CONF-850836--11

PROPOSED PRODUCTION OF A LARGE (~40-μg) SAMPLE OF ^{254}Es*

J. E. Bigelow
C. W. Alexander
L. J. King

Chemical Technology Division
OAK RIDGE NATIONAL LABORATORY
P. O. Box X
Oak Ridge, TN 37831

CONF-850836--11

DE86 008969

MASTER

For presentation at a Symposium on "Heavy Element Research," at the AIChE 1985 Summer National Meeting, Seattle, Washington, August 25-28, 1985

By acceptance of this article, the publisher or recipient acknowledges the U.S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering the article.

*Research sponsored by the Office of Basic Energy Sciences, U.S. Department of Energy, under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

PROPOSED PRODUCTION OF A LARGE (~40-μg) SAMPLE OF ^{254}Es

J. E. Bigelow, C. W. Alexander, and L. J. King

1. Introduction

Much research with the heaviest elements in the chart of the nuclides (Fig. 1) consists of extended periods of bombardment of accelerator targets to make new nuclides one atom at a time, followed by painstaking effort to isolate and detect the products and to measure their properties before they disappear. A recent workshop sponsored by the National Research Council[1] has made it clear that the key to further substantial progress in heavy element research is the expanded use of 276-day ^{254}Es as target material. Einsteinium-254 has the greatest mass and charge of any nuclide that can be produced in the required multimicrogram quantities in the foreseeable future.

Four major laboratories [Lawrence Berkeley Laboratory, Lawrence Livermore National Laboratory, Los Alamos National Laboratory, and Oak Ridge National Laboratory (ORNL)] active in transplutonium research have collaborated to propose a major new thrust in transplutonium research that will require an order of magnitude more ^{254}Es than is normally available. This project, called LEAP (an acronym for Large Einsteinium Activation Program)

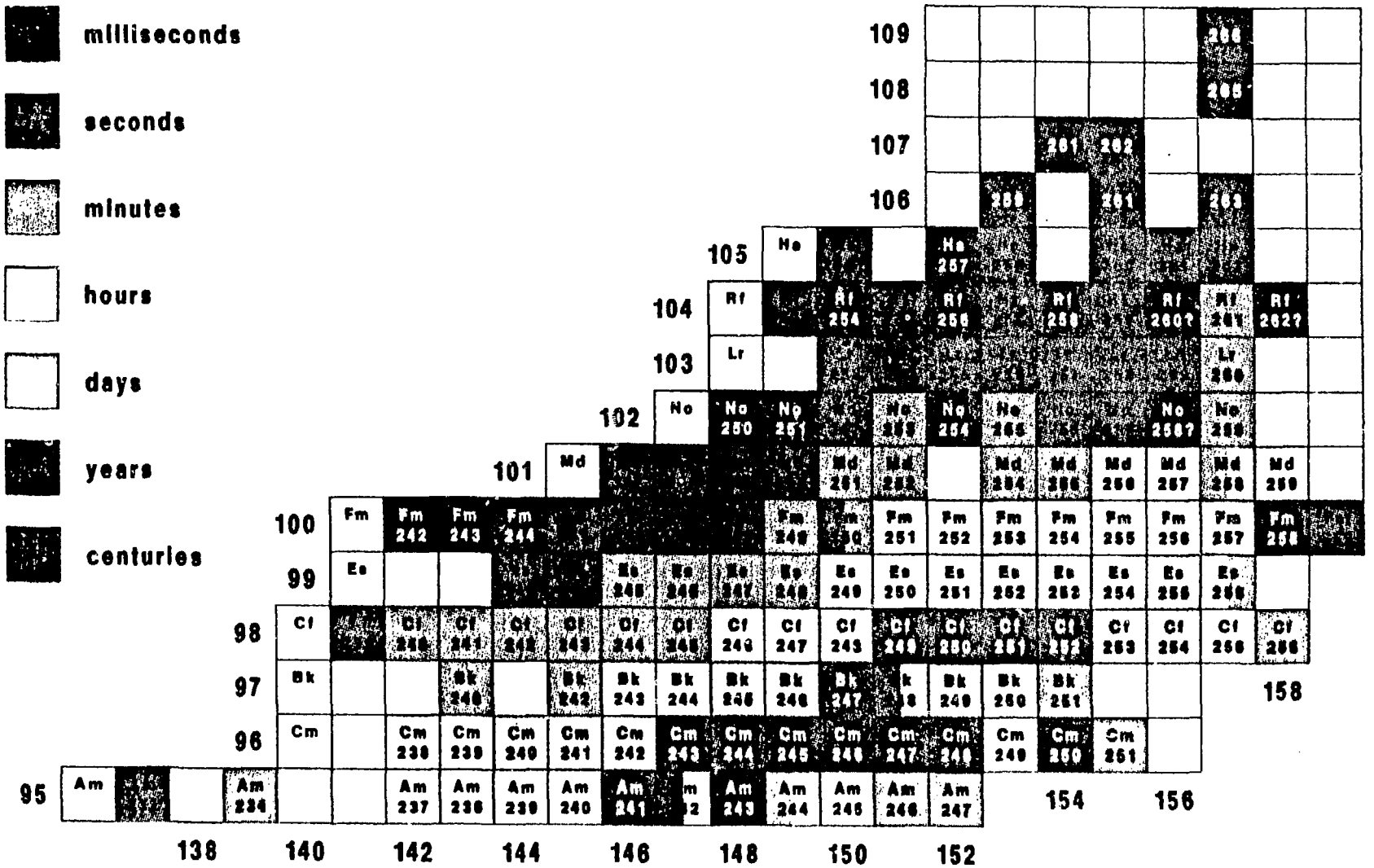

Fig. 1. Chart of the transplutonium nuclides.

has goals of determining the inorganic chemistry and nuclear chemistry and physics of the transeinsteinium elements through atomic number 109, plus a search for superheavy elements. The proposed research is discussed in detail in another paper in this symposium.[2]

LEAP is based on using ~40 μg of ^{254}Es as a target for heavy-ion accelerators. The Transuranium Processing Plant (TRU) of the Chemical Technology Division of ORNL has been given the task of determining the feasibility of producing a 40-μg sample of ^{254}Es and, if later requested, of actually producing the sample.

