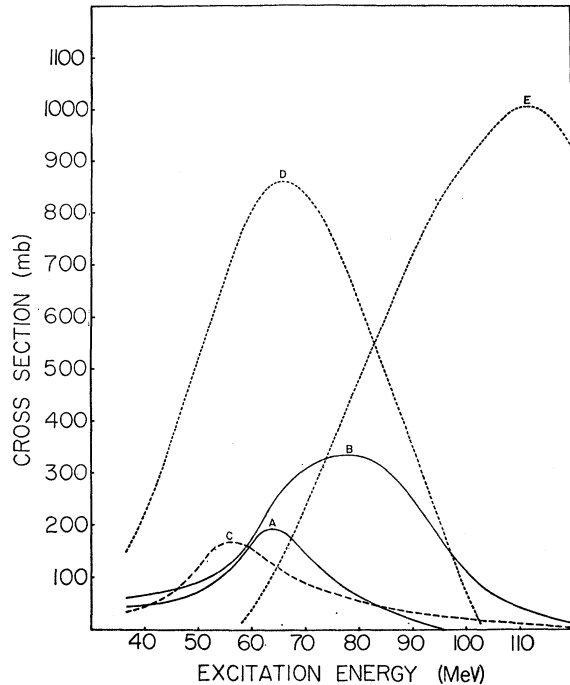


FIG. 3. Excitation function for  $Sn^{124} + O^{16}$ ; A =  $Ce^{135}$ , B =  $Ce^{134}$ , C =  $La^{135}$ , D =  $Ba^{135m}$ , E =  $Ba^{138m}$ .

TABLE III. Charged particle excitation function data for  $\text{Sn}^{124} + \text{O}^{16}$ .

$E$ (MeV)	$p4n$ $\sigma$ (mb)	$\pm \epsilon_\sigma$ (mb)	$E$ (MeV)	$\alpha n$ $\sigma$ (mb)	$\pm \epsilon_\sigma$ (mb)	$E$ (MeV)	$\alpha 3n$ $\sigma$ (mb)	$\pm \epsilon_\sigma$ (mb)
37	36	9	37	150	23	58	15	[8] <sup>a</sup>
45	64		45	354		63	74	
52	152		52	588		67	162	
60	160	35	58	768	115	69	214	
63	125		60	818		72	266	
67	104		63	848		76	377	[143] <sup>a</sup>
69	94		65	858	173	78	436	
73	81		67	858		80	495	
76	71		69	843		85	598	
80	54	12	72	818		87	654	[203] <sup>a</sup>
81	47		76	738	[214] <sup>a</sup>	89	702	
85	35		78	688		94	790	
86	33		80	628		96	831	
91	29		85	519		98	864	207
91	29		87	459	[174] <sup>a</sup>	103	931	
95	27		89	389		107	982	
97	25		94	249		112	1004	151
100	20	4	96	180		116	979	
102	17		98	120	[56] <sup>a</sup>	120	931	112
105	10		103	15				
107	12							
112	10							
116	5	1						
120	2							

<sup>a</sup> Assumes validity of curve resolution method.

the tellurium carbon system but overlap badly in the tin-oxygen system.

The method for the resolution of these two activities was to assume that the low-energy side of the  $\text{Ba}^{133m}$  curve had the same shape as the low-energy side of the  $\text{Ba}^{135}$  curve. Subtraction of this from the gross barium activity gave the high-energy side of the  $\text{Ba}^{135m}$  curve. A comparison of the percentage width at half-maximum energy of the two separated activity vs energy curves acted as an internal check on this method of resolution, since it would be expected that the percentage widths of the two isotope curves would be approximately equal. Obviously, this method will introduce error not only in the cross sections for these isotopes but also in the determination of the energy of the excitation function maximum for each isotope. However, in lieu of a better method, this procedure allowed a first approximation of the excitation functions of the barium isotopes.

Following this resolution the activities at the various energies for each isotope were corrected to end of bombardment. These activities in turn were converted into absolute disintegration rates using the total efficiency of the counter,<sup>6</sup> the window absorption,<sup>7</sup> branching ratios,<sup>8</sup> and conversion coefficients.<sup>9,10</sup> Once the absolute disintegration rate of the barium isotopes at end of bombardment was obtained as a function of bombarding energy, the production cross sections were calculated in the usual manner.

<sup>6</sup> S. H. Vegors, L. L. Marsden, and R. L. Heath, AEC Research and Development Report IDO-16370, 1958 (unpublished).

<sup>7</sup> G. R. White, U. S. National Bureau of Standards Circular No. 583, 1957 (unpublished).

<sup>8</sup> D. Strominger, J. M. Hollander, and G. T. Seaborg, Rev. Mod. Phys. **30**, 585 (1958).

<sup>9</sup> J. M. Cork and G. P. Smith, Phys. Rev. **60**, 480 (1941).

<sup>10</sup> R. N. Hill and F. R. Metzger, Phys. Rev. **83**, 455 (1951).

