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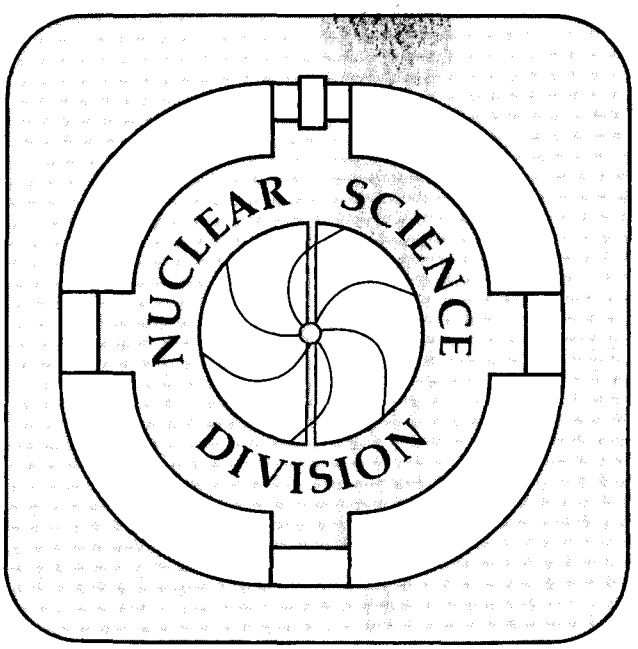

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TRANSFER REACTIONS WITH HEAVY ELEMENTS

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In the last few years there has been a renaissance of interest in transfer reactions with heavy element targets. Some early studies of transfer reactions[1-3] were performed and P. Eskola[3] pointed out in 1975 that in many cases nuclides could be produced in high yield by heavy ion transfer reactions when they could not be produced by compound nucleus reactions. Nevertheless, until recently these reactions remained relatively unexploited in favor of fusion reactions between the heaviest actinide targets available and relatively light projectiles. Such reactions were used in the discovery of the new elements 102 through 106. However, because of the rather high excitation energies of the compound nuclei produced in this way, severe losses from prompt fission and particle emission occur and the production cross section for element 106 was only 0.3 nb for the ^{249}Cf (^{18}O, 4n) reaction to make ^{263}106. The discovery of still heavier elements depended on a rather radical departure to the use of lighter elements and "cold fusion", a method first used by Oganessian and co-workers[4]. This technique was exploited by Münzenberg et al.[5,6,7] to produce elements 107, 108, and 109 using the relatively "light", stable targets Bi and Pb with the heavy ion beams of ^{54}Cr and ^{58}Fe available at GSI.

Like the "cold fusion" reactions, binary transfer reactions are attractive because heavy products can be produced with low excitation energies in reactions between heavy ions and the heaviest actinide targets. This is primarily because many of the reactions to produce heavy actinide products are rather endoergic and energy must be provided in order to make these reactions proceed. In these binary transfer reactions, nucleons or clusters of nucleons are exchanged between target and projectile without complete fusion. In addition to the fact that many of the ground state Q values are negative, the resultant projectile-like fragment carries off much of the kinetic energy and

angular momentum, thus leaving a residual heavy product with low excitation energy and, therefore, a highly reduced probability for fission or particle emission compared to products from the complete fusion process. Eskola[3] also pointed out that the cross sections for products between the target and compound nucleus are larger than for the compound nucleus reactions. This can be both beneficial and detrimental inasmuch as it means that literally a plethora of products between target and compound nucleus can be produced and identification of a specific product can be very difficult.

We "rediscovered" transfer reactions[8] with heavy element targets in 1979 when we used our rotating wheel system, the MG (Merry-Go-round) to detect 1.5-s ^{259}Fm from the reaction of 95-MeV ^{18}O projectiles with a ^{248}Cm target. The ^{259}Fm was presumably produced via an effective transfer of ^{11}Be to the ^{248}Cm with a cross section of about 15 nb. In order to see if this cross section was reasonable and to positively identify and measure the cross sections for other products, we began a systematic study of products of this and other target-projectile systems using off-line radiochemical techniques. Our early results[10] for ^{248}Cm with ^{16}O, ^{18}O, ^{20}Ne, and ^{22}Ne showed that the peaks of the mass-yield curves for Bk through Fm are about 2 mass units larger for ^{18}O and ^{22}Ne than for ^{16}O and ^{20}Ne, thus reflecting the two-neutron excess of the heavier projectiles.

