Production of heavy actinides from interactions of ^{16}O, ^{18}O, ^{20}Ne, and ^{22}Ne with ^{248}Cm

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We have measured cross sections for the production of isotopes of Bk through No in bombardments of ^{248}Cm with ^{16}O, ^{18}O, ^{20}Ne, and ^{22}Ne ions at energies near the Coulomb barrier. In general, the peak of the mass-yield curve for each element is about two mass units larger for ^{18}O and ^{22}Ne than for ^{16}O and ^{20}Ne, reflecting the neutron excess of the projectiles. The production cross sections and maxima of the actinide isotopic distributions are at least as favorable for production of neutron-rich actinides as those measured for irradiation of ^{238}U and ^{248}Cm with very heavy ions. The observation of so many neutron-rich products between the masses of the target and compound nucleus suggests a direct transfer reaction in which the product nuclide is formed with relatively low excitation which minimizes depletion from prompt fission.

[NUCLEAR REACTIONS $^{248}Cm(^{16,18}O, X)$, $(^{20,22}Ne, X)$ $E(^{16,18}O) =$ 98, 97 MeV, $E(^{20,22}Ne = 115, 116$ MeV); measured σ and isotopic distributions for $Z = 97-102$.]

I. INTRODUCTION

Multinucleon transfer from heavy-ion projectiles to target nuclei is a phenomenon that has been known for some time. A variety of techniques[1,2] has been used to measure details of the product kinetic energies and angular distributions. Hahn *et al.*[3] have investigated the reactions of ^{12}C with ^{238}U and ^{239}Pu leading to the production of ^{244}Cf and ^{245}Cf. Demin *et al.*[4] have recently measured excitation functions for production of ^{246}Cf, $^{251,253}Es$, $^{250-254}Fm$, and ^{256}Md from irradiation of ^{249}Cf with ^{22}Ne. Considerable attention has also been devoted to the production of heavy actinides and possible superheavy elements by deep inelastic transfer reactions in bombardments of ^{238}U and other heavy actinide targets with heavier ions[5-13] such as ^{48}Ca, ^{86}Kr, ^{136}Xe, and ^{238}U. Despite this considerable effort, the reaction mechanisms leading to product masses between the target and compound nucleus have not been fully explored. Investigations of transfer-type reactions in the actinide region, where fission represents an important reaction channel, offer a possibility for evaluating the amount of excitation energy transferred to the product nucleus.

Measurements with a broad range of heavy-ion projectiles and actinide targets should enable us to make predictions concerning the production of new isotopes and possibly also of new elements. A comprehensive survey of such reactions might further help to assess the possibilities for reaching the region of superheavy elements using the heaviest available actinide targets and suitable lighter heavy-ion projectiles.

In the current work we have started such a systematic study using radiochemical techniques to investigate the cross sections for the formation of isotopes of Bk through No produced in bombardments of ^{248}Cm with ^{16}O, ^{18}O, ^{20}Ne, and ^{22}Ne ions at energies close to the calculated Coulomb barriers. The isotopic pairs ^{16}O-^{18}O and ^{20}Ne-^{22}Ne

were chosen in order to assess the influence of the extra pair of neutrons in each case.

II. EXPERIMENTAL PROCEDURES

A. Bombarding conditions and target arrangement

Irradiations of ^{248}Cm were performed at the 88-inch cyclotron of the Lawrence Berkeley Laboratory with 98-MeV ^{16}O, 97-MeV ^{18}O, 115-MeV ^{20}Ne, and 116-MeV ^{22}Ne ions. Typical irradiation times were from 6 to 12 h. A diagram of the target arrangement is shown in Fig. 1. The beam intensity was about 1.5 to 2 μA for all irradiations and was monitored and the integrated current recorded every 10 min using a microcomputer. The target contained 0.924 mg/cm^2 of Cm (96.5% ^{248}Cm; 3.5% ^{246}Cm) in the form of CmF_3 deposited with a diameter of 6.5 mm on 2.65 mg/cm^2 Be foil. Gold foils, 19.5 mm in diameter and 2 mg/cm^2 thick, were placed $\approx$3 mm from the target to catch the recoiling products.

