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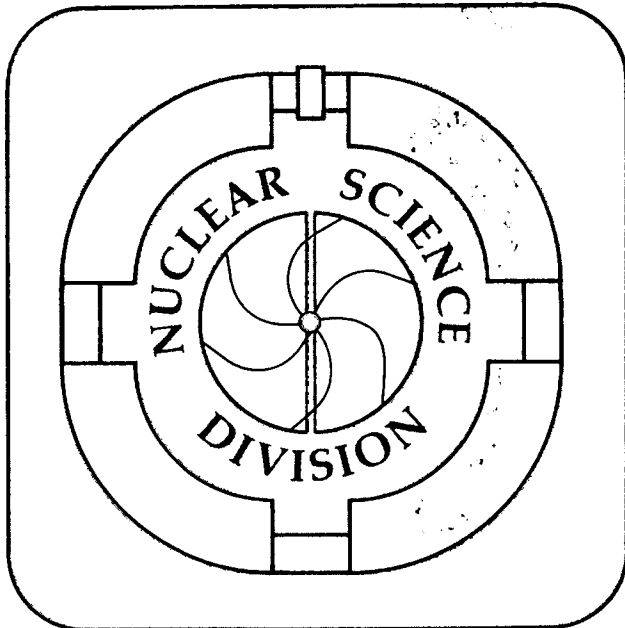
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TRANSURANIUM ISOTOPES

D.C. Hoffman

December 1985

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TRANSURANIUM ISOTOPES

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Abstract

The needs of the research community for the production of transuranium isotopes, the quantities required, the continuity of production desired, and what a new steady state neutron source would have to provide to satisfy these needs are discussed. Examples of past frontier research which need these isotopes as well as an outline of the proposed Large Einsteinium Activation Program, LEAP, which requires roughly ten times the current production of ^{254}Es are given.

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TRANSURANIUM ISOTOPES

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Introduction

Whenever one agrees to take on the responsibility for representing the needs, views, or research interests of a given "community" of scientists (or any other group), one realizes that these can potentially be as diverse as the number of possible investigators to the n^{th} power where n is the number of variables being considered. Also, in this instance, I think there are many scientists who do not necessarily consider themselves as part of the transuranium research community who would nonetheless like to extend their fields of research to the transuranium isotopes or use them in applied studies. Although an intense steady state source of neutrons could be used by the transuranium research community in other ways, e.g., neutron scattering experiments to elucidate the structures of compounds containing transuranium elements, requirements for these studies will no doubt be included in general discussions of structural studies. Certainly, such needs should not be overlooked.

However, in this paper I will concentrate on the research needs for the production of transuranium isotopes, the quantities and purity required, the continuity of production desired, and what a new high flux reactor would need to provide in order to satisfy these needs. Then, as an example of the research that can be performed, I will outline the major features of the Large Einsteinium Activation Program, LEAP, proposed for frontier research on both the chemical and nuclear properties of the heaviest elements. It requires production of some 40 micrograms of ^{254}Es , roughly 10 times the current production.

Production of Transuranium Isotopes

Current production of the heaviest transuranium isotopes at the HFIR-TRU facilities at ORNL is shown in Table I [1]. Macroscopic quantities of some of these elements are available for studies of their nuclear and inorganic chemistry. Others serve as accelerator targets for preparation of other heavy element isotopes for studying their nuclear properties and probing the limits of nuclear stability in this region. Sometimes their chemical properties can be explored if sufficient quantities of isotopes with appropriate half-lives can be produced. Other of these isotopes, such as ^{252}Cf , have found applied uses in medicine and as neutron sources. In fact, sources of 100-200 mg have been requested for use in neutron radiography and neutron activation.

It is apparent from the half-lives that none of these isotopes except ^{248}Cm and ^{249}Cf can be stockpiled and, therefore, the others must be produced on a continuing basis if they are to be available in the future for use in either fundamental research or applied areas. Moreover, the LEAP will need about 10 times the currently produced amount of ^{254}Es ; ^{257}Fm would be still better than ^{254}Es if it could be made available in much larger quantities than can currently be produced.

It is quite obvious that in any proposed successor to HFIR, we would like a reactor that would offer a significantly enhanced production capability for isotopes such as ^{254}Es . Because of the high cross section for destruction of ^{254}Es from thermal neutron-induced fission, Bigelow et al. [2] have found that the production can be greatly enhanced by irradiating the target material (^{252}Cf) wrapped in cadmium which will absorb thermal neutrons but pass the epithermal neutrons. The capture cross section for ^{253}Es for epithermal

neutrons is only slightly lower than for thermal neutrons, but the fission cross section for ^{254}Es is much lower, and therefore, the total amount of ^{254}Es which results is much greater.