Since then the production of heavy actinides from the interactions of ^{248}Cm, ^{249}Bk, ^{249}Cf, and ^{254}Es targets with a variety of projectiles has been studied[11-15] by several collaborations using the accelerators at both GSI and LBL and on-line as well as off-line radiochemical separation techniques. (See Table I.) Excitation functions were measured[11-13] for reactions of ^{248}Cm and ^{249}Cf with ^{18}O ions from energies just below to 50 MeV above the Coulomb barriers and for ^{248}Cm with ^{40}Ca and ^{48}Ca ions with energies from just below to 80 MeV above the Coulomb barriers. The results of these measurements can be summarized briefly as follows:

1. The production of neutron-rich heavy actinides is enhanced by the use of the neutron-rich projectiles ^{18}O and ^{22}Ne, reflecting the 2-neutron excess of the projectiles over ^{16}O and ^{20}Ne.

2. The maxima of the isotopic distributions occur at only 2 to 3 mass numbers larger for ^{48}Ca than for ^{40}Ca reactions with ^{248}Cm.

3. The cross sections decrease rapidly with the number of nucleons transferred.

4. The use of neutron-rich targets favors the production of neutron-rich isotopes.

5. "Cold" heavy products are produced. For most of the heavy isotopes measured, the excitation functions are quite broad and do not decrease rapidly with increasing energy, in contrast to the compound nucleus reactions.

6. Comparisons with simple calculations of the product excitation energies assuming binary transfers indicate that the maxima of the isotopic distributions occur at the lightest product isotope for which the energy exceeds the reaction barrier.

7. The cross sections for transfer of the same nucleon clusters appear to be comparable for a wide variety of systems.

At about the same time these results were becoming available, a workshop was convened in 1983 by the National Research Council at the request of the U.S. Department of Energy to assess[16] the current status and future opportunities in research with transplutonium elements. In the area of nuclear research, the panel concluded that, "the exploration of the limits of nuclear stability is a prime motivation for studying nuclear species with the highest atomic numbers accessible. Improved understanding of nuclear-reaction mechanisms recently achieved gives great promise for reaching uncharted regions of nuclei at the upper end of and beyond the actinides, including presumably longer-lived isotopes of known elements than were previously available and possibly the long-sought superheavy elements of $Z \approx 114$ with neutron numbers near 184."

In the area of chemical research, the panel concluded that, "From the chemist's point of view, the periodic table of elements is the most basic road map and to extend it to its farthest reaches is an obvious goal... Focus on the most basic chemistry for the transeinsteinium elements, including determination of properties of the metallic atoms, range and stability of oxidation states, ionic radii and complexation behavior, and simple binary molecular species." As an imaginative

response to these recommendations, a consortium of four national laboratories in the United States, Lawrence Berkeley Laboratory, Lawrence Livermore National Laboratory, Los Alamos National Laboratory, and Oak Ridge National Laboratory proposed[17] the Large Einsteinium Activation Program or LEAP to prepare a source of ^{254}Es more than 10 times larger than the current production. The LEAP proposal constituted a major initiative to exploit the currently existing expertise in heavy element research and the unique potential for processing ^{254}Es which exists at the HFIR-TRU complex at ORNL.

The program proposes production of the largest ever target of ^{254}Es, at least 30 micrograms, which would permit preparation of a 400 $\mu g/cm^2$ target by depositing the ^{254}Es with a diameter of about 3 mm. The scientific goals of the LEAP are threefold: 1) to produce and identify neutron-rich isotopes of the heaviest elements by bombardment of the target with neutron-rich heavy ions in order to study nuclear properties, especially spontaneous fission, at the extreme limits of nuclear stability; 2) to prepare sufficient quantities of the heaviest actinides and transactinides for studies of their chemistry; 3) to produce superheavy elements by irradiation of the ^{254}Es target with ^{48}Ca, which gives a compound nucleus whose neutron number of 183 is nearer the predicted closed shell at 184 than has previously been achieved. These studies require access to an accelerator which can provide stable and relatively high intensities of neutron-rich light heavy ion beams such as ^{18}O, ^{22}Ne, and ^{48}Ca.

