Excitation functions for production of heavy actinides from interactions of ^{40}Ca and ^{48}Ca ions with ^{248}Cm

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Excitation functions have been measured for production of isotopes of Bk through Fm in bombardments of ^{248}Cm with 234- to 294-MeV ^{40}Ca ions and with 239- to 318-MeV ^{48}Ca ions. The maxima of the isotopic distributions for these elements occur at only 2 to 3 mass numbers larger for ^{48}Ca than for ^{40}Ca reactions. The shapes of the distributions and the half-widths of about 2.5 mass numbers are quite similar to those observed previously for reactions of ^{16}O, ^{18}O, ^{20}Ne, and ^{22}Ne with ^{248}Cm. In general, the excitation functions for ^{40}Ca show maxima near the Coulomb barrier while those for ^{48}Ca are about 20 MeV above the barrier. The cross sections decrease rather slowly with increasing projectile energy over the energy range studied, indicating that the additional projectile energy is not manifested as excitation energy of these actinide products.

I. INTRODUCTION

We have previously reported[1,2] on our measurements of cross sections and excitation functions for production of heavy actinide isotopes from the interaction of light heavy ions with ^{248}Cm and ^{249}Cf targets. In particular we used the isotope pairs ^{16}O-^{18}O and ^{20}Ne-^{22}Ne in order to assess the influence of the extra pair of neutrons in each case. We found that the neutron-rich projectiles such as ^{18}O and ^{22}Ne enhanced the formation of the neutron-rich actinide products and that reactions using these lighter heavy ions gave production cross sections for the heavy actinides which were at least as large as those obtained using the heaviest projectile so far available, that is, ^{238}U. For the measured actinide products from ^{18}O reactions with ^{248}Cm, about half of the projectile kinetic energy appeared[2] to be transferred to the targetlike product, but we were unable to infer any correlation between the number of nucleons transferred and the energy transferred. We have undertaken further experiments of this type using ^{40}Ca and ^{48}Ca projectiles with ^{248}Cm targets in order to see if the very neutron-rich projectile ^{48}Ca would still further enhance the yields of neutron-rich actinides and to determine what effect the eight fewer neutrons in ^{40}Ca would have on the mass distributions. Experiments were conducted with ^{48}Ca at the SuperHILAC at Lawrence Berkeley Laboratory (LBL) and with both ^{40}Ca and ^{48}Ca projectiles at the UNILAC at the Gesellschaft für Schwerionenforschung (GSI) in Germany at energies from just above the Coulomb barrier to about 60 MeV above the barrier for ^{40}Ca, and to 90 MeV above the barrier for ^{48}Ca. Products with larger and smaller Z than the target were measured in these experiments. In this paper we report on the production of Bk, Cf, Es, and Fm isotopes only. The production of lower Z nuclides will be discussed in a later publication.

II. EXPERIMENTAL PROCEDURES

A. Bombarding conditions and target arrangement

Irradiations of ^{248}Cm with ^{48}Ca ions were performed at the SuperHILAC at LBL with incident energies on target of 239, 263, and 288 MeV and at the UNILAC at the GSI with 263 and 318 MeV. Irradiations with 234-, 259-, and 294-MeV ^{40}Ca ions were also conducted at the UNILAC. All projectile energies are quoted in the laboratory system. Two different targets containing 1.5 and 1.7 mg/cm^2 of ^{248}Cm in the form of Cm_2O_3 (96.5% ^{248}Cm; 3.5% ^{246}Cm), prepared by stepwise electrodeposition with a diameter of 9 mm on 2.4 mg/cm^2 Be foil, were used.