The production scheme for ^{254}Es which has been proposed by Bigelow et al. [2] is shown in Fig. 1. Some notable features of this scheme are: a) irradiation of about 1 gram of ^{252}Cf (representing nearly the entire inventory at TRU) in the normal flux of the HFIR for about 20 days followed by cooling for 20 days to allow 18-day ^{253}Cf to decay to ^{253}Es ; b) irradiation in the maximum thermal flux for about 5 days followed by a short decay period; c) irradiation in a cadmium-filtered flux for 24 to 28 days during which time several shutdowns will be required in order to replace the cadmium filters and spent fuel assemblies; d) removal of the target to TRU for isolation and purification of the einsteinium fraction; e) decay for about 6 months to allow 20-day ^{253}Es to decay to approximately equal activity to the 276-day ^{254}Es before preparation of the targets for accelerator irradiations. (At equal activities of ^{254}Es and ^{253}Es , the sample will contain about 3×10^{11} alpha decays per minute in 30 μg . Shielding from the alpha activity is relatively easy, but unfortunately the 3-hour ^{250}Bk daughter has ≈ 1 MeV gamma rays which will give about 10R/h at 5 cm, so considerable shielding will be required.) This entire effort is estimated to require about 15 months.

The proposed production cycle illustrates very well some of the features that are essential for a viable production program for transuranium isotopes for heavy element research. These can be summarized as follows:

1. Fluxes of thermal and epithermal neutrons at least as high, and preferably higher, than currently available at HFIR.

2. Region of high epithermal flux with epithermal to thermal flux ratio higher than in the HFIR.

3. Space for at least as many target positions as currently available.

4. Continuity of production. No protracted shutdowns or unavailability of irradiation ports.

5. Possibility for some flexibility in scheduling and target position so as to be able to perform procedures such as c) above which require several shutdowns, etc.

6. Provision for rapid removal of shorter-lived products if desired.

7. A transuranium processing facility for fabricating the targets and performing the very difficult processing of the highly radioactive products to provide the extremely pure transuranium isotopes currently available. This facility must be located in close proximity to the reactor as rapid transport and reirradiation of products are often required. Shipment of the highly radioactive unseparated targets to another site would be impractical and expensive even if technically possible.

8. Associated research facilities where relevant cross section measurements or possibly other experiments can be performed.

I mentioned that larger quantities of ^{257}Fm would be highly desirable. Currently only a few picograms are available compared to microgram quantities of ^{254}Es . Since there is a factor of about 10^6 in the amount available for targets, the cross sections would have to be larger by that factor in order to make it advantageous to use ^{257}Fm as a target rather than ^{254}Es . Therefore, the production of ^{257}Fm would have to be increased by several orders of magnitude in order to make it feasible to use it as a target in light heavy ion reactions. We have, of course, used it for (t,p) reactions

to make ^{259}Fm and $(t, 2n)$ reactions to make ^{258}Md where the cross sections were of the order of tens of millibarns. Unless the cross sections for production of the desired isotopes are larger by factors of 10^6 (the ratio between the size of the ^{254}Es and ^{257}Fm targets available), or the production of ^{257}Fm can be increased significantly, there is no advantage in using ^{257}Fm as a target material.

Having stated some of the features the transuranium research community would deem desirable in a new high flux reactor, we ask what is the frontier science involved? Why do we want these isotopes and what will we use them for?

Research with Transuranium Isotopes

Much of the frontier research [3] in this region of the periodic table has utilized the products of HFIR-TRU. For example, elements Rf(104), Ha(105), and element 106 were discovered at Berkeley using ^{249}Cf (351 years) as the target for synthesis reactions with light heavy ions. Subsequent experiments to try to produce superheavy elements (SHE's) utilized ^{248}Cm produced by the transplutonium production program at HFIR-TRU. Currently, proposals have been made to use ^{254}Es , the heaviest isotope available in microgram quantities, to make another attempt to produce SHE's via reactions with ^{248}Cm .

Studies of the spontaneous fission (SF) process have also relied upon isotopes provided from this program. Early studies utilized ^{252}Cf to elucidate many of the properties of SF. Later studies of ^{257}Fm showed the first evidence for symmetric mass division in low-energy fission. Subsequently, a picogram of ^{257}Fm was used as a target for the first preparation of 1.5-sec- ^{259}Fm from the (t,p) reaction. It was found that ^{259}Fm fissioned spontaneously into two nearly equal mass fragments with

anomalously high total kinetic energies. Until the very recent discovery [4] of ^{260}Md by irradiation of ^{254}Es with ^{22}Ne , ^{259}Fm was the isotope with the largest number of neutrons so far produced. The observation of symmetric fission in ^{257}Fm and ^{259}Fm led to a renaissance of interest in the SF process because prior to that time it was believed that all low energy fission resulted in asymmetric mass division with total kinetic energies which could be readily extrapolated from lighter systems. A "disaster" in SF half-lives was also found to occur at ^{258}Fm where the SF half-life suddenly dropped from 500 years at ^{257}Fm to 0.38 ms at ^{258}Fm . None of these phenomena was predicted by nuclear theory nor has a comprehensive, dynamic, predictive theory of fission yet been developed.