This task, which has been under way for several years, is directed toward three areas of investigation: (1) experimental determination of the neutron cross sections of certain transplutonium isotopes important to the production of ^{254}Es; (2) selection of a ^{254}Es production scheme; and (3) development of the necessary hardware, followed by an actual test irradiation.

2. Experimental Determination of Neutron Cross Sections

Planning for production of the LEAP sample places a premium on the accurate determination of a number of neutron cross sections. The design of experiments to optimize production of

^{254}Es using ^{252}Cf as a starting material is dependent on a knowledge of all cross sections in both the 253 and 254 mass chains.

Measurements of some of these cross sections were made by Harbour and MacMurdo[3,4] in the early 1970s. However, yields of the 276-day ^{254}Es in targets at TRU were only about 10% of the amount predicted by transmutation calculations using their cross sections.[5] Therefore, an experimental program was required to reevaluate the pertinent neutron cross sections.

The cross sections of interest are those for the production reaction ^{253}Es(n,γ)^{254g}Es (see Fig. 2). Also needed are cross sections for the side reaction ^{253}Es(n,γ)^{254m}Es, an isomeric state which beta decays with a 39-h half-life to ^{254}Fm, and for the parasitic reactions ^{254g}Es(n,γ)^{255}Es and ^{254g}Es(n,fission). (Note that ^{254g}Es is referred to as ^{254}Es elsewhere in this paper.) Multinanogram samples of ^{253}Es and ^{254}Es were prepared at TRU and at the Transuranium Research Laboratory (TRL) of ORNL and then irradiated for short periods in the High Flux Isotope Reactor (HFIR) or the Oak Ridge Research Reactor (ORR). Members of the ORNL Chemistry and Chemical Technology Divisions determined two-group cross sections[6,7] for the above reactions, as shown in Table 1. Part of the technique for performing this type of experiment involves surrounding some of the samples with