A schematic diagram of the target arrangement used at

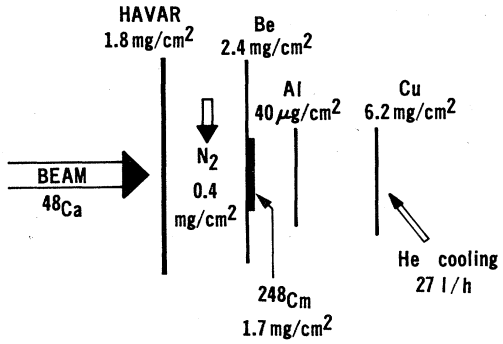

FIG. 1. Schematic drawing of target arrangement used at the SuperHILAC.

the SuperHILAC is shown in Fig. 1. The entrance window and target were cooled with nitrogen gas and the catcher foil with helium gas. A similar arrangement was used at the UNILAC except that the entrance window was 3.9 mg/cm^2 molybdenum. A 40 μg/cm^2 aluminum foil was placed over the Cm target to prevent target atoms knocked out by the beam from reaching the catcher foil. Copper catcher foils of 6 to 10 mg/cm^2 were used to collect recoiling products within an acceptance angle of about 60°. We have assumed that all actinide products recoiled out of the target and were collected. Corrections have been made for the total measured energy losses of about 75 MeV for ^{40}Ca and 97 MeV for ^{48}Ca ions prior to entry into the Cm target. Energy losses in the target were 13 to 16 MeV and the energy range is indicated in the tables and figures.

B. Radiochemical separations

The copper catcher foils were dissolved in concentrated HCl containing a few drops of HNO_3 and the actinides were co-precipitated on ferric hydroxide by the addition of ammonia to complex the copper, leaving it in solution. This step was repeated as necessary to remove the copper. The final precipitate was dissolved in 9 *M* HCl and extracted with di-isopropyl ether to remove the iron. In the later experiments at GSI a modified procedure was used.

TABLE I. Cross sections for the production of heavy actinides in the bombardment of ^{248}Cm with ^{40}Ca of different energies.

		(221–234) MeV[a,b]		(246–259) MeV[a,b]		(281–294) MeV[a,b]	
		Cross section (μb)	s[c] (%)	Cross section (μb)	s[c] (%)	Cross section (μb)	s[c] (%)
Bk	244[d]	64.7	20	60.0	5	47.3	9
	245	675	12	605	5		
	246	2020	4	1900	2		
	248*m*	5470	6	4575	3		
	250	250	8	330	8		
Cf	245	84.7	14	44.2	14		
	246	215	5	185	19	135	2
	248[e]	1800	5	1585	2	1375	3
	250[e,f]	320	63	260	53	175	80
	252	50	82	45	65	40	80
Es	249[g]	150	7			80	7
	250[h]	190	26	145	20	150	10
	252	12.8	23	10.9	37	6.6	30
	253	2.6	20	2.5	40	1.8	40
	254*m*	0.33	33	0.30	20	0.25	32
Fm	250			2.8	14		
	251	7.0	80	5.2	21		
	252	6.85	2	5.2	8	3.3	12
	254	0.39	30	0.31	6	0.22	9
	256	0.004	50	0.007	30		

[a]Range of projectile energy (laboratory system) in target.
[b]Chemical yields were measured to be 11%, 27%, and 21% for the 234-, 259-, and 294-MeV bombardments, respectively.
[c]The standard deviation associated with the quoted absolute cross sections is estimated to be ±12%, in addition to the statistical standard deviation s given in the table, which is based on the analysis of the decay data. The same radiations and abundances were used as in Ref. 1. It was assumed that all actinide products recoiled out of the target and were collected.
[d]An intensity of 0.5 was assumed for the measured 892-keV gamma ray.
[e]Corrected for contribution from decay of Bk parent.
[f]Corrected for contribution from decay of Es parent.
[g]An intensity of 0.40 was used for the measured 379-keV gamma ray.
[h]Only the 8.6-h isomer was measured and a ratio of 0.5 for the yield of metastable to ground state isomers was used to calculate total cross section.