The isotopes produced at HFIR-TRU have also been important for studies of chemical properties. The availability of macroscopic quantities of the elements curium, berkelium, californium, and einsteinium have made studies of their chemical properties possible. Since the longest-lived isotopes of Bk and Es have half-lives of less than a year, they must be produced on a continuing basis if such studies are to continue. These studies have shown that the actinide series clearly ends at lawrencium (element 103) and a new series begins at element 104 as required by Seaborg's formulation of a 14-electron 5f series beginning at actinium (element 89). The ground electronic state of fermium has even been experimentally determined to be $5f^{12}7s^2$ in an atomic beam experiment with ^{254}Fm derived from 39-hour ^{254}Es from a special preparation at HFIR-TRU. The actinide elements also present a unique opportunity for solid-state physics studies of the bonding properties of the 5f electron orbitals.

LEAP

The Large Einsteinium Activation Program (LEAP) was proposed [5] in 1984 by four national laboratories--Lawrence Berkeley (LBL), Lawrence Livermore (LLNL), Los Alamos (LANL), and Oak Ridge (ORNL). This joint proposal was prepared in an effort to launch a major initiative to exploit the currently existing expertise in heavy element research and the unique potential for producing very heavy actinide target materials such as 285-day ^{254}Es at the High Flux Isotope Reactor (HFIR) at ORNL. It was also envisioned that major new techniques and instrumentation would be developed to enhance our ability to identify and study both chemical and nuclear properties of new neutron-rich heavy element isotopes at the limits of nuclear stability and even superheavy elements (SHE's). A workshop convened in 1983 by the National Research Council at the request of the U.S. Department of Energy had assessed the current status and future opportunities in research with transplutonium elements [6].

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." The LEAP proposal constituted an imaginative response to some of these challenges, a response which would expand, as well as utilize, our current capabilities.

The program proposes production of the largest ever target of ^{254}Es , at least 30 micrograms, which would permit preparation of a $400\ \mu\text{g}/\text{cm}^2$ 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 . These can be provided at the 88-Inch Cyclotron and SuperHILAC at LBL.

Production of Heavy Element Isotopes

In order to study the chemical or nuclear properties of the heaviest elements we must first devise suitable production reactions. Traditionally, compound nucleus reactions involving complete fusion of projectile and target

nuclei, followed by particle emission, have been used. However, because of the high excitation energies of these compound nuclei and their high fissionability, losses due to prompt fission and neutron emission are excessive. Furthermore, it is difficult to produce the neutron-rich heavy element isotopes which we predict may have longer half lives. Fortunately, recent studies [7], [8], [9], [10] have shown that binary transfer reactions between neutron-rich, light heavy-ion projectiles and neutron-rich actinide targets can be used to produce neutron-rich products of elements up to four protons heavier than the target in relatively high yields. Because the Q values for these reactions are negative, the desired products can be produced with relatively low excitation energy and the excitation functions are quite broad as shown in Fig. 2 for production of Es and Fm isotopes from reactions of ^{48}Ca projectiles with ^{248}Cm [11].

Comparison of effective transfer of Be fragments for several different systems indicates comparable cross sections for these rather diverse systems. Based on systematics of this type and assuming similar cross sections for transfer of the same fragments to ^{254}Es , we estimate the production rates shown in Table II for isotopes of interest for chemical studies. Production cross sections for many of these isotopes have now been measured and indeed the comparison for ^{254}Es and ^{248}Cm targets given in Table III shows that much large quantities of these actinide isotopes can be produced [12] using ^{254}Es with ^{22}Ne projectiles. Currently, we are trying to measure cross sections for $Z = 5$ and 6 transfers to lighter actinide targets in order to predict what they may be for ^{254}Es .

Chemical Studies

Studies of the chemical properties of the heaviest elements are of particular interest because of the strong relativistic effects caused by the electric field resulting from the high nuclear charge. For example, calculations [13] show that because of relativistic stabilization of the p orbitals the electronic configuration for Lr ($Z = 103$) could be $5f^{14} 7s^2 7p_{1/2}$ rather than the $5f^{14} 6d^1 7s^2$ expected on the basis of simple extrapolation from the lighter actinides. Lr might then have a stable +1 oxidation state, although earlier studies [14] did show its extraction behavior was consistent with that of a trivalent actinide. We have installed a helium jet transport system at the 88-Inch Cyclotron at LBL and have successfully transported reaction products recoiling out of the target and attached to KCl aerosols to a chemistry laboratory some 80 meters away. The products are collected on aluminum foil, picked up in water and adsorbed on a tiny cation exchange resin column to investigate the elution position of 3-min. ^{260}Lr by elution with ammonium alpha-hydroxyisobutyrate. From the position of Lr relative to added rare earth tracers, the ionic radius can be deduced. A small ^{254}Es target ($20 \mu\text{g}/\text{cm}^2$) is being used and it is obvious from the low production rates that the large target will be required for more extensive studies. Plans to use other techniques such as thermochromatography, preparation of volatile fluorides of the transactinides using the powerful oxidizing and fluorinating agents FOOF and KrF_2 , continuous liquid-liquid extractions, and various other systems to study the chemical properties of the transactinides are also being made. Separations based on the difference in chemical properties between actinides and transactinides will also be useful for separations prior to studies of nuclear properties.

