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Excitation functions for production of heavy actinides from interactions of ^{16}O with ^{249}Cf

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ABSTRACT

Excitation functions have been measured for the production of isotopes of Bk through Fm in bombardments of ^{249}Cf with 90- to 150-MeV ^{16}O ions. A comparison of the maxima of the mass-yield curves measured in this experiment with those for the reactions of ^{18}O ions with ^{249}Cf shows different shifts from those that have been measured for reactions of the $^{16,18}O$ and $^{20,22}Ne$ ion pairs with ^{248}Cm. However, the shifts appear similar to those recently measured for reactions of these ion pairs with ^{254}Es.

NUCLEAR REACTIONS $^{249}Cf(^{16}O,X)$, $E(^{16}O)=90, 96, 106, 115, 122, 139, 150$ MeV; measured cross sections and isotopic distributions for Z=97-100.

This work was supported by the Director, Office of Energy Research, Division of Nuclear Physics of the Office of High Energy and Nuclear Physics of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

I. INTRODUCTION

In an earlier publication[1], we reported on the production of actinides with atomic number greater than the target from interactions of ^{16}O, ^{18}O and ^{20}Ne, ^{22}Ne projectiles with ^{248}Cm. These studies showed that neutron-rich projectiles enhance the formation of neutron-rich actinide isotopes. The maxima of the mass-yield curves were about two mass numbers heavier for reactions with the heavier ion of each projectile pair. In these transfer-type reactions product nuclei are formed with low excitation energy and, therefore, are able to survive prompt fission and/or particle emission. The production cross sections were found to decrease with increasing Z of the products and ranged from a few mb for Bk and Cf isotopes to a few μb for Es and Fm isotopes; they were of the order of nanobarns for Md and No isotopes. We have also investigated[2] the excitation functions for the interactions of ^{18}O projectiles with ^{248}Cm and ^{249}Cf targets. The cross sections for transfer of the same projectile fragments were found to be similar for ^{18}O reactions with both targets. The formation of neutron-rich isotopes was enhanced for reactions with the more neutron-rich ^{248}Cm target. The maxima of the measured excitation functions were consistent with calculated product excitation energies based on reaction Q values and Coulomb barriers. In general, products with lower excitation energy reached their maximum cross sections at higher projectile energies because fission and/or neutron evaporation does not occur until the product excitation energy exceeds 5 to 6 MeV.

In the present study, we have measured the excitation functions for reactions of ^{16}O projectiles with ^{249}Cf to compare with those for ^{18}O projectiles. We wished to ascertain whether the mass-yield curves showed the two mass number difference observed for the ^{248}Cm target with ^{16}O and

^{18}O projectiles. We also wished to ascertain if the shapes of the excitation functions were again consistent with the calculated excitation energies and to estimate how much of the excess kinetic energy of the projectile appears as excitation energy of the heavy product.

II. EXPERIMENTAL

The target arrangement was identical to that used in our previous work.[1,2] A 0.334 mg/cm^2 target of ^{249}Cf ($\approx$100% ^{249}Cf; $\approx 10^{-3}$ wt. % ^{252}Cf) as Cf_2O_3 electroplated with a diameter of 6.5 mm on 2.75-mg/cm^2 Be foil was prepared and used. The ^{249}Cf target was irradiated at the Lawrence Berkeley Laboratory 88-Inch Cyclotron with 90-, 96-, 106- 115-, 122-, 139-, and 150-MeV ^{16}O ions. (All quoted energies are the average energy in the target in the laboratory system after correction for a total energy loss of 11 to 15 MeV in the Havar window, nitrogen cooling gas, and Be target backing. The energy loss in the Cf_2O_3 target was always less than 0.6 MeV.) The recoiling reaction products were stopped in 1.4 mg/cm^2 Au catcher foils.

The radiochemical separations were identical to those described earlier[1], except that after the AG-1 anion exchange resin column an MP-50 cation exchange resin column of 2-mm diameter and 4-cm length was used to effect a group separation of the trivalent actinides from the lanthanides by eluting with saturated HCl. Individual actinides were then separated by elution from a cation exchange resin column with ammonium alpha-hydroxyisobutyrate as described earlier[1]. The chemical yield for trivalent actinides was determined for each run by addition of ^{241}Am tracer.