Since that proposal was prepared and presented the laboratories involved have proceeded with "pre-LEAP" experiments which are confirming the prediction that transfer reactions between heavy ions and an ^{254}Es target can provide access to the unexplored region of neutron-rich heavy element isotopes of the trans-einsteinium isotopes. (See Fig. 1.)

Recently, the previously unknown ^{260}Md was produced and isolated [18] from the products of ^{18}O and ^{22}Ne reactions with a 40 $\mu g/cm^2$ ^{254}Es target. The cross section for its production was measured to be 0.3 microbarns for both ^{18}O and ^{22}Ne reactions with ^{254}Es, consistent with predictions for a ^{6}He transfer based on the systematics for He transfers for a variety of target and projectile systems. The half-life was measured to be 32 days, much longer than

predicted. (See Table II.) The mass was determined by electromagnetic separation and the element assignment was based on the production cross section[19] and the predicted decay properties for isotopes of mass 260. Subsequent experiments indicated that ^{260}Md decays primarily by spontaneous fission rather than by electron capture or beta decay.

The use of transfer reactions with ^{254}Es has also permitted the extension of measurements[20] of mass and kinetic-energy distributions for the spontaneous fission (SF) of a number of additional heavy element isotopes and has revealed that although the mass distributions for ^{254}Fm, ^{259}Md, ^{260}Md, ^{258}No, and ^{260}104 are all symmetric, the full-widths at half-maximum vary from 8 mass units for ^{258}Fm and ^{260}Md to 36 mass units for ^{260}104. Asymmetric tailing in the total kinetic energy distributions was observed at both higher and lower energies than the most probable total kinetic energy. These observations have been interpreted[20] as resulting from a mixture of liquid-drop and fragment-shell controlled fission.

^{260}Lr (half-life = 3 min) has been prepared by ^{22}Ne reactions (^{6}Be transfer) with an ^{254}Es target and by ^{18}O reactions with a ^{249}Bk target. We have determined its elution position from a cation exchange resin column with alpha hydroxyisobutyrate has been determined relative to the rare earth pairs Yb-Er and Tm-Ho. Its elution position was very close to that of Er and from the data obtained from many runs in which only about an atom every other run was detected, the ionic radius was deduced to be 0.885 + 0.004 angstroms. This is in excellent agreement with the prediction of 0.885 made by Goldman & Morss[21]. Additional experiments to prepare the predicted +1 oxidation state are planned but would be much easier with the proposed large ^{254}Es target.

We are continuing our investigations of transfer reactions with different target and projectile systems and have recently measured the actinide production cross sections for ^{18}O reactions with ^{249}Bk and ^{249}Cf targets with both our on-line rotating wheel system and with off-line radiochemical techniques. (See Fig. 2.) The on-line measurements indicate the production of both ^{256}Lr and ^{260}Lr from the ^{249}Cf target, effective transfers of ^{7}B and ^{11}B, with cross sections of about 10 and 40 nb, respectively. These are at the optimistic end of the range predicted earlier (See Table III) for B

transfers. Pending the availability of a large ^{254}Es target, it may be much easier to produce ^{260}Lr via a ^{11}B transfer to ^{249}Cf rather than by the ^{18}O (alpha, 3n) reaction with ^{249}Bk which has a cross section of only 20 nb. If the B transfer cross sections are this large, it would also be feasible to produce sufficient quantities for chemical studies of isotopes of element 104 from a large ^{254}Es target.