Nuclear Properties

The use of a large ^{254}Es target (400 g/cm^2) and transfer reactions from neutron-rich light heavy ions ($Z \leq 20$) should allow production of a large number of new neutron-rich heavy element isotopes for study. Those predicted to be produced with cross sections greater than a nanobarn are shown as cross-hatched areas in Fig. 3. Compound nuclei for reactions of ^{18}O with ^{248}Cm , ^{252}Cf and ^{254}Es are given in parentheses. Estimated production rates and half lives for some of these isotopes are given in Table IV. Spontaneous fission (SF) will ultimately limit the production of the heavy element isotopes and the half lives for SF still cannot be predicted accurately, especially for nuclides containing an odd proton or neutron. It has been shown [15] that the odd particle can greatly inhibit SF. Hindrance factors, particularly for high spin particles, can be as high as 10^7 as shown in Fig. 4. Half lives for odd-odd isotopes are expected to be especially hindered.

Recently, ^{260}Md , which has both an odd proton and an odd neutron, has been produced [4] and shown to have an SF half life of at least 30 days, confirming this extra hindrance. This makes it hopeful that longer-lived isotopes of No(102) and especially of Lr(103) can be produced. Studies of their SF properties will be particularly interesting since SF properties change very rapidly in the region of $Z = 100$ and $N = 158$. The most probable mass division is symmetric in this region and unusually high total kinetic energies which approach the Q value for fission have been observed. There is also some indication of two different kinds of symmetric mass division, one with high and one with "normal" total kinetic energy. Production of longer-lived isotopes of these elements should also make studies of chemical properties much more viable if the cross sections are large enough.

One of the problems with transfer reactions is that they are not specific and a wide variety of products results. Identification of the isotopes which spontaneously fission is especially difficult since the fission process rather effectively destroys the information on the Z and A of the fissioning species. A new detector system, the Heavy Element Fission Tracker (HEFT) is being developed to measure half lives as short as nanoseconds and simultaneously identify the fissioning isotope. It consists of an array of modules each consisting of four elements (Fig. 5) to characterize the energy deposited by the fission fragment. The first records the x-y position of the fragment and provides a start signal. The second gives the energy loss. The third again provides an x-y position and a timing signal. The fourth element stops the fragment and gives information concerning its residual kinetic energy. The mass of the fission fragment is then determined from the kinetic energy and velocity and the atomic number from the rate of energy loss. An array of perhaps five pairs of these modules would permit efficient detection of coincident fission fragments. By summing information on the mass and charge of coincident fragments, it should be possible to make definitive assignments from some 10 to 100 recorded events.

Superheavy Elements

Use of the ^{254}Es target and ^{48}Ca projectiles would give a compound nucleus with 183 neutrons, closer to the postulated stable shell at 184 neutrons than has previously been achieved. In addition to fast chemical separation techniques, SASSY-II, a higher efficiency version of the Small Angle Separating System will be used to search for Superheavy Elements (SHE's) formed in such compound nucleus reactions. Half lives as short as

microseconds can be detected with crude mass resolution. Final identification will depend on observation of alpha decay chains resulting in previously identified nuclides.

Experiments with small ^{254}Es targets show production of much larger amounts of trans-einsteinium isotopes than is possible with ^{248}Cm , but larger targets will be required for studies of the chemical properties of elements beyond lawrencium, element 103. Studies of nuclear properties to date indicate that there should be longer-lived, neutron-rich isotopes with odd protons and neutrons which will be accessible. The possibilities for extending our understanding of both the nuclear and chemical properties of the heaviest element appears extremely promising. There is also a unique opportunity for the collaboration of diverse teams of researchers from foreign countries as well as the U. S. in exploring this frontier of nuclear and chemical science.

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**Table 1. Mainline HFIR-TRU production
(elements 96-100)**

Isotope	Half-life (principal decay mode)	Production (amount/year)
Curium-248	3.397 x 10 ⁵ years (alpha)	150 mg
Berkelium-249	320 days (beta)	50 mg
Californium-249	350.6 years (alpha)	from ²⁴⁹ Bk decay
Californium-252	2.64 years (alpha) ^a	500 mg
Einsteinium-253	20.4 days (alpha)	2 mg
Einsteinium-254	275.7 days (alpha)	4 μg
Fermium-257	100.5 days (alpha)	1 pg

^a ²⁵²Cf has a neutron output from spontaneous fission of 2.3 x 10¹² neutrons/s/g.

Table II

PRODUCTION RATES FOR CHEMICAL STUDIES
 (400 $\mu\text{g}/\text{cm}^2$ ^{254}Es , 1 p μA beam)

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

Table III. Ratio of Heavy Actinide Yields from Reactions of ^{22}Ne with ^{254}Es Relative to ^{248}Cm Targets.