The Bk fractions were assayed with a Ge(Li) gamma-ray spectrometer system. The Fm and Md fractions were measured with a Si(Au) spectrometer system which recorded spontaneous fission events and alpha spectra simultaneously. The Cf and Es fractions were first assayed with the Ge(Li) system to obtain data on the gamma-emitting isotopes and then later were moved to the fission-alpha system. All samples were counted continuously for two weeks and then at appropriate intervals for the next four weeks so that half lives could be determined and positive isotope identification could be achieved. The decay modes, energies, and abundances of the radiations measured for each isotope are listed in Table I.

The gamma-ray spectra were analyzed for major peaks using the SAMPO code[3] on the Lawrence Berkeley Laboratory CDC-7600 computer. Minor peaks were integrated by hand. The alpha spectra were analyzed by integrating the desired peaks. Least-squares computer analyses of the activities were performed to obtain disintegration rates at the end of bombardment. As described in Ref. 1, the cross-sections were calculated from the measured activities, chemical yields, detector efficiencies, the ^{249}Cf atoms in the target, and the integrated beam intensities, assuming that 100% of the measured products recoiled out of the target and were caught in the Au catcher foils. As before, we estimate a standard deviation of 12% in the calculated absolute cross sections in addition to the statistical standard deviations quoted in Table II which are due only to the counting data and decay analysis. Due to transfer problems with the yield tracer, the standard deviation was estimated to be 22% rather than 12% for the 150-MeV measurements.

III. RESULTS AND DISCUSSION

The results of the cross-section measurements are summarized in Table II and plotted in Figures 1-4. The ^{244}Bk cross sections are lower limits only since the absolute gamma-ray abundances are not known and the cross sections were obtained by assuming 100% abundance for the 217.6-keV gamma ray. Cross sections were not obtained for ^{247}Cf from the 106- and 115-MeV runs due to problems with the Ge(Li) spectrometer system during these runs.

The cross sections, as expected, increase with bombarding energy as the impact parameter for Rutherford trajectories which can result in collisions increases. At some point, the increased probability of fission or neutron emission as more excitation energy is deposited in the target-like fragment should cause a decrease in the observed cross sections. The observed maxima in the excitation functions would, therefore, be expected to occur at higher energies for reactions with more negative excitation energies. This results in excitation functions (see Figures 1-3) comparable to those measured for the same transfers for ^{18}O reactions with ^{248}Cm and ^{249}Cf.

The energies, E_M, at which maxima in the excitation functions for the Bk, Es, and Fm isotopes occur, were obtained by inspection of Figures 1, 2, and 3, and are listed in Table III together with the excitation energies, E_X, calculated assuming simple binary transfer reactions[4]. These calculations are based on the ground-state Q values for the reactions, and the initial and final Coulomb barriers. In general, the isotopes with positive excitation energies should have the maxima in their excitation functions near the estimated Coulomb barrier for the reaction (about 96 MeV) since the fission barriers[5] are only about 5.5 MeV. However, the maxima of the excitation functions for isotopes with negative excitation energies should occur at correspondingly higher energies above the Coulomb barrier.

The excitation functions for the Bk isotopes are shown in Fig. 1. Their maxima are all above the Coulomb barrier, consistent with their negative calculated excitation energies. However, the maximum for ^{244}Bk is much higher than might be expected on this basis. Perhaps a mechanism other than simple binary transfer is occurring. As was observed previously[2] for reactions of ^{249}Cf with ^{18}O, the cross sections for formation of products with one proton less than the target are about a factor of 100 smaller than the cross sections for products with one proton more than the target.

Excitation functions were measured for production of Cf isotopes of masses 247 and 248. Yields are high (tens of mb) for ^{248}Cf which can be produced by the (n, 2n) reaction as well as by removal of a neutron. Yields up to mb's were observed for ^{247}Cf which can be produced by removal of 2 neutrons from the target. Information on ^{252}Cf and ^{253}Cf is not reported because of the unknown corrections required for the $\simeq 10^{-3}$ wt. % of ^{252}Cf in the target.