B. Radiochemical separations

The Au catcher foils were dissolved in concentrated HCl containing a small amount of nitric acid. This solution was passed through a 3-mm diam$\times$40-mm long column of AG-1, X10 anion exchange resin which had been pretreated with 10 M HCl. The Au was completely removed by adsorption on the resin while the actinides passed through. The actinide elements from Bk through Md were individually separated by elution from a

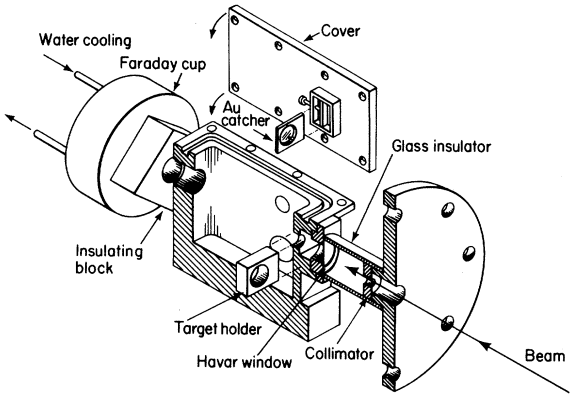

FIG. 1. Expanded view of target arrangement. The target and Au catcher foils are cooled by low pressure N_2 and He gases, respectively.

2-mm$\times$45-mm column of Dowex-50, X12 cation exchange resin (7$-$10 μm) with hot (80°C) 0.5 M ammonium alpha-hydroxyisobutyrate at a pH of 3.71. The resulting actinide fractions were collected directly on Pt discs which were then dried and used for the subsequent analyses of alpha, fission, and gamma activity. The average time for complete processing was about 2 h and the chemical yield was determined to be (80$\pm$5)%.

In separate experiments, the No fraction was isolated by evaporating the eluate from the anion exchange column and redissolving it in 0.05 M HNO_3, followed by extraction with an equal volume of 0.4 M 2-diethylhexyl orthophosphoric acid in heptane to remove $+3$ actinides and lanthanides.[14] Nobelium and other $+2$ ions remained in the aqueous phase which was evaporated on a Pt disc for measurement of the alpha and spontaneous fission activity. The chemical yield was determined to be approximately 80% with ^{133}Ba tracer which was added before dissolution of the Au foil. The average separation time was about 20 min.

C. Measurements of radioactivity and data analysis

The Bk isotopes were assayed with a Ge(Li) gamma-ray spectrometer system. The other actinide samples were measured with a fission-alpha spectrometer system which used four Si(Au) surface-barrier detectors in conjunction with a computer based multichannel analyzer. Both spontaneous fission events and alpha spectra were recorded simultaneously. Typically, spectra were taken continuously for the first two weeks and then at appropriate intervals over a period of six months so that the half lives as well as alpha energies could be used for identification.

A simple computer code was used for integration of the alpha peaks. The gamma-ray spectra were analyzed by a procedure described previously.[15] Least-squares computer analyses of the growth and decay data were performed. Detector efficiencies and geometries were determined from measurements of NBS standard sources with estimated standard deviations of 3%.