			<u>Transfer</u>
Fm: (254)	$\frac{3 \times 10^3 \mu\text{b}}{3 \mu\text{b}} \approx 10^3$		p vs. Be
Md: (256)	$\frac{10^3 \mu\text{b}}{0.01 \mu\text{b}} \approx 10^5$		He vs. B
No: (259)	$\frac{4 \mu\text{b}}{<0.03 \mu\text{b}} \geq 10^2$		Li vs. C
Lr: (259,260)	$1 \mu\text{b}$ No data from ^{248}Cm		Be vs. N

Table IV

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

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

Figures

- Fig. 1. Changes of composition during irradiation of ^{252}Cf to produce ^{254}Es .
- Fig. 2. Excitation functions for Es and Fm isotopes (from Ref. 11).
- Fig. 3. Portion of table of isotopes showing neutron-rich trans-Am isotopes. Hatched regions indicate new actinide isotopes postulated to be produced with $>1\text{-nb}$ cross sections from transfer reactions to a $400\ \mu\text{g}/\text{cm}^2$ ^{254}Es target with a projectile beam of $1\ \mu\text{A}$. Compound nuclei for reactions of ^{18}O with ^{248}Cm , ^{252}Cf , and ^{254}Es are shown in parentheses.
- Fig. 4. Odd proton and odd neutron hindrance factors.
- Fig. 5. Detector module to be used in the Heavy Element Fission Tracker (HEFT) System. The four active regions are used to determine the position, velocity, rate of energy loss, and total energy for fission fragments.

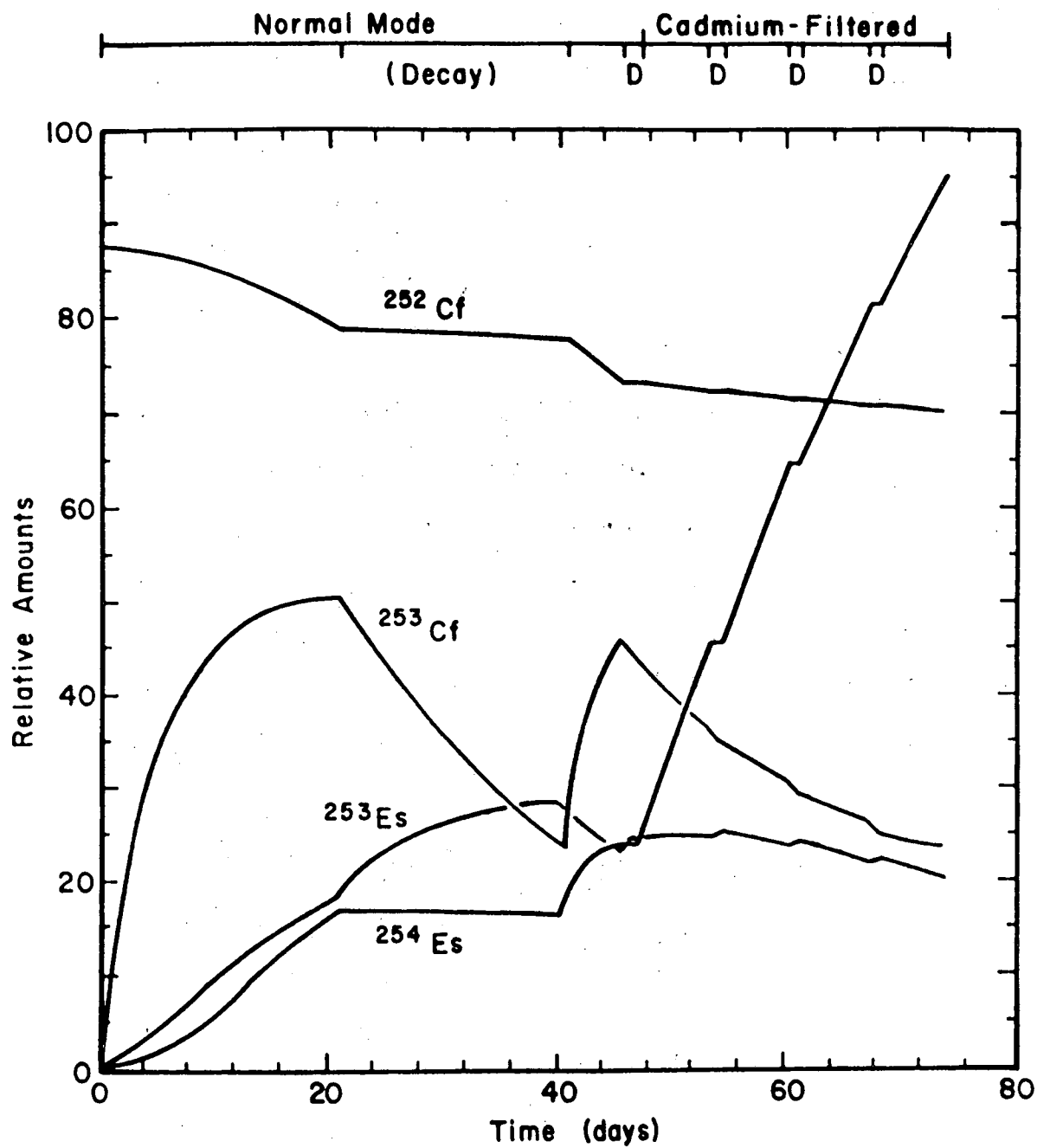


Fig. 1. Changes of Composition During Irradiation of ^{252}Cf to Produce Large Einsteinium Sample