Comparison of E_M and E_X (Table III) for the Es isotopes shows that ^{250}Es has a maximum near the Coulomb barrier, consistent with its positive excitation energy. The Es isotopes with masses 248, 249, and 251 have maxima from 5 to 10 MeV above the Coulomb barrier consistent with their E_X's which are negative by a few MeV. Even though E_X for ^{253}Es is -23 MeV, its E_M is no higher than for the other isotopes. However, as shown in Fig. 2, its production cross sections are orders of magnitude smaller (μb's) and perhaps some mechanism other than binary transfer has become important.

Similar comparisons can be made for the Fm isotopes, and the E_M's are generally consistent with the calculated excitation energies, except for ^{256}Fm for which E_X is -31 MeV. Again, the production cross sections are extremely small, only tens of nanobarns.

Figure 4 compares the mass-yield curves for Bk, Es, and Fm obtained from the maxima of the measured excitation functions for reactions of ^{16}O with ^{249}Cf with previously measured[2] mass-yield curves for ^{18}O with ^{249}Cf. The full-widths at half maximum (FWHM's) of 2, 1 1/2, and 2 mass numbers for

Bk, Es and Fm, respectively, for the mass-yield curves for ^{16}O reactions, appear to be similar to the FWHM's of about 2-mass numbers observed for ^{18}O reactions. Similar comparisons of mass-yield curves have been made[1,6] for the reactions of the $^{16,18}O$, $^{20,22}Ne$, and $^{40,48}Ca$ ion pairs with ^{248}Cm where the FWHM's were about 2.5 mass numbers. In the studies on ^{248}Cm, a shift of two mass numbers toward heavier nuclides was observed for the heavier ion of the lighter two pairs, reflecting their 2-neutron excess. A shift of only two to three mass numbers was observed[6] for the $^{40,48}Ca$ reactions which only partially reflected the neutron difference of the projectiles. For reactions of the $^{16,18}O$ ion pair with ^{249}Cf, the Bk mass-yield curves show the expected two mass-number shift, but the Es ($\Delta Z=+1$) curves show only a 1/2 to 1 mass-number shift and the Fm ($\Delta Z=+2$) curves show only a 1 to 1 1/2 mass-number shift. Recent studies performed by Schädel et al.[7] of reactions of $^{16,18}O$ and $^{20,22}Ne$ projectiles with ^{254}Es also show smaller shifts in the maxima of the mass-yield curves. Their results indicate that the peak position in the mass-yield curves for Fm ($\Delta Z=+1$) and Md ($\Delta Z=+2$) is shifted by 0 and 1/2 mass numbers, respectively, which does not reflect the difference in neutron number of the projectiles. However, the two-neutron number difference is observed for the No ($\Delta Z=+3$) and Lr ($\Delta Z=+4$) mass-yield curves.[7]

IV. SUMMARY

In summary, we can draw the following conclusions from our results.

(a) The measured cross sections for production of isotopes of Bk through Fm from the $^{16}O + ^{249}Cf$ reaction are of the same order of magnitude as for similar transfer reactions of light-heavy ions on other actinide targets.

(b) The shapes of the excitation functions are similar to those measured earlier for $^{18}O + ^{249}Cf$ and $^{16,18}O + ^{248}Cm$.

(c) The cross sections decrease much more rapidly with projectile energy than those measured[6] for $^{40,48}Ca$ reactions with ^{248}Cm, but appear to be similar if projectile energy per nucleon is considered.

(d) The FWHM's of about 2 mass numbers for the mass-yield distributions for reactions of $^{16}O + ^{249}Cf$ are similar to those for ^{18}O reactions with ^{249}Cf and for $^{16,18}O$, $^{20,22}Ne$, and $^{40,48}Ca$ reactions with ^{248}Cm.

(e) The mass-yield shifts for $\Delta Z=+1$ and $\Delta Z=+2$ transfers for the $^{16,18}O$ ion pair on ^{249}Cf are less than the 2-mass number shift expected from past studies with ^{248}Cm targets. However these shifts are in agreement with recent studies[7] of these light-heavy ion pairs with ^{254}Es.

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TABLE I. Decay modes, energies, and abundances of measured radiations.