The copper catcher foils were dissolved in 8 *M* HCl containing KNO_2, H_2O_2, and ^{243}Am as a tracer for the trivalent actinides. Th, U, and Pu were removed by extraction into hydrogen di(2-ethylhexyl)orthophosphate (HDEHP) in heptane. The aqueous phase containing the lanthanides and trivalent actinides was then boiled to remove excess peroxide. The resulting solution (8–9 *M* in HCl) was passed through an anion exchange resin column to remove Cu and other reaction products and the actinides were eluted with 9 *M* HCl. They were separated from alkali and alkaline earth elements in 0.2 *M* HCl by absorption and elution from a column consisting of a polymer containing HDEHP. (The material used was Levextrel; it is currently available from Bayer-Leverkusen, under the trade name Lewatit.) They were separated from lanthanides by elution from a cation exchange resin column[3] with 20% ethanol saturated with HCl gas. Final separation of the individual actinides was accomplished by elution from a cation column with 0.5 *M* ammonium alpha-hydroxyisobutyrate at pH 3.71 as described previously.[1] After elution of Md, Fm, Es, and Cf, the pH was changed to 3.89 to elute Bk, Cm, and Am. The final fractions were evaporated to dryness and the resulting samples were counted for alpha, spontaneous fission, and gamma activities, and data analysis was performed as described before.[1]

Chemical yields for the more extensive actinide separation procedures used at GSI were determined from ^{243}Am tracer added to the initial dissolving solutions. (See Tables I and II.) No tracer was used in the experiments at LBL, but the chemical yields for the actinide separations were estimated to be 75% for the runs with 239- and 263-MeV ^{48}Ca ions for which a procedure similar to that described[1] earlier was used. Experiments with 263-MeV ^{48}Ca ions were performed at both GSI and LBL, but because the chemical yield was measured only in the GSI experiment, only those results are shown in Table II. The duplicate experiments for 263 MeV agree within the statistical standard deviations if a chemical yield of 75% is used for the LBL experiment. A yield of 50 ± 10% was

TABLE II. Cross sections for the production of heavy actinides in the bombardment of ^{248}Cm with ^{48}Ca of different energies.

	(223–239) MeV[a,b]		(247–263) MeV[a,c]		(272–288) MeV[a,d]		(304–318) MeV[a,c]	
Nuclide	Cross section (μb)	s[e] (%)	Cross section (μb)	s[e] (%)	Cross section (μb)	s[e] (%)	Cross section (μb)	s[e] (%)
Bk 245	12	20	40	40	67	20	110	30
246	73	15	360	10	480	15	480	10
248*m*	730	20	2900	6	2680	15	3180	20
250	690	10	2520	5	2920	10	2220	5
Cf 246	0.25	5	1.8	2	1.0	20	0.69	15
248[f]	100	9	260	2	210	6	180	10
250[f,g]	415	20	2380	7	1935	2	1250	15
252	51	20	225	4	220	15	145	15
253	0.81	20	12	20	4.0	15	2.5	40
254	0.32	30	1.5	30	1.0	25	0.7	40
Es 250[h]			6.6	40			5.4	45
252	9.5	7	30	15	24	5	12.7	20
253	4.1	20	10	10	7.8	15	5.1	40
254*m*	0.74	30	2.0	15	1.4	15	0.85	6
Fm 252	0.02	40	0.11	35	0.06	20	0.05	60
254	0.28	8	0.81	5	0.71	8	0.42	10
255	0.21	7	0.9	30	0.62	10	0.38	15
256	0.06	30	0.24	20	0.14	15		

[a]Range of projectile energy (laboratory system) in target.

[b]Cross sections from irradiation at LBL. Chemical yield assumed to be 75%, the same as measured previously (Ref. 1) for a comparable procedure.

[c]Cross sections from irradiation at GSI; chemical yields were measured to be 30% and 27%, respectively. It was assumed that all actinide products recoiled out of the target and were collected.

[d]A chemical yield of 50 ± 10% was estimated for this untraced experiment at LBL as explained in the text.

[e]The standard deviation associated with the quoted absolute cross sections is estimated to be ±12%, in addition to the statistical standard deviation s given in the table, which is based on the analysis of the decay data. The same radiations and abundances were used as in Ref. 1.

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

[g]Corrected for contribution from decay of Es parent. Es contribution estimated for irradiations for which Es yields were not measured.