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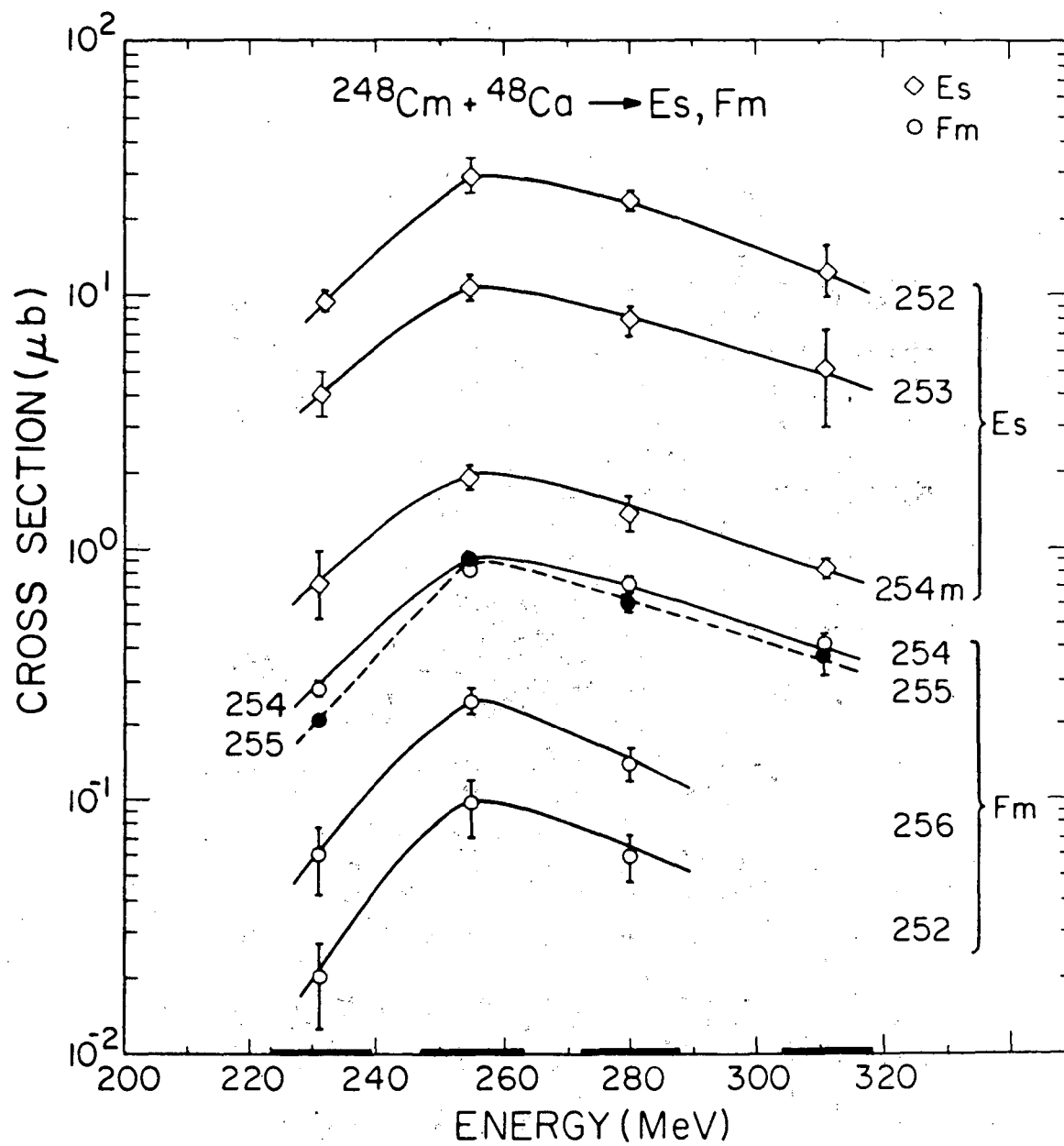