The cross sections were calculated from the measured activities, the number of ^{248}Cm atoms in the target, the integrated beam intensities, and assuming that 100% of the products of interest recoiled out of the target and were caught in the Au catcher foils. Based on preliminary recoil

range experiments,[16] it appears that no more than 5% of these actinides remain in the target. The target was assumed to be uniform, with an estimated standard deviation of 10%. Corrections were made for growth and decay and beam intensity fluctuations during irradiation with an estimated standard deviation of 3%. Combining the uncertainties in the absolute detection efficiencies, target uniformity, current stability and integration, and chemical yield, by standard statistical methods, gives a standard deviation of 12% in the calculated absolute cross sections in addition to the statistical standard deviation of the counting data and decay analysis which is given in Tables I and II. These standard deviations can then be combined in the usual way, but we have not done this in order that

TABLE I. Cross sections for heavy actinides from bombardment of ^{248}Cm with ^{16}O and ^{18}O. (The standard deviation associated with the quoted absolute cross sections is estimated to be $\pm 12\%$ in addition to the s given in the tables which is based on the decay analysis. See discussion in text and footnote a.)

Nuclide	98-MeV ^{16}O Cross section (μb)	s[a] (μb)	s[a] %	97-MeV ^{18}O Cross section (μb)	s[a] (μb)	s[a] %	Decay mode	Measured radiation Energy (MeV)	Abundance[b]
Bk 245	11	5.0	45	0.4	0.2	50	EC-γ	0.253	0.31
246	81	15	19	6.7	0.6	9	EC-γ	0.799	0.61
248*m*	1600	150	9	120	11	9	β^--γ	0.551	0.05
250	200	11	6	1100	39	4	β^--γ	0.989	0.45
251	≥ 9.7[c]	4.0	41	≥ 24[c]	3	13	β^--γ	0.177	≤ 0.7[c]
Cf 246	6.2	0.4	7	0.04	0.02	50	α	6.76	1.00
248	500[d]	100	20	45[d]	3	7	α	6.26	1.00
249	1700	370	22	280	58	21	α	5.8	0.96
250	110[d]	230	21	850[d]	160	19	α	6.03	1.00
251				980	200	20	α	5.68	0.43
252	4.3	0.4	9	280	14	5	α	6.12	0.97
253	0.5	0.1	20	44	9	20	$\beta^- \rightarrow \alpha$	6.63[e]	1.00
254	0.005	0.003	60	3.5	0.3	9	SF		1.00
Es 251	38	5	8	0.1	0.05	50	α	6.49	0.005
252	21	7	33	5.7	1.1	19	α	6.63	0.73
253	7.4[f]	1.4	19	11[f]	2.1	19	α	6.63	0.98
254*m*	0.16	0.07	44	6.6	1.3	20	$\beta^- \rightarrow \alpha$	7.19[e]	1.00
255	0.006	0.003	50	0.2	0.1	50	$\beta^- \rightarrow \alpha$	7.01[e]	0.91
Fm 251	4.9	2.5	51	0.5	0.1	20	α	6.83	0.016
252	3.5	0.7	20	1.5	0.4	27	$\alpha \rightarrow \alpha$	6.26[e]	1.00
253	3.9	0.5	13	3.1	0.9	29	EC$\rightarrow \alpha$	6.63[e]	0.86
254	1.3[g]	0.2	15	2.8[g]	0.4	14	α	7.06–7.19	1.00
255	0.3	0.1	33	0.7	0.3	43	α	6.89–7.02[h]	0.99
256	0.02	0.005	25	0.3	0.06	20	SF		0.92
Md 256	0.005	0.003	60	0.010	0.005	50	EC$\rightarrow$SF[e]		0.83
No 259	≤ 0.001			0.030	0.01	33	α	7.44–7.67	0.77

[a]The statistical standard deviation, s, based on the decay analysis of the measured radiations, is given in both μb and % to show the relative precision of the measurements.

[b]Abundances calculated from data given in Ref. 17.

[c]No absolute gamma ray-intensitiy data are available, but our measurements show that the 0.177 level (Ref. 18) is deexcited by a 0.177-MeV γ transition (70%) and a 0.153 (30%) transition.

[d]Corrected for contribution from Bk parent.

[e]Radiation from daughter activity was measured.

[f]Corrected for contribution from decay of Cf parent.

[g]Corrected for contribution from decay of Es parent.