[h]Both isomers were measured. An abundance of 0.136 was used for the 989-keV gamma transition from the 2.2-h isomer and 0.223 for the 303-keV gamma transition from the 8.6-h isomer.

estimated for the 288-MeV ^{48}Ca experiment at LBL in which the actinides were co-precipitated on $Fe(OH)_3$. This yield is based on chemical yield determinations of the procedure used in that experiment.

III. RESULTS AND DISCUSSION

The results of the experiments for bombardment of ^{248}Cm with ^{40}Ca and ^{48}Ca projectiles are given in Tables I and II, respectively. Production yields for Bk through Fm isotopes are plotted in Fig. 2 for 246- to 259-MeV ^{40}Ca and 247- to 263-MeV ^{48}Ca ions. No yields for heavier elements were obtained except for an upper limit of 0.005 μb (95% confidence level) for the yield of ^{256}Md for 247- to 263-MeV ^{48}Ca ions. The cross sections for most of the measured isotopes are near their maxima for these energies, which are about 1.1 of the calculated Coulomb barriers of 235 and 236 MeV, respectively.

The positions of the maxima of the mass yield curves for the reactions with ^{40}Ca are approximately at masses 247, 248, 250, and 251 to 252 for Bk, Cf, Es, and Fm, respectively. Those for the reactions with ^{48}Ca are at about masses 249, 250, 252, and 254 to 255, respectively. The yields at the maxima of the isotopic distributions for Bk and Cf isotopes are not significantly different for ^{40}Ca and ^{48}Ca, but for Es and Fm they are about a factor of 5 larger for ^{40}Ca than ^{48}Ca. However, the yields for the most neutron-rich isotopes are much larger for the ^{48}Ca than for ^{40}Ca reactions. The differences between the positions of the maxima of the mass-yield curves for Bk, Cf, and Es are about 2 mass numbers. However, for Fm they are about 3 mass numbers larger for reactions with ^{48}Ca than with ^{40}Ca. Thus the eight-neutron excess of ^{48}Ca over ^{40}Ca is certainly not reflected in these products. The effect of the 3.5% ^{246}Cm in the target would be to increase the observed yields of neutron-deficient isotopes, but no significant effect can be seen in these data. The shapes of all of the isotopic distributions are quite similar and rather symmetric with a full-width at half maximum (FWHM) of about 2.5 mass numbers. This width is similar to those of 2 to 2.5 mass numbers reported previously[1,2] for light ion reactions with ^{248}Cm and only slightly smaller than that of about 3 mass numbers reported[4] for ^{238}U reactions with ^{248}Cm. These relatively narrow widths indicate[5] that heavy actinide nuclides produced in heavy ion transfer reactions only survive in collisions where the targetlike fragments have low excitation energies.

The excitation functions for the production of isotopes of Bk, Cf, Es, and Fm from ^{40}Ca bombardments of ^{248}Cm are given in Figs. 3—5. In general, they are all similar in shape, showing the highest cross sections for the near-barrier energy and decreasing slowly with increasing projectile energy. Thus the mass-yield curves for the individual elements and their widths change very little over the energy range investigated. Even at about 60 MeV above the barrier this decrease in cross section, with the possible exception of that for ^{245}Cf, is no more than a factor of 2. Since the fission barriers are only 5 to 6 MeV, it is apparent that this extra projectile kinetic energy above the reaction barrier is not manifested as excitation energy of these actinide products. The cross sections for most of these products are already near their maxima at the Coulomb barrier, consistent with the positive reaction energies calculated[6] for these nuclides based on ground state