Nuclide		Decay Mode	Energy of Radiation (MeV)	Abundance
Bk	244	EC, γ	0.2176	a
	245	EC, γ	0.2528	0.313
			0.3808	0.026
	246	EC, γ	0.7987	0.610
			0.8335	0.500
			1.0814	0.580
	248m	β−, γ	0.5507	0.046
Cf	247	EC, γ	0.2941	0.0098
			0.4179	0.0034
	248	α	6.26	1.00
Es	248	α	6.87	0.0025
	249	EC, γ	0.3795	0.404
			0.8132	0.092
	250(6+)	EC, γ	0.3033	0.223
			0.3494	0.204
			0.3837	0.140
			0.8290	0.736
	250(1−)	EC, γ	0.9891	0.134
			1.0319	0.106
	251	EC, γ	0.1529	0.0091
			0.1777	0.014
	253[b]	α	6.63	0.98
Fm	250	α	7.43	0.93
	251	α	6.83	0.85
	252[c]	α ➤ α	6.26	1.00
	253[c]	EC ➤ α	6.63	0.86
	254	α	7.06 - 7.19	1.00
	256	SF		0.92

a. ^{244}Bk absolute γ-ray abundances have not been measured; 100Δ abundance was assumed for the 0.2176 MeV γ-ray and thus the cross sections are lower limits.

b. Corrected for contribution from decay of Cf parent.

c. Radiation from daughter was measured.

TABLE II. Cross sections for the production of heavy actinides from bombardments of ^{249}Cf with ^{16}O of different energies.

	90 MeV[a] Cross sections (μb)	s[b] (%)	96 MeV[a] Cross sections (μb)	s[b] (%)	106 MeV[a] Cross section (μb)	s[b] (%)	115 MeV[a] Cross section (μb)	s[b] (%)	122 MeV[a] Cross section (μb)	s[b] (%)	139 MeV[a] Cross section (μb)	s[b] (%)	150 MeV[a] Cross section (μb)	s[b] (%)
Bk														
244[c]	-	-	1.3	17	4.8	4.4	6.9	7.5	4.3	2.8	4.3	12	3.1	14
245	1.7	26	39	3.8	104	3.5	64	32	74.6	1.2	83.5	5.0	46.7	6.4
246	19.9	5.0	150	1.2	159	2.0	136	4.8	96.3	0.8	113	4.4	69.6	3.7
248m	1.9	-	4.2	-	5.3	-	9.1	-	1.5	-	3.9	-	9.8	-
Cf														
247	105	9.2	2860	2.0	-	-	-	-	3310	3.5	3050	1.2	2540	2.7
248	5215	0.1	27400	0.1	23200	0.1	21200	0.1	14700	1.7	17700	0.1	13000	3.9
Es														
248	5.5	16	44	8.8	241	6.2	237	7.3	77	7.8	35	50	5	9.9
249	43.5	5.6	1800	1.9	3550	1.7	2800	1.3	2480	0.6	2160	1.6	1660	1.3
250(6+)	588	0.8	5130	0.3	4650	0.5	2580	0.4	1750	0.4	1280	0.5	1170	0.5
250(1+)	834	3.4	7460	1.8	7170	1.1	4370	1.3	3300	2.2	2470	2.5	1890	2.7
251	66	13	850	24	633	28	554	8.4	783	3.7	541	28	413	18
253	1.5	20	3.4	3.5	6.6	1.5	2.1	4.3	2.4	2.1	1.3	3.7	1	12
Fm														
250	13	8.3	1241	0.4	904	1	584	1.2	385	1.1	32	1.8	11	3
251	165	6.8	4165	1.9	3805	1.2	1554	4.2	1473	2.9	377	4	339	6.8
252	54	1.4	1467	0.4	1272	0.4	1089	15	444	0.3	148	0.9	144	2.2
253	42	1.5	420	0.8	269	0.6	137	0.4	95	0.8	27	0.6	29	1.4
254	0.37	13	32.5	0.9	20	0.6	8.8	1.7	3.3	5.5	1.4	6.3	1.5	2.6
256	≤0.00004[d]	50	0.036	11	0.046	8.7	0.047	11	0.049	2.0	0.011	9.1	0.008	30

a. Average projectile energy (laboratory system) in the target.