Our on-line measurements of the ^{249}Bk target showed evidence for both No and Lr isotopes (B and C transfers), but because of the rapid growth of ^{249}Cf into the ^{249}Bk, large corrections for ^{249}Cf were necessary and confirmation awaits additional measurements on a freshly separated ^{249}Bk target. We are particularly interested in measuring yields of some transfer reactions with odd Z targets such as ^{249}Bk to see if there is a pronounced effect on the yields of various types of transfers. Schädel et al.[14] compared their results for Li and Be transfers to ^{254}Es (odd-Z) with our earlier results[10,11] for ^{248}Cm (even Z) interactions with ^{18}O and ^{22}Ne projectiles. The cross sections are roughly comparable for the ^{22}Ne reactions, but the yields are considerably lower for the ^{18}O reactions. However, since the ^{18}O energy used was only about at the Coulomb barrier and the reactions to produce most of the measured No and Lr isotopes are quite endoergic, the excitation functions need to be investigated before a firm conclusion can be reached. Our measured excitation functions[11] for ^{18}O reactions with ^{248}Cm and ^{249}Cf to produce Es and Fm isotopes show maxima which are related to the calculated[22] reaction barriers. The maxima of the excitation functions tend to be at higher energies for those isotopes with lower calculated excitation energies. Furthermore, it appears that the cross sections for endoergic reactions remain relatively low even when the projectile energy is increased. Perhaps this can be partially explained if the energy is apportioned according to the fraction of projectile mass transferred. Then for these reactions somewhat less than half of the incident kinetic energy above the Coulomb barrier would be expected to result in internal energy of the target-like product. This may account for our observation for the reactions of ^{248}Cm with ^{48}Ca that the excitation functions[13] reached a maximum some 20 MeV above the Coulomb barrier (consistent with our calculations indicating quite negative excitation energies), but

decreased only slowly with projectile energies as high as 80 MeV above the barrier. However, our earlier attempts to correlate the fraction of projectile energy transferred for the $^{18}O + ^{248}Cm$ and ^{249}Cf systems with the fraction of projectile mass transferred indicated that about half the projectile kinetic energy was transferred for up to 8 nucleons transferred. Obviously additional experiments are required in order to better understand the energy partition for these systems.

All of the "pre-LEAP" experiments conducted to date have confirmed the potential for performing additional exciting experiments with a still larger target of ^{254}Es. It also appears to be feasible to produce[23] such a target and we are hoping that we will soon be able to take this LEAP forward!

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Table I

Recent Radiochemical Studies of Transfer Reaction with Transuranium Targets

Targets	Projectiles	Energies MeV
^{248}Cm	$^{16,18}O$	90-150
	$^{20,22}Ne$	115-116
	$^{40,48}Ca$	234-318
^{249}Bk	^{18}O	102, 109
^{249}Cf	$^{16,18}O$	91-150
^{254}Es	$^{16,18}O$	98
	$^{20,22}Ne$	121, 126
	^{48}Ca	266

Table II

ESTIMATED PRODUCTION RATES FOR NEW N-RICH ISOTOPES
(400 μg/cm^2 ^{254}Es, 1 p μA beam)

NUCLIDE	ESTIMATED $T_{1/2}$	REACTION	RATE ATOMS/m
Md-260	1-10h β	(^{6}He)T	800
261	2-200 μs, SF	(^{7}He)T	80
262	≈0.2 s, SF	(^{8}He)T	8
No-260	< ps SF	(^{6}Li)T	1000
261	1.1 h α	(^{7}Li)T	80
Lr-261	5 m α	(^{7}Be)T	400
262	14 m α	(^{8}Be)T	120
263	0.7 h α	(^{9}Be)T	20
264	2.3 h α	(^{10}Be)T	8
Rf-262	ms, SF	(^{8}B)T	4
263	1.5 m α	(^{9}B)T	4
Ha-263	0.6 s α	(^{12}C, 3n)	4
264	4 s α	(^{10}C)T	0.4

Table III

TRANSFER REACTIONS MAXIMUM YIELDS

TRANSFERRED FRAGMENT	CROSS SECTION (μb)
^{1,2}H	2000-3000
^{2,3}He	1000-2000
^{4,5}Li	30
^{6,7}Be	2-3
$^{7-9}$B	0.01-0.10 *
$^{9-11}$C	0.001-0.01 *

*Wild Extrapolations!!