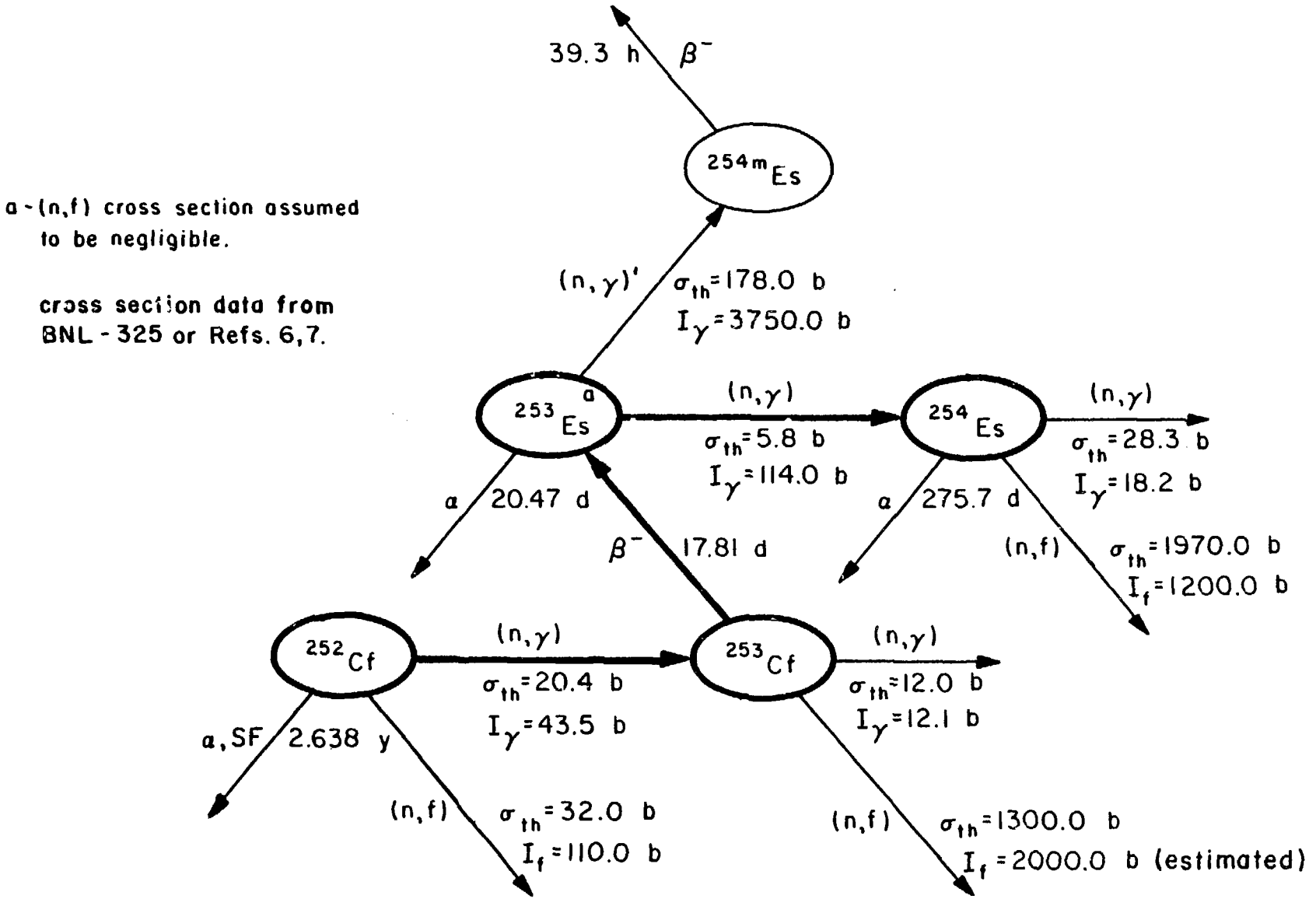

Fig. 2. Production of ^{254}Es by irradiation of ^{252}Cf.

Table 1. Neutron cross sections for the 253 and 254 mass chains[a]

Reaction	σ_{th}[b] (barns)[c]	I[d] (barns)[c]
$^{253}Es(n,\gamma)^{254g}Es$	5.8 ± 0.7	114 ± 7
$^{253}Es(n,\gamma)^{254m}Es$	178 ± 15	3750 ± 200
$^{254g}Es(n,\gamma)^{255}Es$	28.3 ± 2.5	18.2 ± 1.5
$^{254g}Es(n,fission)$	1970 ± 200	1200 ± 250

[a] J. Halperin et al.[6,7]

[b] 2200-m/s cross section.

[c] 1 barn = 10^{-28} m^2.

[d] I is the resonance integral, which is a cross section averaged over the 1/E neutron distribution.

a cadmium wrapper. The cadmium acts like a filter, strongly absorbing thermal neutrons but passing epithermal neutrons with little attenuation. Thus, the resonance integral is determined from the transmutation rate of the wrapped samples. The thermal cross section is determined from the difference in transmutation rates between the wrapped samples and the bare ones.

This two-group treatment of reactor neutron flux spectra has been remarkably successful[5] for predicting reaction rates in well-moderated nuclear reactors, such as the ORR and the HFIR target island (where the ^{254}Es will be produced). As a result of the reevaluation of these cross sections, the predicted values agree with the measured content of ^{254}Es in standard HFIR/TRU production targets within 20%.

3. ^{254}Es Production Scheme

Several ^{254}Es production schemes were analyzed in which virtually the entire inventory of ^{252}Cf at TRU (~1 g) would be fabricated into special targets and irradiated in the HFIR. The ^{252}Cf would be transmuted successively to ^{253}Cf, ^{253}Es, and, finally, ^{254}Es by two neutron captures separated by a beta decay, as shown in Fig. 2. Note that there is an isomeric state, ^{254m}Es, which is also produced in the irradiation in much higher yields. Unfortunately, this nuclide is a short-lived beta emitter that does not undergo isomeric transition and thus makes no contribution to the production of the desired ground-state isomer, ^{254g}Es. The irradiation conditions would include a period in the normal flux in the HFIR target island followed by a period in a cadmium-filtered flux. The irradiated targets

would then be returned to TRU for isolation and purification of the einsteinium, using well-known processes and techniques.[8,9] A decay period of ~6 months would be required to permit the 20-day ^{253}Es to decay sufficiently (to approximately equal activities) to allow preparation of accelerator targets using the 276-day ^{254}Es. The entire production effort, including fabrication of the ^{252}Cf HFIR targets and decay of the interfering ^{253}Es, would require ~15 months.

The preferred target material for producing the ^{254}Es is ^{252}Cf (see Fig. 2). The heavier nuclides, ^{253}Cf and ^{253}Es, have half-lives too short to permit their separate production and refabrication into targets. On the other hand, nuclides lighter than ^{252}Cf would require more target rods to contain the requisite amount of feed and the reactor time needed for the conversion would be increased. Also, the additional material would complicate the subsequent chemical processing operations.

The idea of using cadmium-filtered neutrons to prepare this sample was proposed a number of years ago; however, reliable two-group cross sections have only become available recently. Using the new data, an optimized irradiation scheme has been developed which takes into account the HFIR operating schedule as well as the time scale of the transmutation reactions. The resulting schedule is shown in Fig. 3, where the shaded bars