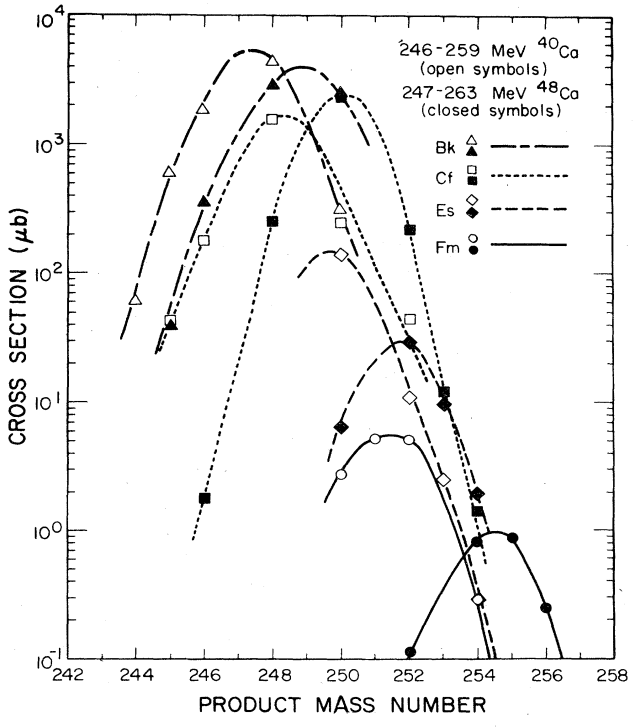

FIG. 2. Comparison of heavy actinide yields from ^{40}Ca and ^{48}Ca reactions with ^{248}Cm. Projectile energies in the target are from 5% to 10% above the Coulomb barriers.

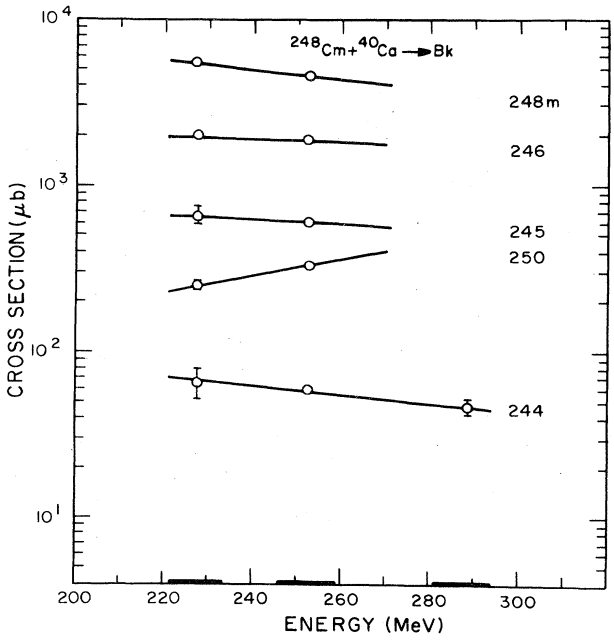

FIG. 3. Excitation functions for Bk isotopes produced in the bombardment of ^{248}Cm with ^{40}Ca.

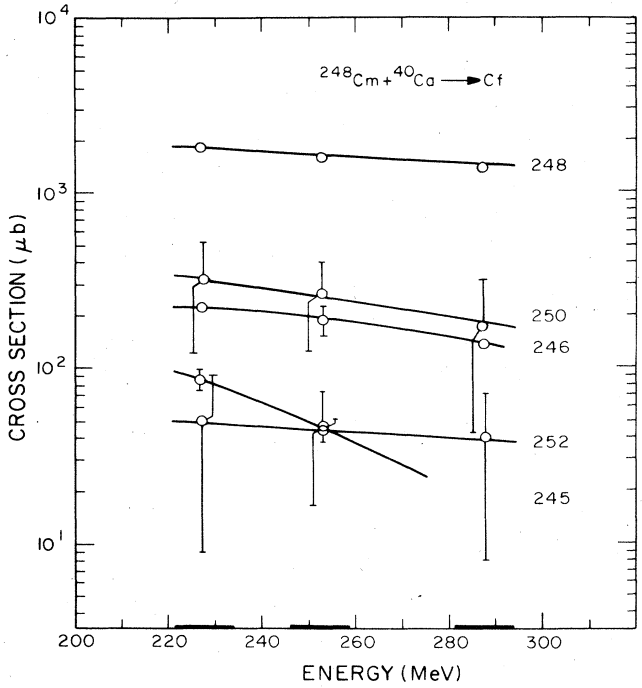

FIG. 4. Excitation functions for Cf isotopes produced in the bombardment of ^{248}Cm with ^{40}Ca.