[h]Corrected for contribution from 7.04-MeV alpha group of ^{252}Fm.

TABLE II. Cross sections for heavy actinides from bombardment of ^{248}Cm with ^{20}Ne and ^{22}Ne. (The standard deviation associated with the quoted absolute cross sections is estimated to be ±12% in addition to the s given in the tables which is based on the decay analysis. See discussion in text and footnote a.)

	115-MeV ^{20}Ne			116-MeV ^{22}Ne			Measured radiation		
	Cross section	s[a]		Cross section	s[a]		Decay mode	Energy	
Nuclide	(μb)	(μb)	%	(μb)	(μb)	%		(MeV)	Abundance[b]
Bk 245	18	2	11				EC-γ	0.253	0.31
246	94	4	4	2.2	0.6	27	EC-γ	0.799	0.61
248*m*	695	11	2	61	5.3	9	β^--γ	0.551	0.05
250	7	2	29	61	1.6	3	β^--γ	0.989	0.45
251				≥0.31[c]	0.10	32	β^-−γ	0.177	≤0.7[c]
Cf 246	9.5	0.9	9	0.25	0.08	32	α	6.76	1.00
248	480[d]	67	14	40[d]	11	28	α	6.26	1.00
249	2670	890	33	360	70	19	α	5.8	0.96
250	2010[d]	190	9	720[d]	160	22	α	6.03	1.00
251	1020	510	50	1040	520	50	α	5.68	0.43
252	54	20	37	77	14	18	α	6.12	0.97
253	0.14	0.07	50	9.8	1.8	18	$\beta^-\rightarrow\alpha$	6.63[e]	1.00
254				0.57	0.10	18	SF		1.00
Es 251	100	10	10	0.59	0.30	51	α	6.49	0.005
252	42	14	33	2.2	0.4	18	α	6.63	0.73
253	5.3[f]	1.1	21	6.0[f]	0.7	12	α	6.63	0.98
254*m*	0.12	0.02	17	1.6	0.2	13	$\beta^-\rightarrow\alpha$	7.19[e]	1.00
255	0.016	0.006	38	0.090	0.008	9	$\beta^-\rightarrow\alpha$	7.01[e]	0.91
Fm 251				0.022	0.010	45	α	6.83	0.016
252	16	1.6	10	0.14	0.06	43	$\alpha\rightarrow\alpha$	6.26[e]	1.00
253	12	1.1	9	0.80	0.09	11	EC$\rightarrow\alpha$	6.63[e]	0.86
254	2.0[g]	0.2	10	3.1[g]	0.4	13	α	7.06−7.19	1.00
255	0.22	0.10	45	1.1	0.2	18	α	6.89−7.02[h]	0.99
256	0.06	0.04	67	0.15	0.02	13	SF		0.92
Md 256	0.024	0.012	50	<0.001			EC$\rightarrow$SF[e]		0.83
No 259	<0.001			<0.001			α	7.44−7.67	0.77

[a]The statistical standard deviation, s, based on the decay analysis of the measured radiations, is given in both μb and % to show the relative precision of the measurements.
[b]Abundances calculated from data given in Ref. 17.
[c]No absolute gamma ray-intensity data are available, but our measurements show that the 0.177 level (Ref. 18) is deexcited by a 0.177-MeV γ transition (70%) and a 0.153 (30%) transition. We have used 0.7 for the absolute intensity of the 0.177-MeV gamma ray, but this value is an upper limit since there may be decay via other levels or to the ground state.
[d]Corrected for contribution from Bk parent.
[e]Radiation from daughter activity was measured.
[f]Corrected for contribution from decay of Cf parent.
[g]Corrected for contribution from decay of Es parent.
[h]Corrected for contribution from 7.04-MeV alpha group of ^{252}Fm.

the relative magnitude of these can be seen. The relative cross sections are, of course, better known than the absolute values, but in some cases the statistical errors of the counting data constitute the major uncertainty.