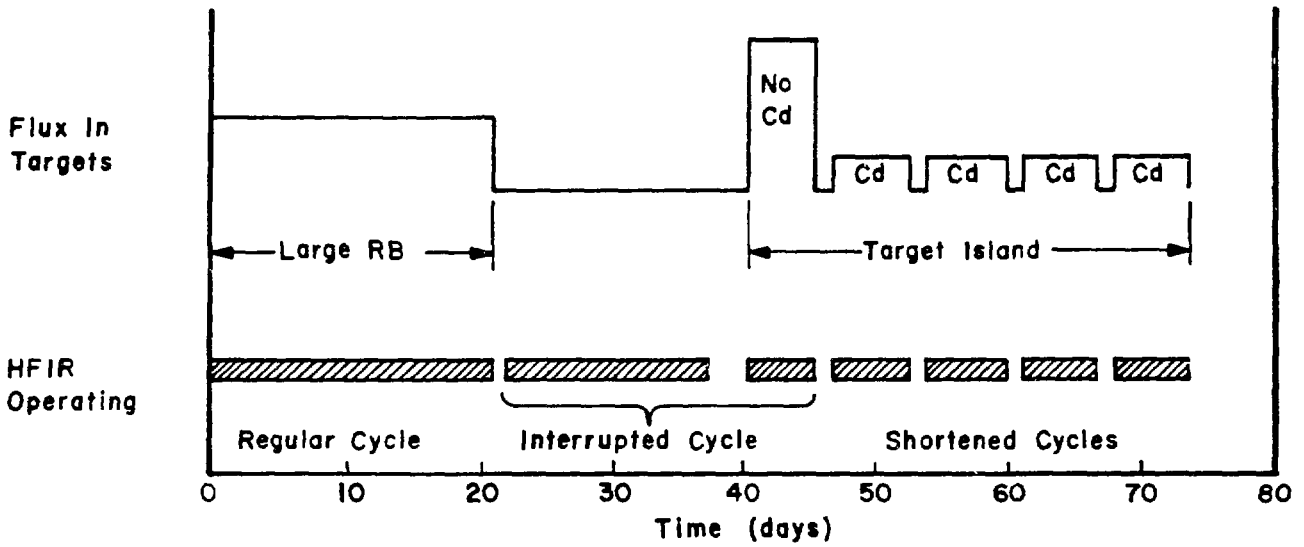

Fig. 3. Proposed irradiation schedule for ^{254}Es production in ^{252}Cf targets.

denote HFIR operation and the gaps signify downtime for refueling. The upper curve indicates what is happening to the ^{252}Cf targets: During the first 21 days, the targets are located in the beryllium reflector where they are exposed to a moderate amount of flux. The targets are removed from the reactor for the next ~20 days and allowed to cool to permit ^{253}Cf to decay to ^{253}Es. Then, they are inserted into the target island for ~5 days, where they are exposed to the maximum thermal flux. Finally, after a short (~36-hour) decay period, the targets are placed in the cadmium filters and exposed for 24-28 days to the cadmium-filtered flux. Several shutdowns are necessary during this period to renew the cadmium filters as well as to replace spent fuel assemblies.

During the seventy-plus days of the irradiation program, significant changes in composition are occurring in the target rods, as illustrated in Fig. 4. When reading Fig. 4, keep in mind that ^{252}Cf ranges from 0.7 to 0.9 g, ^{253}Cf and ^{253}Es peak at 5 and 3 mg, respectively, and that ^{254}Es is measured in μg, rising to 90 μg at reactor discharge (although only about half of this amount is ultimately available as product because of the final decay period). In the initial irradiation period, a supply of ^{253}Cf is accumulated in the targets. Although the ^{253}Cf continually decays to ^{253}Es, the ^{253}Es is partly consumed through