Table IV

PRODUCTION RATES FOR CHEMICAL STUDIES
(400 μg/cm^2 ^{254}Es, 1 p μA beam)

NUCLIDE	REACTION	$T_{1/2}$	ATOMS/MIN.
^{256}Md	(^{18}O or ^{22}Ne)T	1.3 h	8 x 10^5
^{259}Md	"	1.6 h	4 x 10^4
^{259}No	(^{22}Ne)T	58 m	2 x 10^3
^{260}Lr	"	3 m	3 x 10^2
^{261}Rf	(^{11}B, 4n)	1 m	(20)
^{262}Ha	(^{12}C, 4n)	34 s	(8)

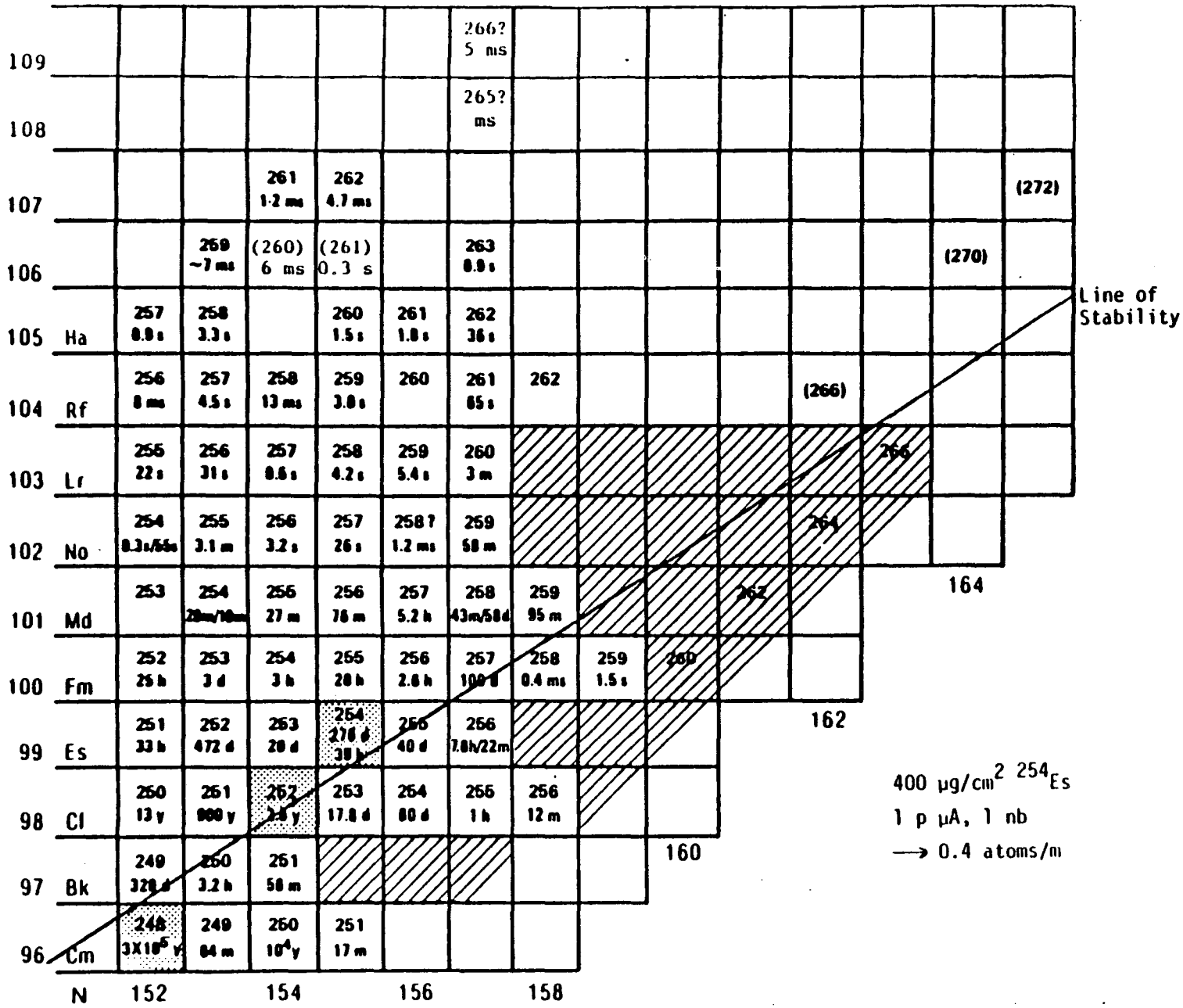
Figures

Fig. 1. Portion of table of isotopes showing neutron-rich trans-Am isotopes. Hatched regions indicate new actinide isotopes postulated to be produced with >1-nb cross sections from transfer reactions to a 400 $\mu g/cm^2$ ^{254}Es target with a projectile beam of 1 p A. Compound nuclei for reactions of ^{18}O with ^{248}Cm, ^{252}Cf, and ^{254}Es are shown in parentheses.