III. RESULTS AND DISCUSSION

The cross sections for production of heavy actinides in reactions of (98±3)-MeV ^{16}O, (97±3)-MeV ^{18}O, (115±3)-MeV ^{20}Ne, and (116±3)-MeV

^{22}Ne on ^{248}Cm are shown in Tables I and II and plotted in Figs. 2 and 3, respectively. Corrections were made for the contribution to the measured activities of ^{248}Cf and ^{250}Cf from decay of ^{248}Bk and ^{250}Bk, for contribution to ^{253}Es from decay of ^{253}Cf, and to ^{254}Fm from decay of ^{254}Es during bombardment and prior to chemical separation. No corrections were made for possible contributions to Fm isotopes from electron capture decay of Md isotopes. We estimate the yields of these Md isotopes to be lower by at least an order of magnitude so the corrections should be very small.

The projectile center-of-mass energies for ^{16}O, ^{18}O, ^{20}Ne, and ^{22}Ne ions of 92, 90, 106, and 106 MeV, respectively, are close to the estimated reaction barriers of 89, 88, 108, and 108 MeV. The Coulomb barriers were calculated in the standard way using nuclear radii of $1.41\times M^{1/3}$ fm. Since ^{248}Cm atoms are deformed in the ground state, this calculation can be considered only as an approximation. It has been shown by other investigators[4] that reactions of ^{22}Ne with ^{249}Cf to make isotopes of Cf through Md proceed well below the Coulomb barriers estimated in this way. Our preliminary results[18] on the energy dependence for the reaction of ^{18}O with ^{248}Cm are consistent with their observations.

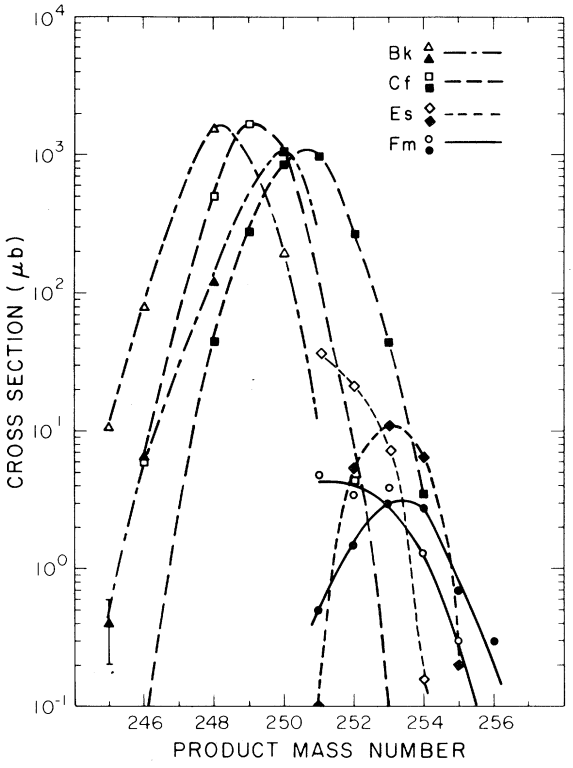

FIG. 2. Isotopic distributions measured for 98-MeV ^{16}O and 97-MeV ^{18}O bombardments of ^{248}Cm. ^{16}O data are open symbols; ^{18}O data are solid symbols.