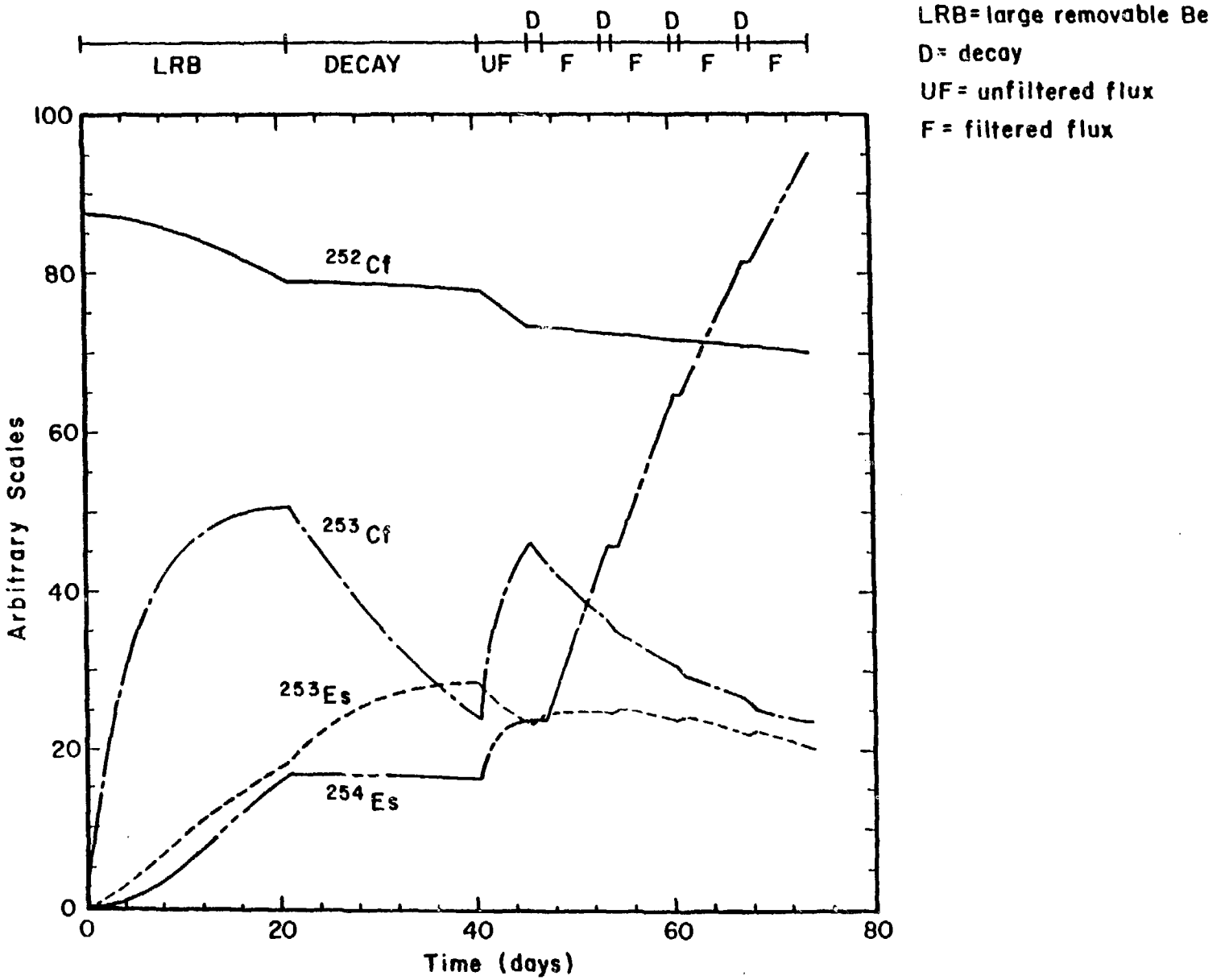

Fig. 4. Changes of composition during irradiation to produce large einsteinium sample.

neutron capture during the period that the targets are in the reactor. Thus, the ^{253}Es content grows more rapidly during the intentional decay period. The short (5-day) irradiation in the target island restores the maximum level of ^{253}Cf so that a nearly constant supply of ^{253}Es is available during the final phase of the irradiation. This final phase, the cadmium-filtered irradiation, results in a remarkable increase in the ^{254}Es.

The sharp increase in the rate of production of ^{254}Es is explained by an examination of some ratios of cross sections. The time-dependence of the target composition is quite complex, but a simple relationship governs the limiting abundance of ^{254}Es relative to ^{253}Es:

$$\frac{N_4^\infty}{N_3^\infty} \approx \frac{\sigma_{c,3}^{eff}}{\sigma_{a,4}^{eff}} ,$$

where

N_i^∞ = number of atoms of ^{253}Es or ^{254}Es after an extended period in the reactor,

σ^{eff} = effective cross sections of the two isotopes,

$\sigma_{c,3}$ = capture cross section of ^{253}Es to produce ^{254}Es, and

$\sigma_{a,4}$ = absorption cross section of ^{254}Es, which is the sum of all capture and fission events.

If the flux is highly thermalized, we can substitute σ^{th} for σ^{eff} and the limiting ratio becomes (data from Table 1):

$$\frac{N_4^\infty}{N_3^\infty} \approx \frac{5.8}{1970 + 28.3} = 2.9 \times 10^{-3} .$$

In a region where cadmium has been used to filter out all thermal neutrons, we can substitute the resonance integral, I, for the σ_{eff}. The limiting ratio then becomes:

$$\frac{N_4^\infty}{N_3^\infty} \approx \frac{114}{1200 + 18.2} = 9.4 \times 19^{-2} ,$$

which would be a 32-fold enhancement. There are practical reasons why an enhancement of this magnitude cannot be achieved, but the results of the studies reported here indicate that a 10-fold enhancement may be possible.

Several computer calculations were performed to determine the sensitivity of the production scheme to the cross-section values in the 253 and 254 chains according to the reported uncertainties in their measured values. In the case of the ^{253}Cf(n,fission), where the resonance integral has never been measured but only estimated, both the thermal fission cross section and the resonance integral were arbitrarily varied from 50 to 200% of their nominal values. While the calculated amount of ^{254}Es produced did fluctuate, the amount calculated for most combinations exceeded the requested 40 μg, thereby indicating a fair probability of success.

Although a production scheme for producing a large sample of ^{254}Es has been identified, numerous areas of concern still exist. The use of cadmium filters entails a substantial extrapolation of the two-group neutron cross-section treatment that has been verified through our considerable experience with transplutonium element production in the HFIR. We do not know whether our transmutation model will be valid under macroscopic conditions in the reactor when the cadmium filters are used. Further, there are strong interactions between the intense neutron flux normally present in the target island and the cadmium, which is a very effective neutron absorber because of the 20,000-barn thermal cross section of ^{113}Cd. Somehow, these conflicting factors must be reconciled to achieve a reasonable balance between the criticality of the reactor, the effective lifetime of the HFIR fuel assembly, and the burnup of the ^{113}Cd in the filters.

The cadmium in the target island of HFIR constitutes a large thermal neutron poison, similar in effect to the reactor control rods being inserted. Therefore, the quantity of cadmium must be limited to that which will permit the HFIR to go critical ($k_{eff} = 1$); at the same time, it must be sufficient to adequately filter the thermal flux in the ^{252}Cf targets to minimize thermal fission of the ^{254}Es.

For purposes of optimizing the production scheme, it was assumed that the HFIR would go critical, the fuel cycle duration would be 6 days, and there would be enough ^{113}Cd to last through the fuel cycle and adequately filter the thermal neutron flux. Attempts are being made to confirm these assumptions. One- and three-dimensional Monte Carlo[10] and Discrete Ordinates[11] neutron transport calculations are being made to more carefully evaluate reactor operating parameters. Very preliminary calculations indicate that the assumptions are valid and, possibly, conservative. In addition, ^{113}Cd burnup experiments are being conducted to confirm the calculations of the model predicting the burn-through.

4. Hardware Development and Test Irradiation

A prototype LEAP target assembly has been designed and fabricated by the Engineering Technology Division of ORNL.[12] A LEAP target assembly is quite different from a normal HFIR target assembly. First, the LEAP target rod is loaded with ^{252}Cf rather than Am-Cm. The ^{252}Cf is also concentrated in the middle 250 mm of the rod in order to take advantage of the peak of the axial neutron flux distribution. The LEAP target assembly also has provisions for using a cadmium filter. This capability

necessitated an increase in the overall dimensions of the target assembly to the degree that one LEAP target assembly occupies the same space as three normal HFIR target assemblies. To accommodate the LEAP target assembly, a trefoil geometry was designed for the aluminum support structure. The trefoil geometry accomplishes two tasks: (1) it mates with the hexagonal-shaped upper ends of normal HFIR target assemblies, and (2) it reduces the amount of water surrounding the ^{252}Cf (this reduces the thermalization of epithermal neutrons). Both the cadmium filter and the ^{252}Cf target can be removed from the LEAP target assembly by remote handling techniques.