Fig. 2. Isotopic distributions measured for 104-MeV ^{18}O reactions with ^{249}Cf.

Fig. 3. Mass-yield curves for Fm from ^{16}O and ^{18}O Reactions with ^{248}Cm and ^{249}Cf.

Fig. 4. Excitation functions for Es and Fm isotopes from the interaction of ^{48}Ca with ^{248}Cm.



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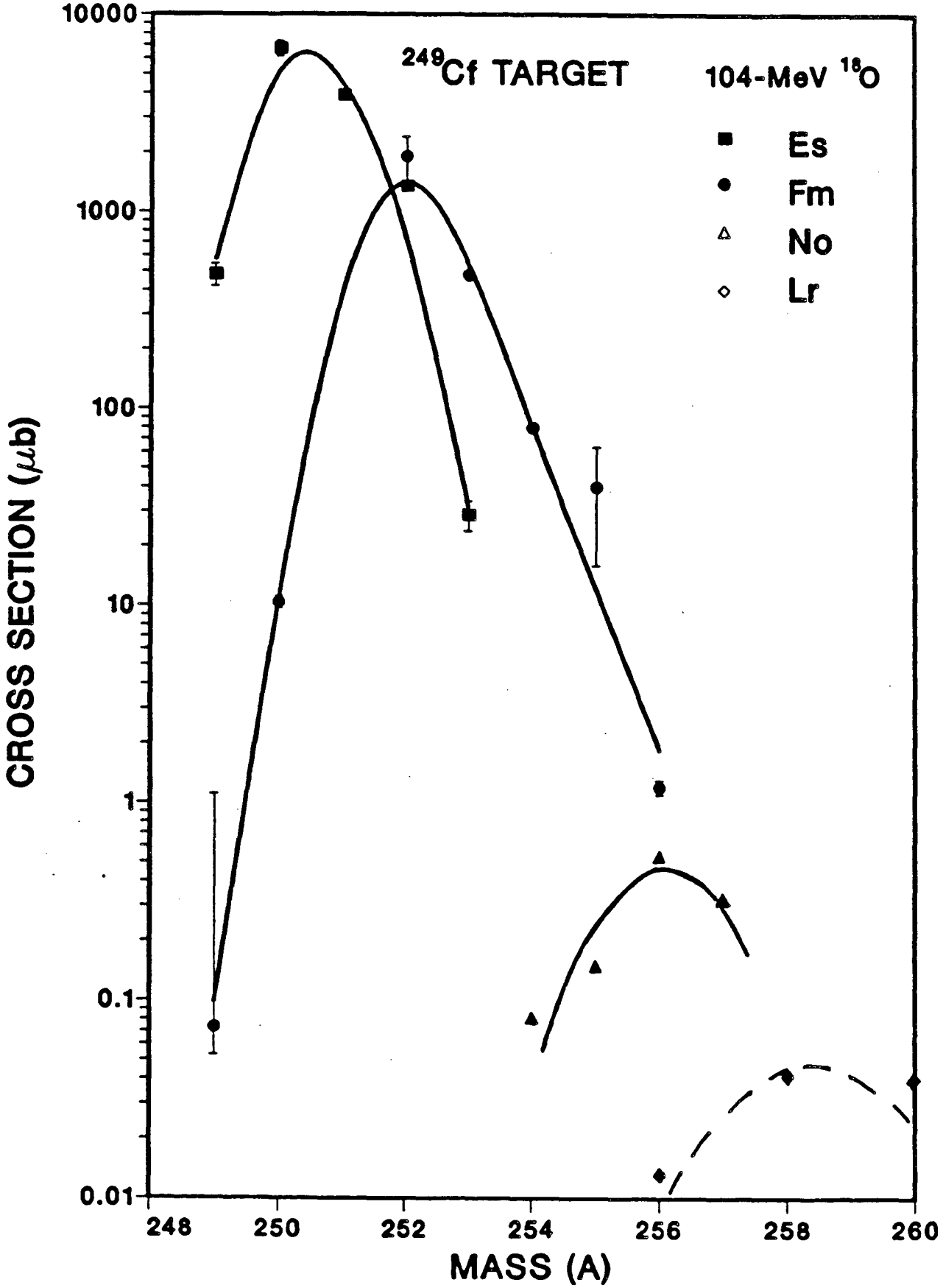