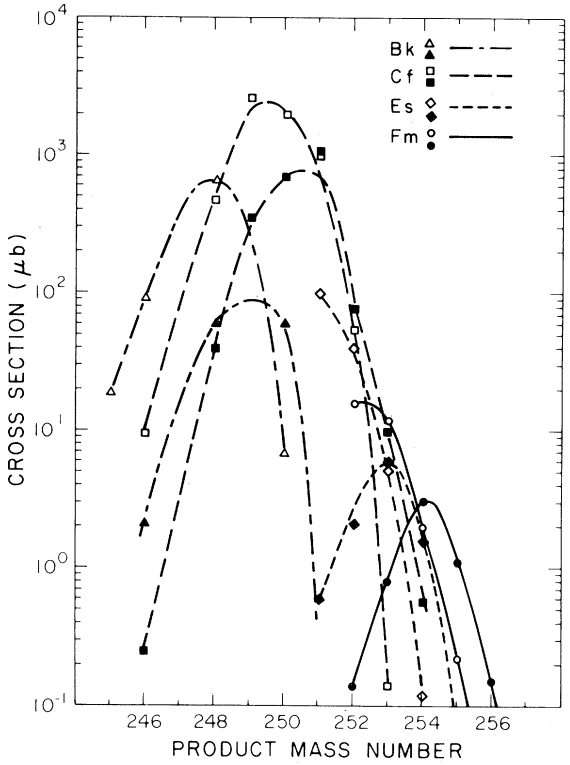

FIG. 3. Isotopic distributions measured for 115-MeV ^{20}Ne and 116-MeV ^{22}Ne bombardments of ^{248}Cm. ^{20}Ne data are open symbols; ^{22}Ne data are solid symbols.

The maxima of the isotopic yield distributions for each element are found at approximately the same mass numbers for the reactions with ^{16}O and ^{20}Ne and for those with ^{18}O and ^{22}Ne. A comparison of the results for ^{18}O with those for ^{16}O shows that the maxima of the mass-yield curves for each element are shifted to the neutron-rich side by about 2 mass units, reflecting the neutron excess of the projectile. A comparison of the ^{20}Ne and ^{22}Ne shows a similar result. Calculations[19] based on the ground state Q values for the reactions, the Coulomb barriers, and apportionment of the velocity (kinetic energy) of the projectile according to the fraction of mass transferred to the target show that this 2-neutron shift is to be expected for this target and these projectiles. For bombarding energies close to the Coulomb barrier as these are, the fraction of energy transferred to the target atoms is relatively unimportant.

The experimental yield curves appear to be rather symmetric and do not drop off sharply on the

neutron-rich side. The shapes of all the isotopic distributions are similar and have approximately the same full-width at half maximum of about 2.5 u. The shapes seem to be rather independent of the projectile, thus indicating that different reaction paths do not greatly influence the final distributions. The production cross sections and maxima of the actinide mass-yield distributions observed for ^{18}O and ^{22}Ne on ^{248}Cm are at least as favorable for production of neutron-rich heavy actinides as those measured for deeply inelastic transfer reaction products from irradiation[7,8] of ^{238}U targets with ^{86}Kr, ^{136}Xe, and ^{238}U and from irradiation[11,12] of ^{248}Cm targets with ^{86}Kr and ^{136}Xe. Detalied comparisons of maximum yields cannot be made until information concerning the excitation functions is obtained.

Several reaction channels may in principle lead to the same observed final reaction products. Unfortunately, it is not possible from these integral radiochemical experiments to indicate a definite reaction path. However, the identification of the final products is unambiguous. The observed cross sections are largest when only a small number of nucleons is transferred, as is the case with the isotopes of Bk and Cf. The cross sections decrease rapidly when the number of transferred particles increases, as can be seen for Es, Fm, Md, and No. Md and No can, of course, also be produced via compound nucleus reactions.

In summary, the peak yields are relatively large for all of these reactions, ranging from a few mb for Bk and Cf to a few μb for Fm. This is to be compared with peak yields of about a mb to a few tenths of a μb, for example, in the bombardment[11,12] of ^{248}Cm with ^{136}Xe. The production of so many rather neutron-rich isotopes between the target and compound nucleus with relatively high yields suggests a direct transfer-type reaction in which the product nucleus is formed with low enough excitation energy so that it is not all lost to prompt fission. These "transfer-type" reactions appear to offer a method for producing new neutron-rich isotopes of known elements and, if properly chosen, perhaps even new heavier elements.

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