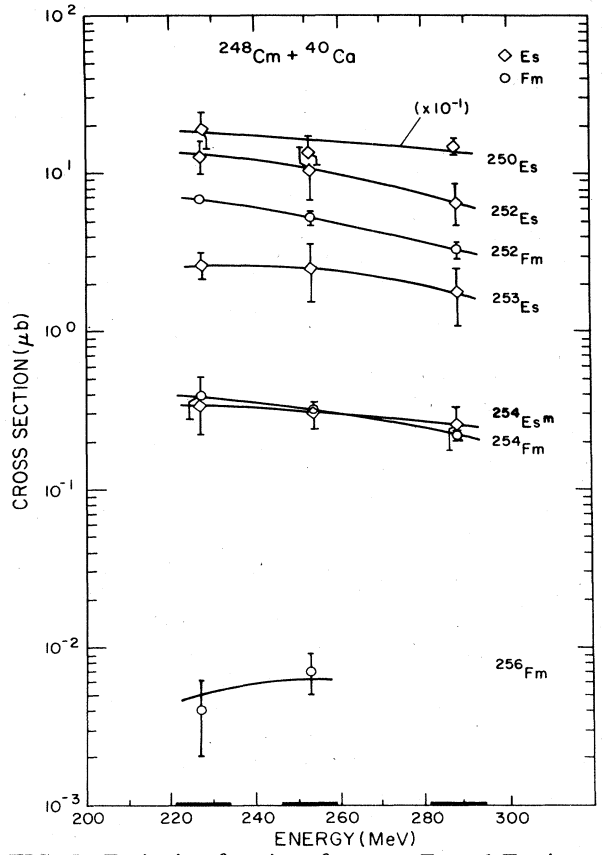

FIG. 5. Excitation functions for some Es and Fm isotopes produced in the bombardment of ^{248}Cm with ^{40}Ca. (Data for ^{249}Es and ^{251}Fm are not plotted since their yields fall nearly on top of adjacent isotopes.)

Q values and the differences between the ingoing and outgoing Coulomb barriers, calculated assuming touching spherical nuclei.

The excitation functions for the production of isotopes of Bk, Cf, Es, and Fm for ^{48}Ca bombardments of ^{248}Cm are given in Figs. 6–8. In general, the maxima are about 20 MeV above the Coulomb barrier, consistent with the calculated reaction energies which are negative by around 10 MeV. The cross sections for Bk do not decrease over the energy range investigated and those for Cf, Es, and Fm do not drop off rapidly even at energies 60 MeV above the maxima, indicating that these actinide products were not formed with high excitation energies.

It is also of interest to compare the cross sections for actinide production from other neutron-rich projectiles, e.g., ^{18}O ($N/Z=1.25$), ^{22}Ne ($N/Z=1.20$), ^{136}Xe ($N/Z=1.52$), and ^{238}U ($N/Z=1.59$), with ^{48}Ca ($N/Z=1.40$) and to heavy actinide targets. Data for these systems indicate that the net transfer of the same numbers of protons and neutrons proceeds with similar cross sections provided the reaction energy is not negative. In general, the yields drop off rather rapidly with the total number of transferred nucleons. Maximum cross sections for transfer of various mass Be fragments from ^{18}O, ^{48}Ca, and ^{238}U to ^{248}Cm to produce Fm isotopes and from ^{136}Xe to ^{238}U to produce Cm isotopes are shown in Fig. 9. These may actually be transfers of four protons from projectile to target with neutrons being transferred either way. (Similar curves can be constructed for H, He, and Li transfers.) It is somewhat surprising to note that these cross sections are of the same order of magnitude for projectiles ranging from ^{18}O to ^{238}U. These similarities may support the general idea of a binary transfer mechanism which leads to actinide products with very low excitation energy, i.e., "cold" nuclei which are not destroyed by prompt fission or particle emission, and are, therefore, observed in these studies.