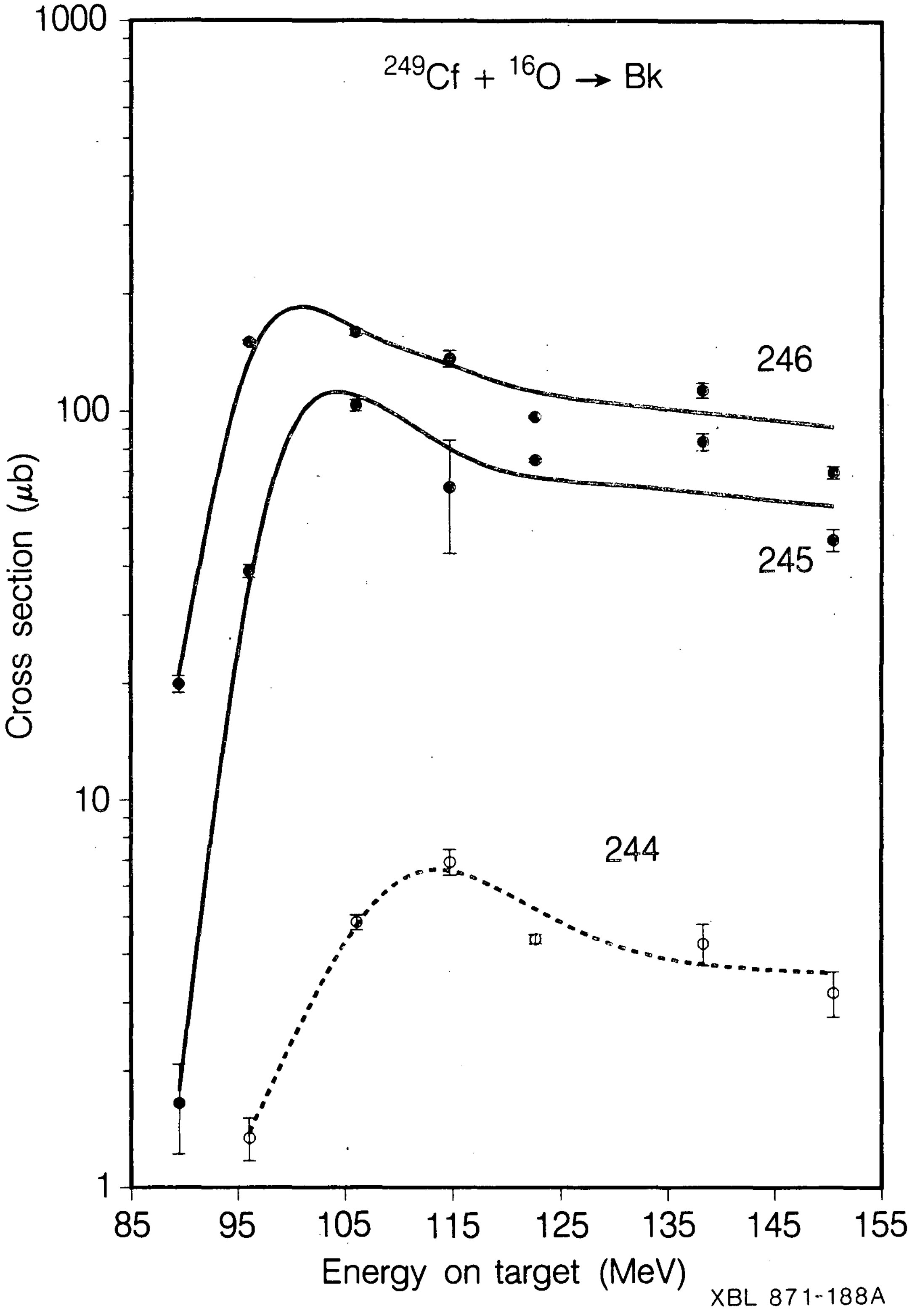
b. The standard deviation associated with the quoted absolute cross sections is estimated to be +/-12 in addition to the listed statistical standard deviation, which is based on the decay analysis. This added error is estimated to be +/-22 for the 150-MeV data.

c. ^{244}Bk cross sections are only lower limits since the absolute abundances of the gamma transitions are unknown for this isotope. The shape of the curve is unaffected.

d. Lower limits only since no activity was detected.

TABLE III. Comparison of maxima, E_M, estimated from experimental excitation functions for ^{249}Cf + ^{16}O with calculated[4] excitation energies, E_X.