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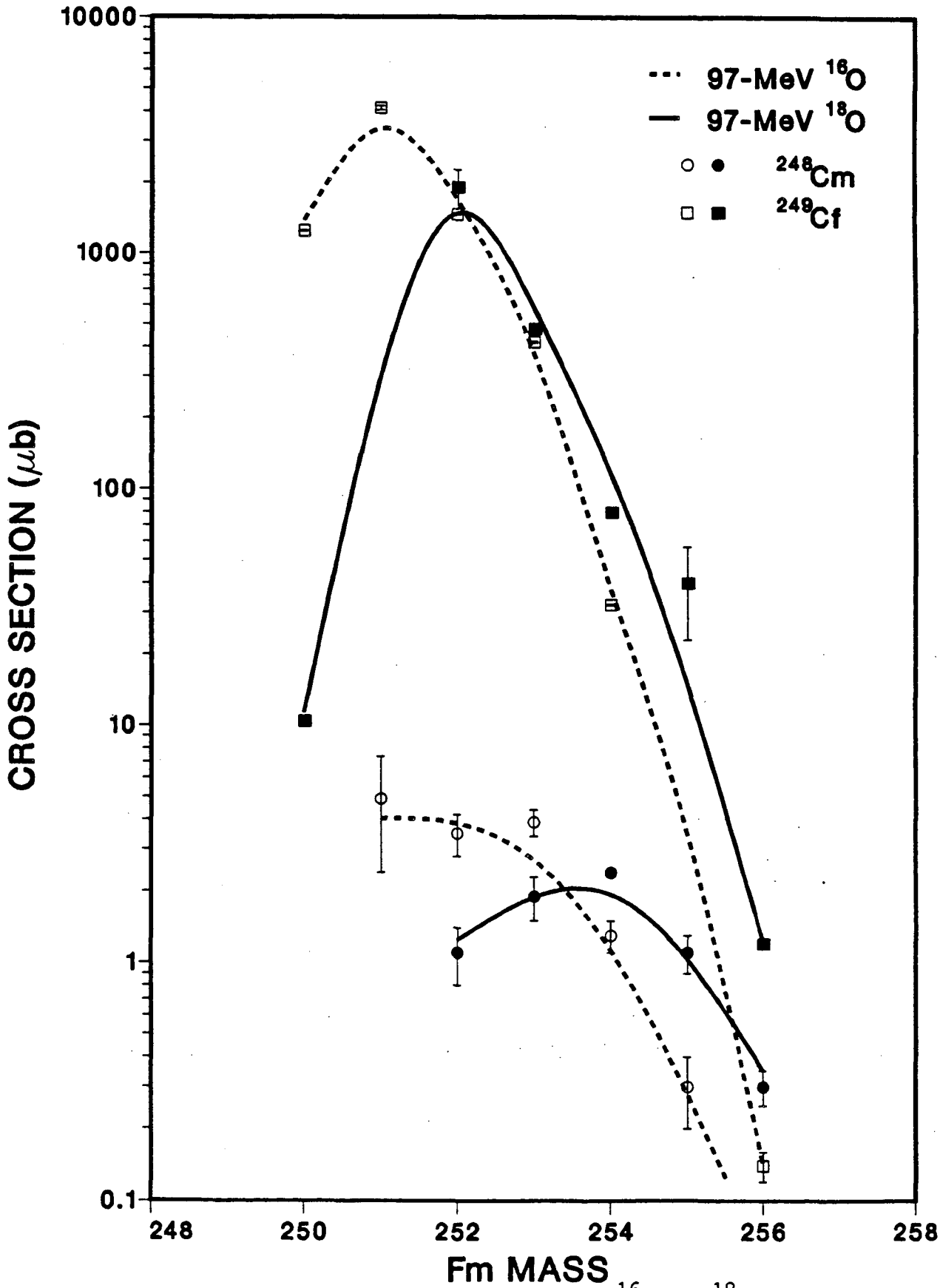