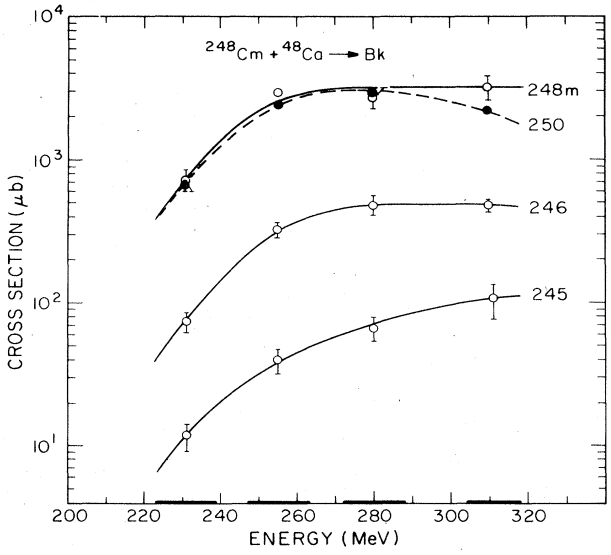

FIG. 6. Excitation functions for Bk isotopes produced in the bombardment of ^{248}Cm with ^{48}Ca.

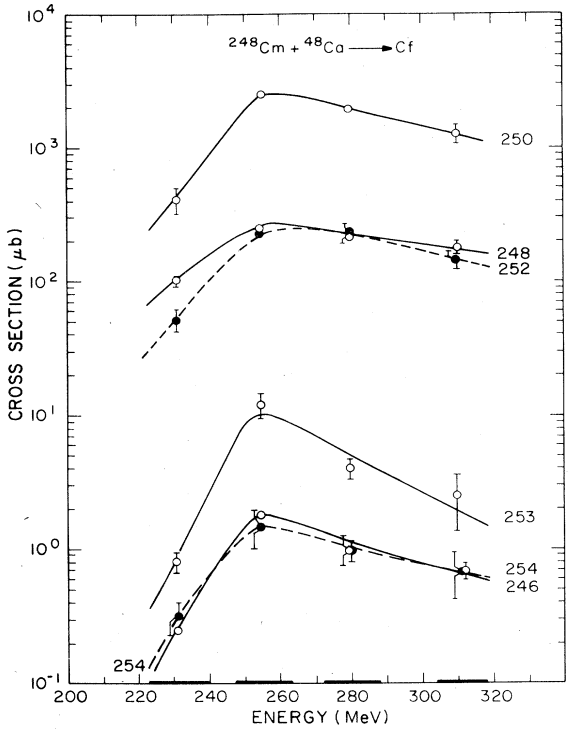

FIG. 7. Excitation functions for Cf isotopes produced in the bombardment of ^{248}Cm with ^{48}Ca.

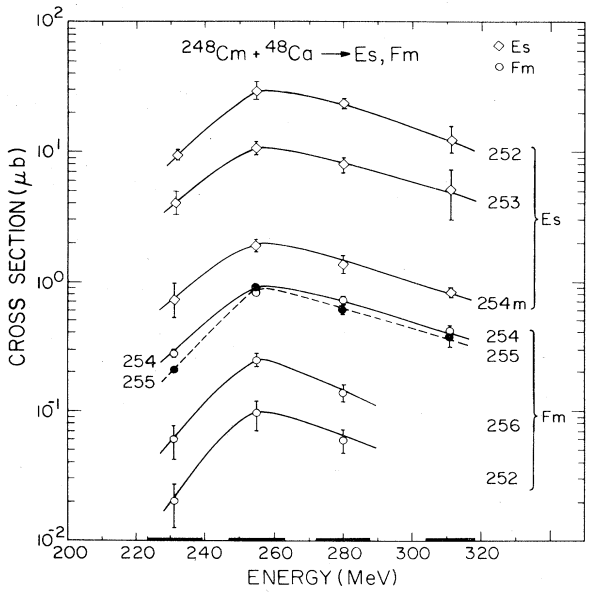

FIG. 8. Excitation functions for Es and Fm isotopes produced in the bombardment of ^{248}Cm with ^{48}Ca.