The physical specifications for the prototype LEAP target assembly are given in Table 2, and a cross-sectional view is shown in Fig. 5. The actual prototype target assembly is shown in Figs. 6 and 7. For comparative purposes, a normal HFIR target rod is shown in Fig. 8.

A test irradiation is planned that will consist of installing three of the cadmium filters within their trefoil holders in the HFIR, along with special target rods containing several small californium samples and flux monitors. The HFIR target island arrangement is shown in Fig. 9. There may be additional standard HFIR target rods in other positions also containing

Table 2. Specification for LEAP target assembly

Target Rod	
Target material	$^{252}Cf_2O_3$-Al matrix; Al clad
^{252}Cf loading	0.3 g/rod
Active length of ^{252}Cf region	~250 mm (10 in.)
Target rod OD	9.19 mm (0.362 in.)
Inner Water Gap	1.02 mm (0.040 in.)
Cadmium Filter Assembly	
Inner Cladding:	
Material	Stainless steel
ID	11.23 mm (0.442 in.)
OD	11.99 mm (0.472 in.)
Neutron Filter:	
Material	Cadmium
ID	11.99 mm (0.472 in.)
OD	17.07 mm (0.672 in.)
Outer Cladding:	
Material	Stainless steel
ID	17.07 mm (0.672 in.)
OD	17.83 mm (0.702 in.)
Outer Water Gap	1.52 mm (0.060 in.)
Trefoil Support Structure:	
Material	Aluminum
ID	20.9 mm (0.822 in.)
OD	Irregular
Cross-sectional area	374 mm^2 (0.585 $in.^2$)

ORNL DWG 85-13735

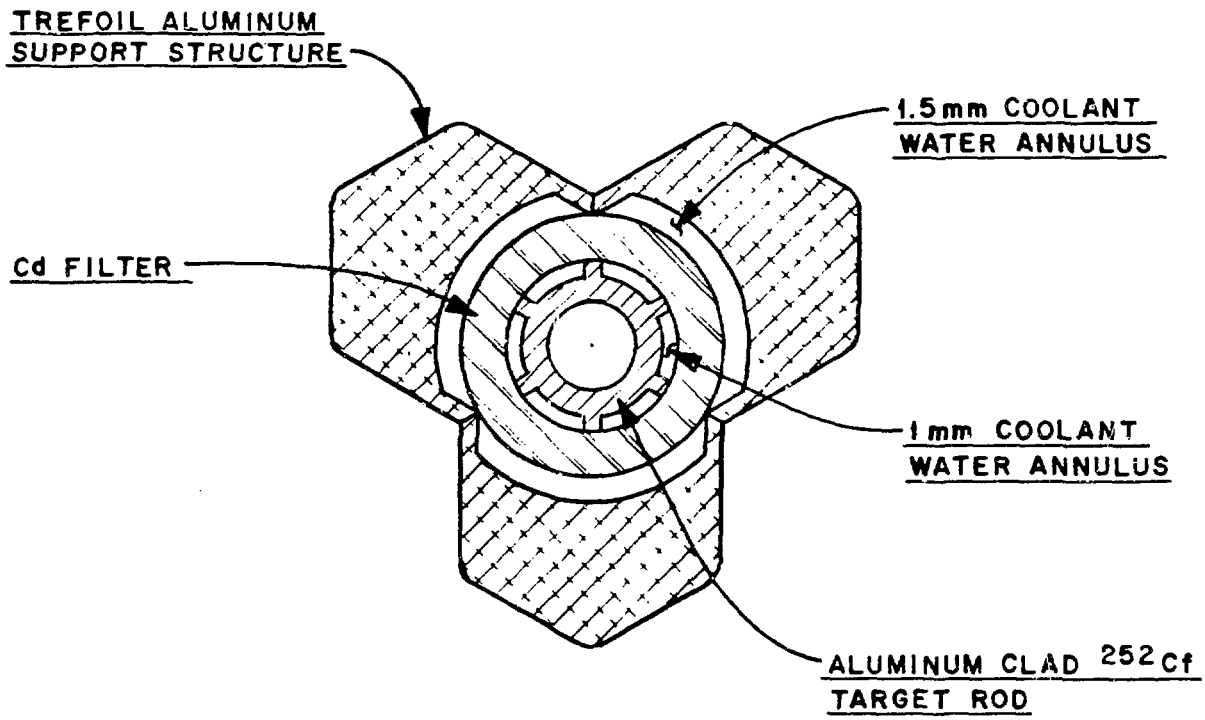

Fig. 5. LEAP target assembly.