Nuclide	E_M	E_X
Bk-244	110	-3.8
245	101	-5.4
246	101	-7.0
Es-248	107 MeV	-2.4 MeV
249	102 MeV	-1.3 MeV
250	98 MeV	1.9 MeV
251	101 MeV	-2.6 MeV
253	106 MeV	-22.6 MeV
Fm-250	97 MeV	2.0 MeV
251	98 MeV	6.6 MeV
252	98 MeV	5.3 MeV
253	98 MeV	5.4 MeV
254	99 MeV	-7.2 MeV
256	105 MeV	-31.1 MeV



^{249}Cf + ^{16}O → Bk
246
245
244
Cross section (μb)
1000
100
10
1
85
95
105
115
125
135
145
155
Energy on target (MeV)
XBL 871-188A

Fig. 1

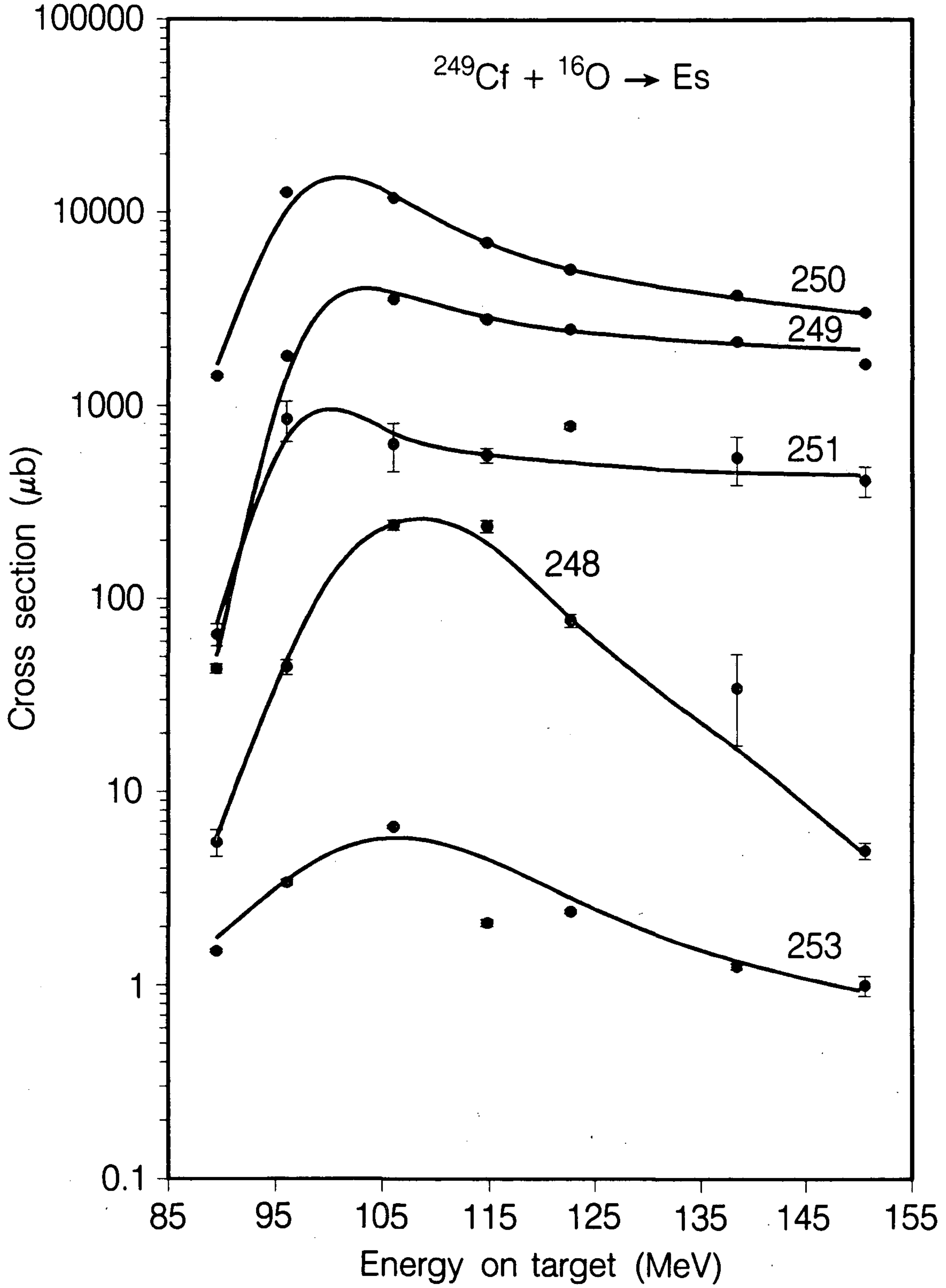