## RESULTS

The excitation functions for the formation of  $\text{La}^{135}$ ,  $\text{Ba}^{133m}$ , and  $\text{Ba}^{135m}$ , as well as  $\text{Ce}^{134,135,137}$ , are shown as a function of excitation energy of the compound nucleus in the center-of-mass system in Figs. 1, 2, and 3. For clarity, these excitation functions are shown as smooth curves without experimental points and error limits. The data from which these curves were constructed are shown in Tables I, II, and III. The values of the maximum cross sections are given in Table I for the production of the cerium, lanthanum, and barium nuclides. The position of the maximum in the  $\text{La}^{135}$  curves is rather sensitive to the value of the  $\text{Ce}^{135}$  production cross section, since the calculation of the  $\text{La}^{135}$  cross section involves correction for the  $\text{Ce}^{135}$  decay. The fact that the calculated  $\text{La}^{135}$  maximum occurs at a lower energy than the  $\text{Ce}^{135}$  maximum for the  $\text{Sn}^{124} + \text{O}^{16}$  system may be due to the inaccuracy of the  $\text{Ce}^{135}$  cross section plus the large overlap of the two curves. The relationship of the  $\text{Ce}^{135}$  and  $\text{La}^{135}$  for the  $\text{Te}^{128}$  and  $\text{Te}^{130}$  systems would seem more reasonable. The error limits listed are based on the counting correction factors, etc., and are almost certainly too small considering the uncertainties in the parent-daughter correction.

## DISCUSSION

In the case of the lanthanum excitation function curves the maxima are all less than the maxima of the corresponding cerium curves, although their shapes are similar. The energetics of the reactions producing the  $\text{La}^{135}$  by either a compound nucleus mechanism or a direct interaction mechanism agree equally well with the experimental excitation functions. Because of the nature of the experimental technique used in this work, the mechanism of the reaction could not, therefore, be established uniquely. However, on the basis of the shape, height, and position of the lanthanum excitation function curves relative to the cerium curves, we feel that the  $p(x-1)n$  reactions proceed primarily via compound nucleus formation with subsequent nucleon evaporation.

The excitation functions for the barium isotopes, on the other hand, are quite different from those for the production of either the cerium or lanthanum isotopes. The barium excitation function curves for all three systems peak at cross sections which are three to four times higher than those for the cerium curves. In the  $\text{Sn} + \text{O}^{16}$  reactions,  $\text{Ba}^{135m}$  can be produced by

TABLE IV. Maximum values of the cross sections in mb.

System	$5n$	$p4n$	$\alpha n$	$7n$	$p6n$	$\alpha 3n$
$\text{Te}^{128} + \text{C}^{12}$	240	150	790	...	...	1100
$\text{Te}^{130} + \text{C}^{12}$	...	...	...	390	180	1220
$\text{Sn}^{124} + \text{O}^{16}$	195	165	865	...	...	1000

$\text{Sn}^{124}(\text{O}^{16}, 2p3n)$  and  $\text{Sn}^{122}(\text{O}^{16}, 2pn)$  where  $\text{Sn}^{124}$  is in 6.1% and  $\text{Sn}^{122}$  in 4.8% abundance.  $\text{Ba}^{133m}$  can be produced by  $\text{Sn}^{124}(\text{O}^{16}, 2p5n)$ ,  $\text{Sn}^{122}(\text{O}^{16}, 2p3n)$ , and  $\text{Sn}^{120}(\text{O}^{16}, 2pn)$  with  $\text{Sn}^{120}$  in 32% abundance in the natural tin target. It is not possible to apply any trustworthy corrections to resolve the relative contributions of the different reactions to the total production as could be done in the reactions forming cerium.<sup>1</sup> Consequently, it is not possible to place any significance in the width of the peaks in the Sn+O reactions until the reactions have been studied for the separated tin isotopes.

Here again, the energetics of the systems are equally well accounted for by either a compound nucleus mechanism or a direct interaction mechanism. Knox, Quinton, and Anderson<sup>11</sup> and Britt and Quinton<sup>12</sup> have investigated the production of light charged particles from heavy-ion induced reactions using targets both lighter and heavier than those used in this study. These workers have observed very large production cross sections for both protons and alpha particles and found that they were primarily produced by a direct interaction mechanism. Thus, it would not seem unreasonable to assume that the barium products of the reactions studied here are produced by some direct interaction mechanism involving ejection of an alpha particle.

<sup>11</sup> W. J. Knox, A. R. Quinton, and C. E. Anderson, Phys. Rev. **120**, 2120 (1960).

<sup>12</sup> H. C. Britt and A. R. Quinton, Phys. Rev. **124**, 877 (1961).

Since  $\text{Ba}^{133g}$  and  $\text{Ba}^{135g}$  direct production was not measured, these experimental cross sections may actually be lower limits. However, the magnitude of the cross section maxima is indicative that  $\text{Ba}^{133g}$  and  $\text{Ba}^{135g}$  formation must be quite a bit less than that of  $\text{Ba}^{133m}$  and  $\text{Ba}^{135m}$  or that they are formed at different (presumably lower) energies. The difference of 15 MeV for the maxima for  $\text{Tb}^{149g}$  and  $\text{Tb}^{149m}$  is an example of the latter possibility<sup>13</sup> and is, presumably, due to an effect of angular momentum.<sup>1</sup>

In summary, charged-particle emission accounts for a significant fraction of the total reaction cross section for nuclear reactions induced by heavy ions. The proton-out reaction cross sections (in comparison to the neutron-only-out cross sections) are about as large as expected from simple compound nucleus-nucleon evaporation theory, when a possible contribution from direct interaction processes is included. However, the cross sections involving alpha emission are much larger and would seem to be due to a direct interaction mechanism.

We wish to acknowledge the financial support of the U. S. Atomic Energy Commission. The operating crew of the Heavy Ion Linear Accelerator of the Lawrence Radiation Laboratory as well as Dr. I. Perlman, Dr. B. G. Harvey, Dr. A. Ghiorso, Dr. T. Sikkeland, Mrs. Roberta Garrett, and Ray O'Dea have made this research possible by their generous assistance in many ways.

<sup>13</sup> R. D. Macfarlane, Phys. Rev. **126**, 274 (1962).