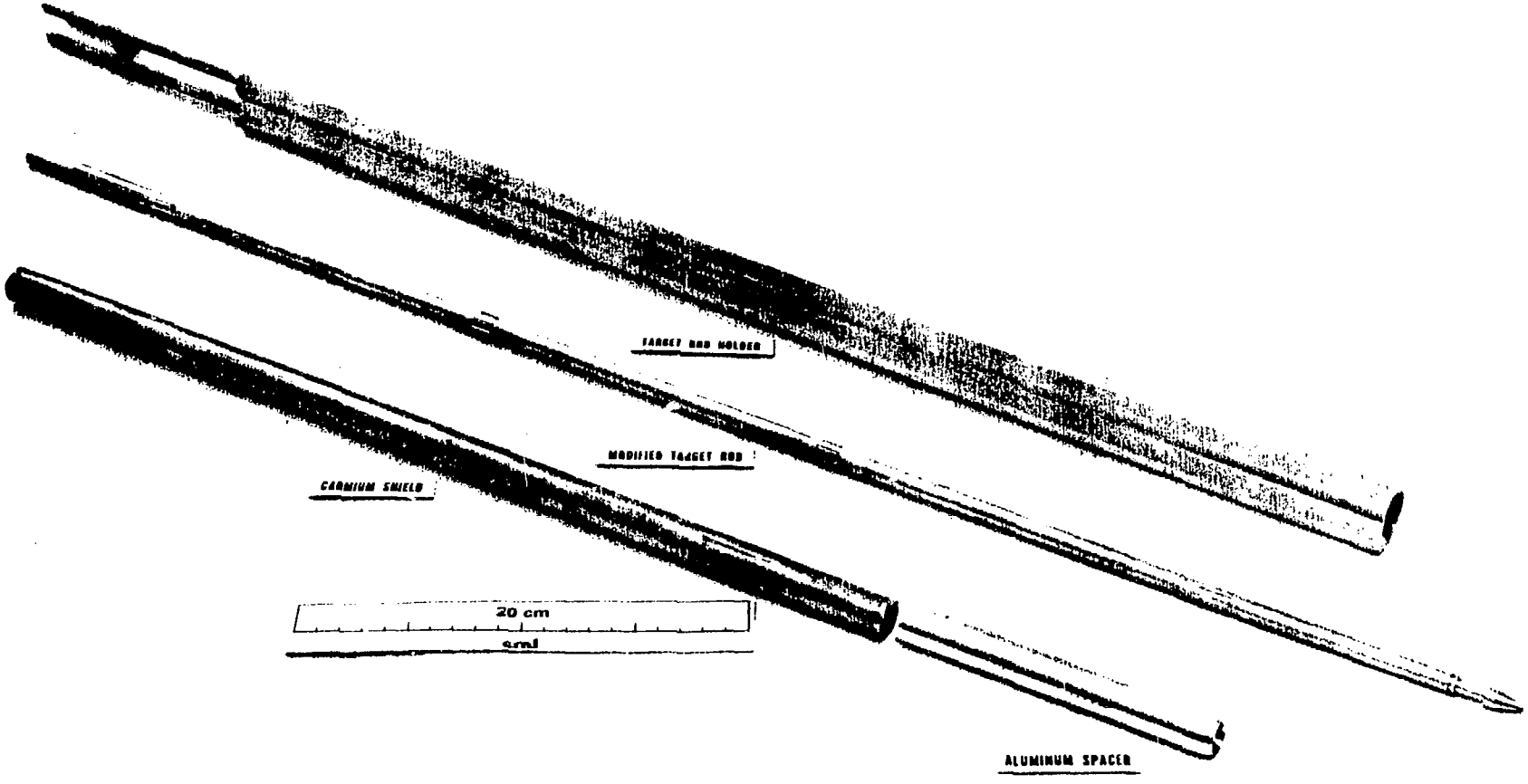

Fig. 6. HFIR LEAP irradiation capsule.