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Fig. 2

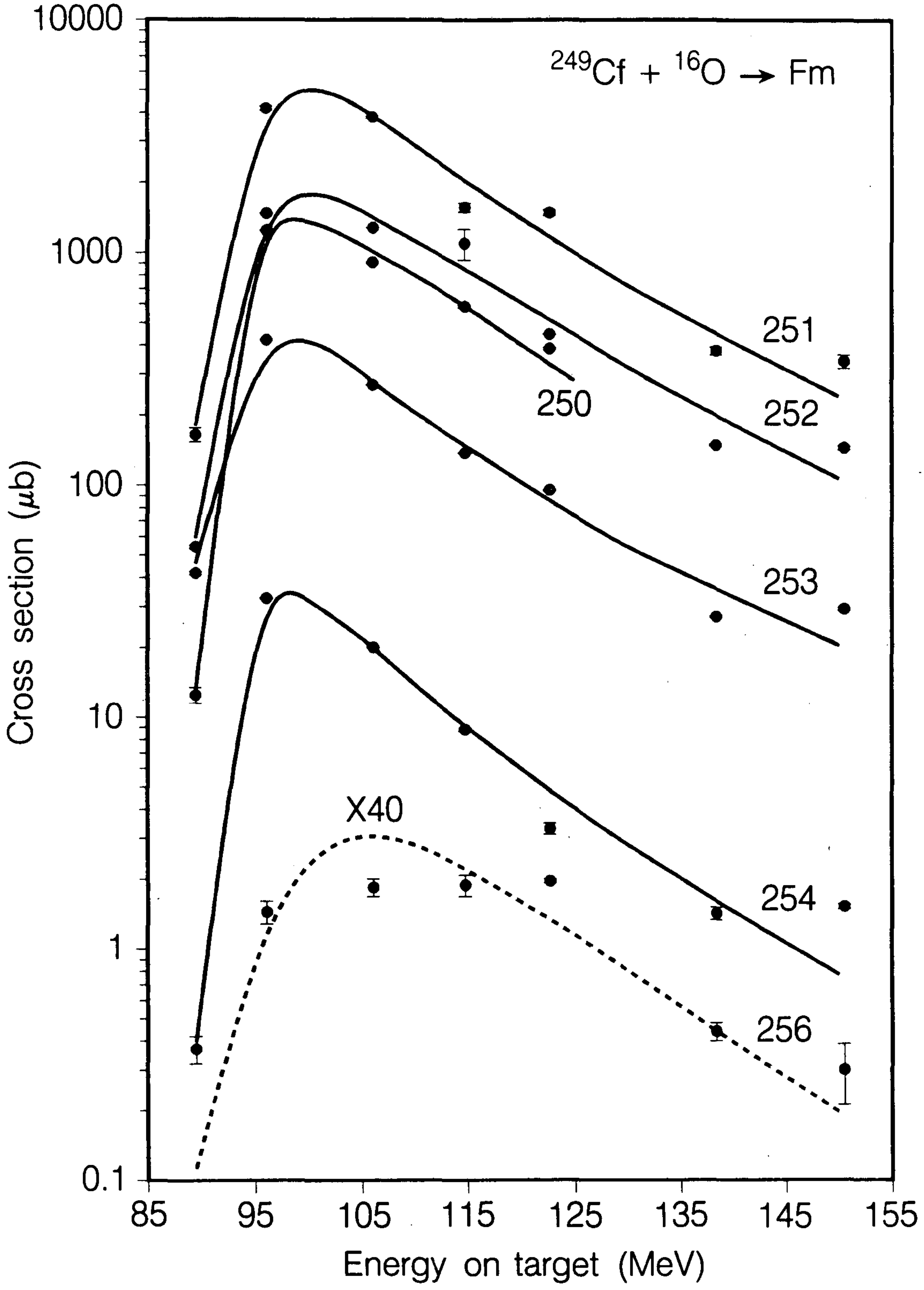

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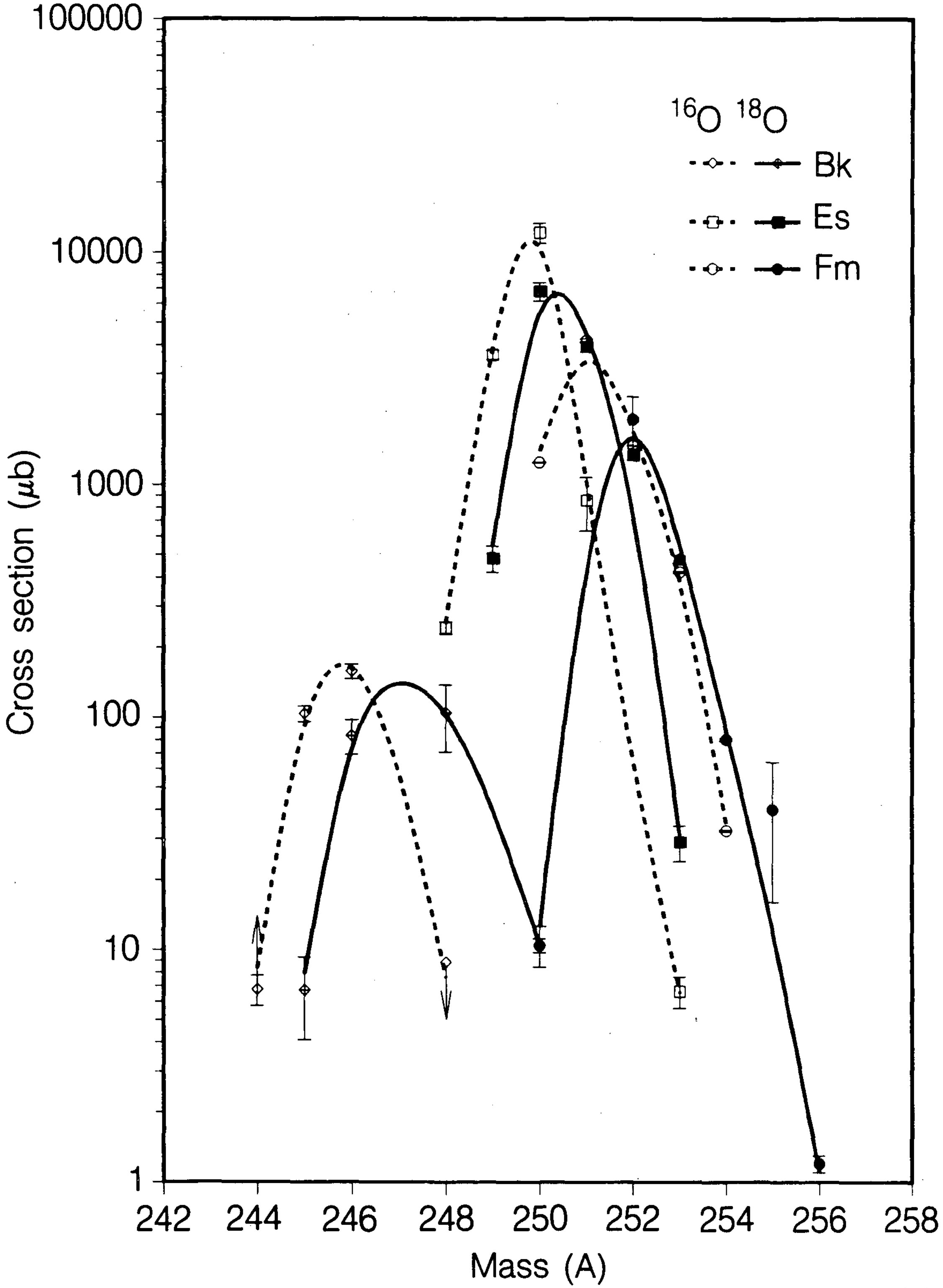
Fig. 3



XBL 871-189

Fig. 4

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