Fig. 2. Excitation Functions for Es and Fm Isotopes

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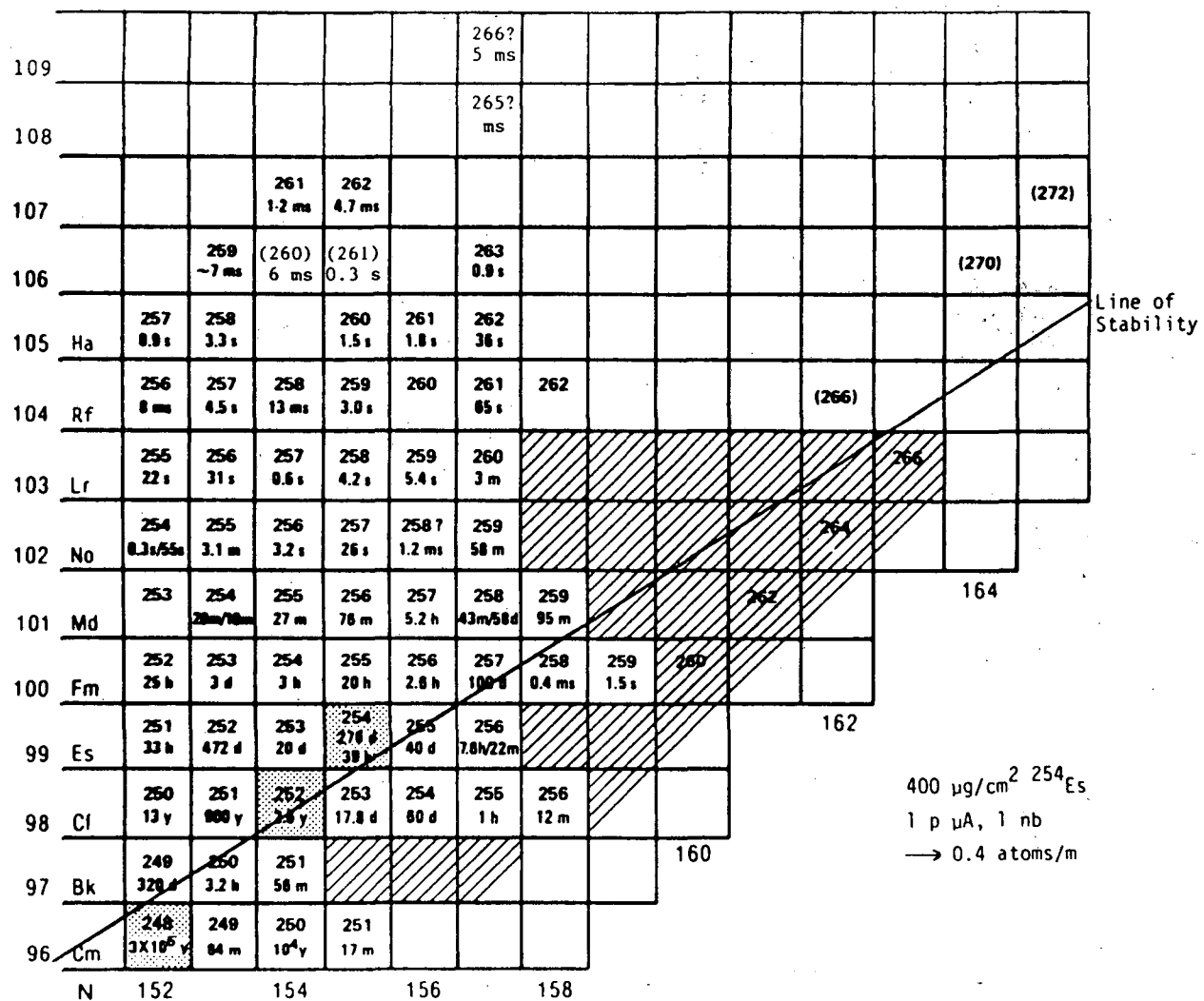