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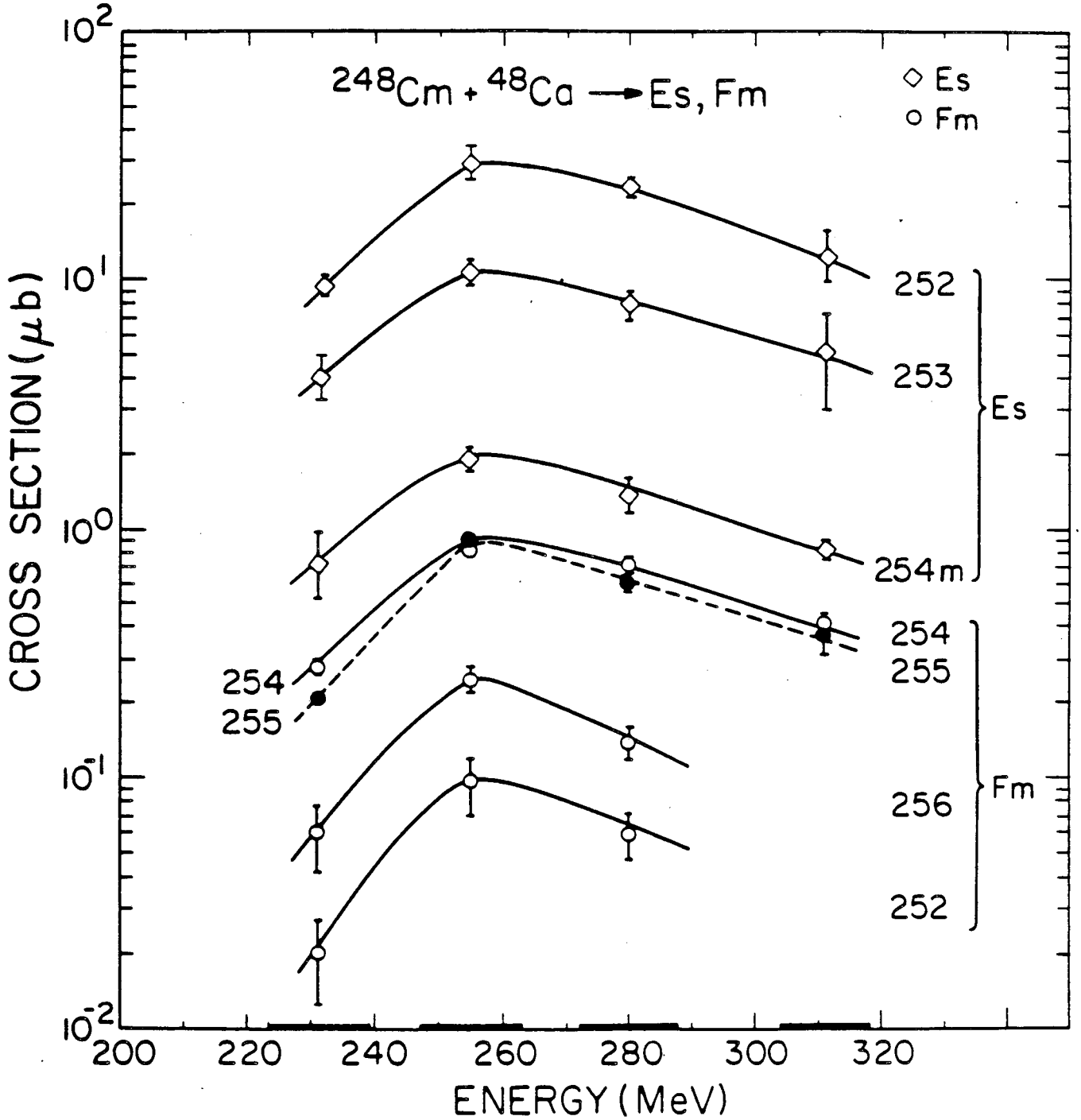

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