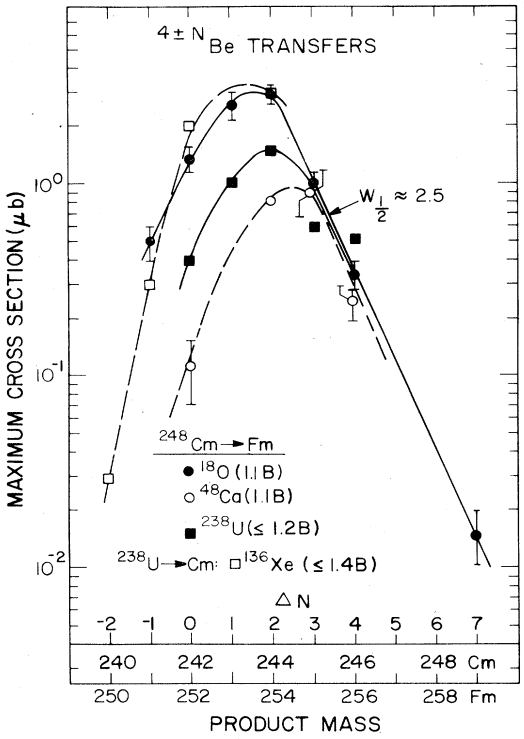

FIG. 9. Cross sections for various mass Be transfers from ^{18}O, ^{48}Ca, and ^{238}U projectiles to ^{248}Cm to produce Fm isotopes and from ^{136}Xe projectiles to ^{238}U to produce Cm isotopes. Values in parentheses give the ratio of projectile energy to Coulomb barrier. (Data from Refs. 1, 2, 4, and 7.)

An alternative mechanism leading to rather small changes in cross section as the bombarding energy is increased has been suggested[4,5,7] for heavier projectiles such as ^{136}Xe or ^{238}U. It has been noted that there is a broad distribution of dissipated energies associated with these damped or partially damped collisions. The surviving actinides may be those products which are associated with the smaller dissipated energies. In this description, the expected increase in primary production cross sections due to the heavier projectiles is largely offset by the decrease in survivability due to sequential fission which is caused by less favorable Q_{gg} values. Changes in the residual cross sections for neighboring actinides by factors of 5 to 10 may be associated with the nuclear structures of the colliding nuclei and details of the N/Z equilibration in the various systems.[5,7] The observation of relatively high yields of neutron-rich actinide products in these reactions of neutron-rich projectiles with heavy actinide targets is quite encouraging for production of new neutron-rich heavy element isotopes for study.

IV. SUMMARY

In summary, we can draw the following conclusions from the results presented in this paper.

(a) Cross sections are largest for transfers of a few nucleons and decrease rapidly with an increase in the total number of transferred nucleons.

(b) A shift of only 2 to 3 mass numbers in the mass-yield curves is observed between ^{40}Ca and ^{48}Ca reactions. Thus, the eight-neutron difference between the two projectiles is only partially reflected in the products.

(c) The shapes of the isotopic distributions are quite similar and rather symmetric (FWHM$\approx$2.5 mass numbers).

(d) Reactions with ^{40}Ca are favorable for the production of neutron-deficient isotopes, those with ^{48}Ca for neutron-rich products.

(e) "Cold" heavy products are observed in the reactions with ^{40}Ca and ^{48}Ca in the energy range investigated.

(f) The cross sections for the reactions with ^{40}Ca are near their maxima at the Coulomb barrier, consistent with calculated positive reaction energies.

(g) The excitation functions for ^{48}Ca show maxima about 20 MeV above the Coulomb barrier, but decrease only very slowly with increasing projectile energy.

(h) Cross sections for the production of actinides from transfer of the same nucleons are very similar for projectiles ranging from ^{18}O to ^{238}U.

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