Fig. 3

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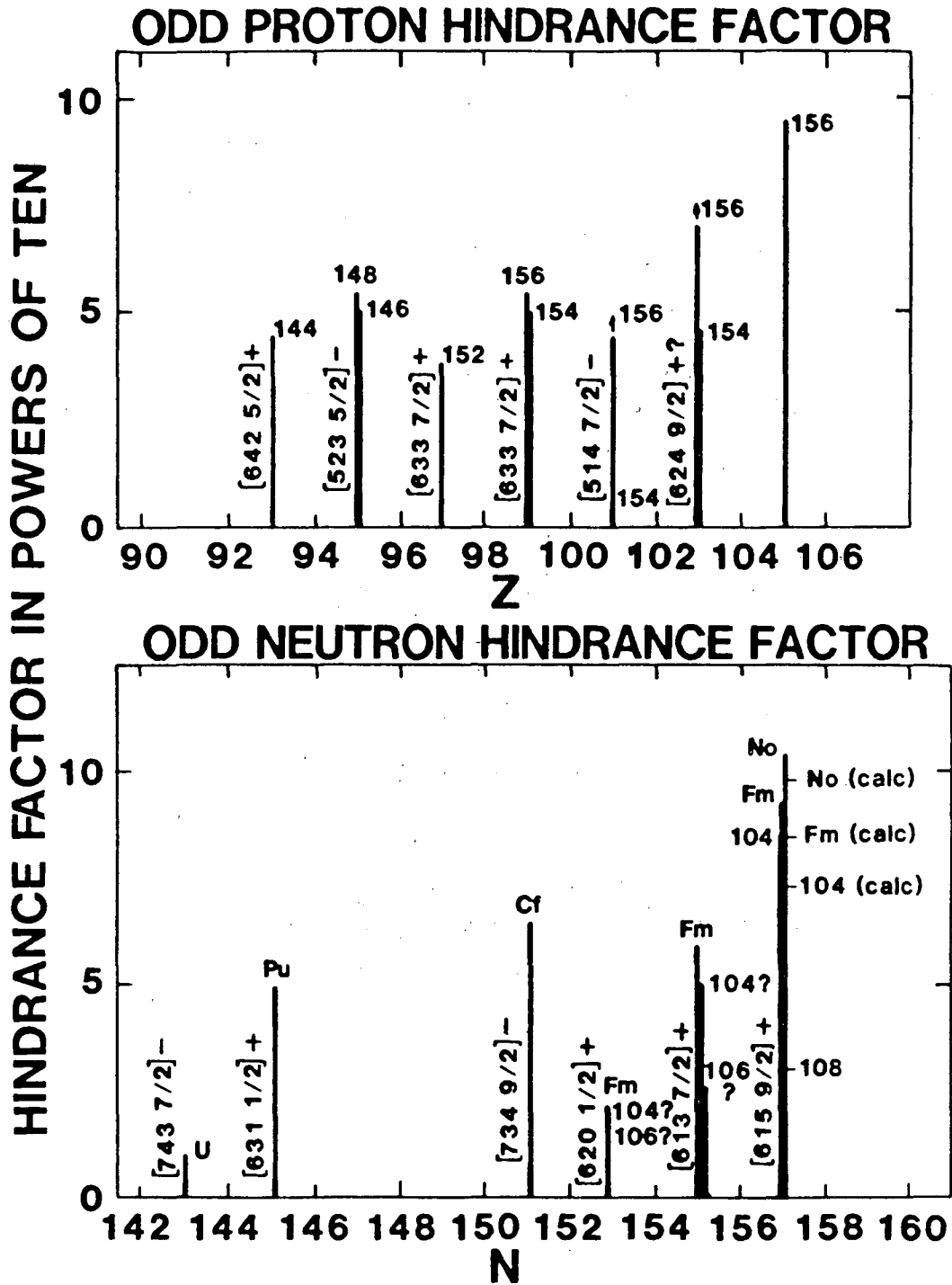


Fig. 4. Odd Proton and Neutron Hindrance Factors

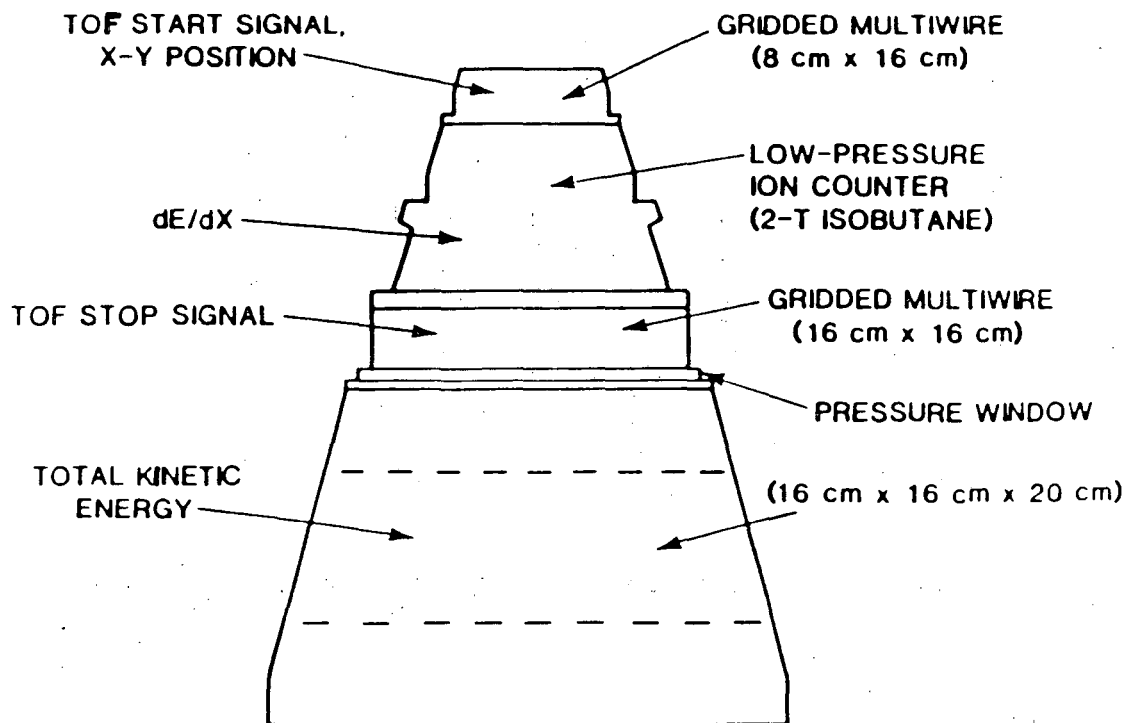


Fig. 5. Detector module used in the Heavy Element Fission Tracker (HEFT) System. The four active regions are used to determine the position, velocity, rate of energy loss, and total energy for fission products.

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