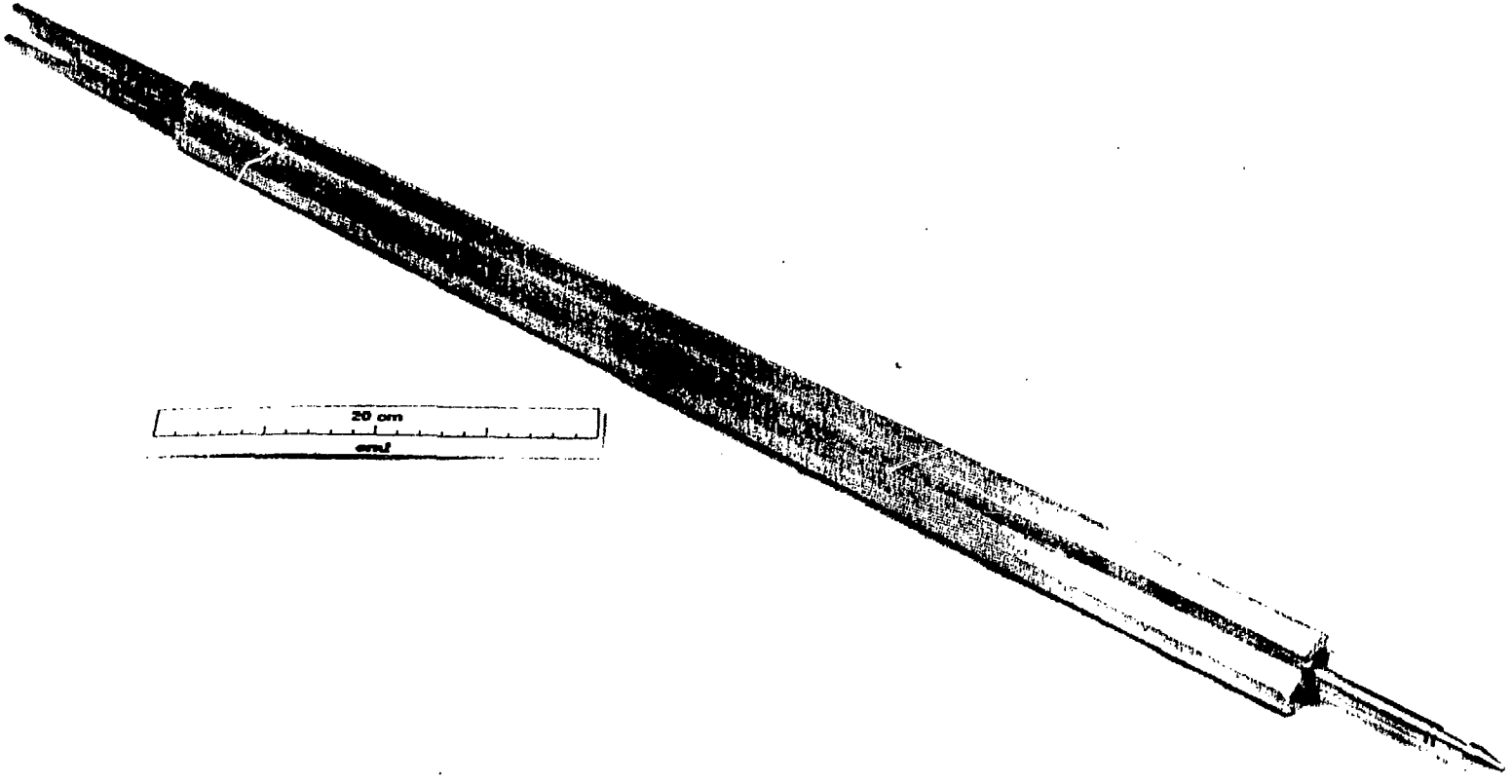

Fig. 7. HFIR LEAP target assembly.

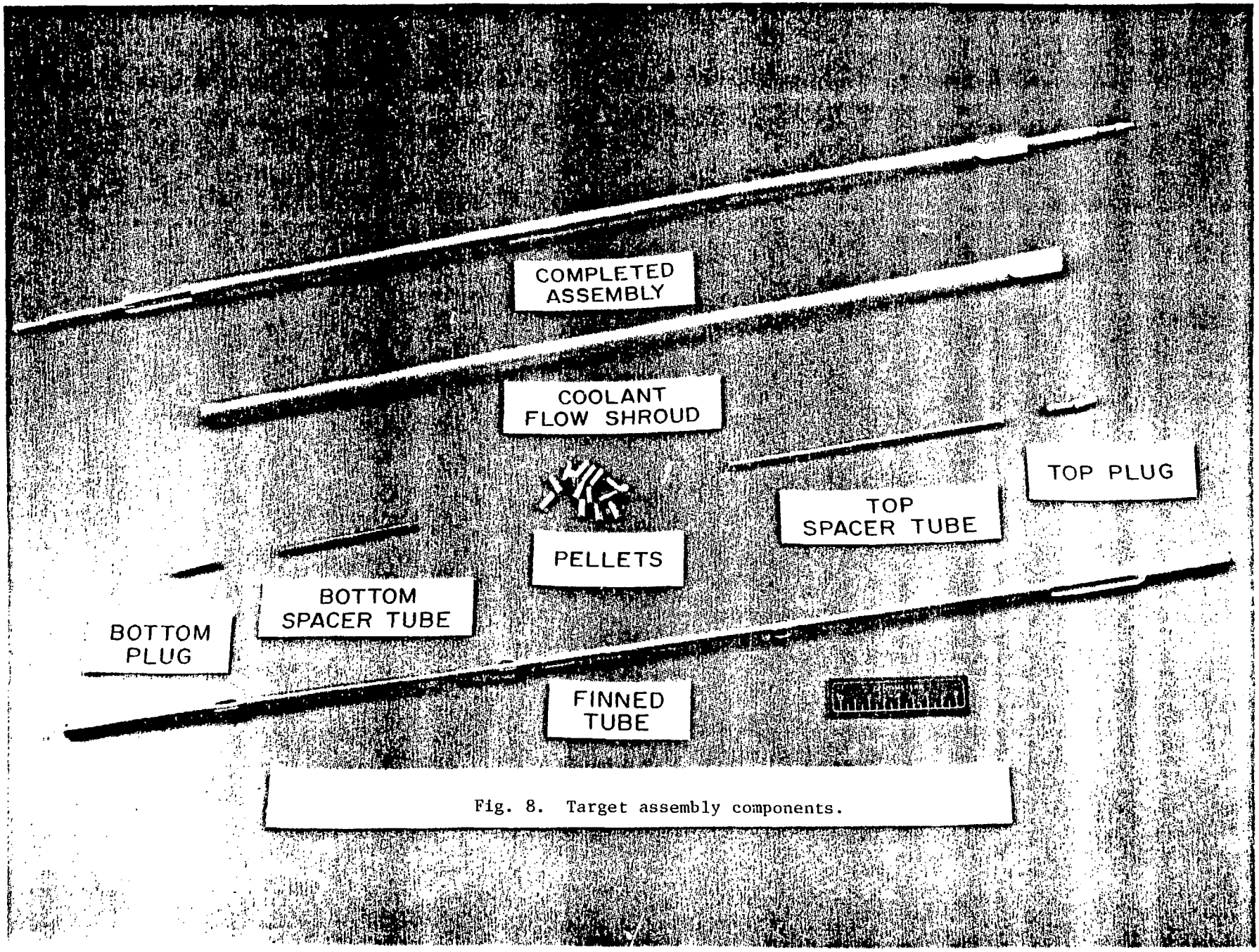

Fig. 8. Target assembly components.

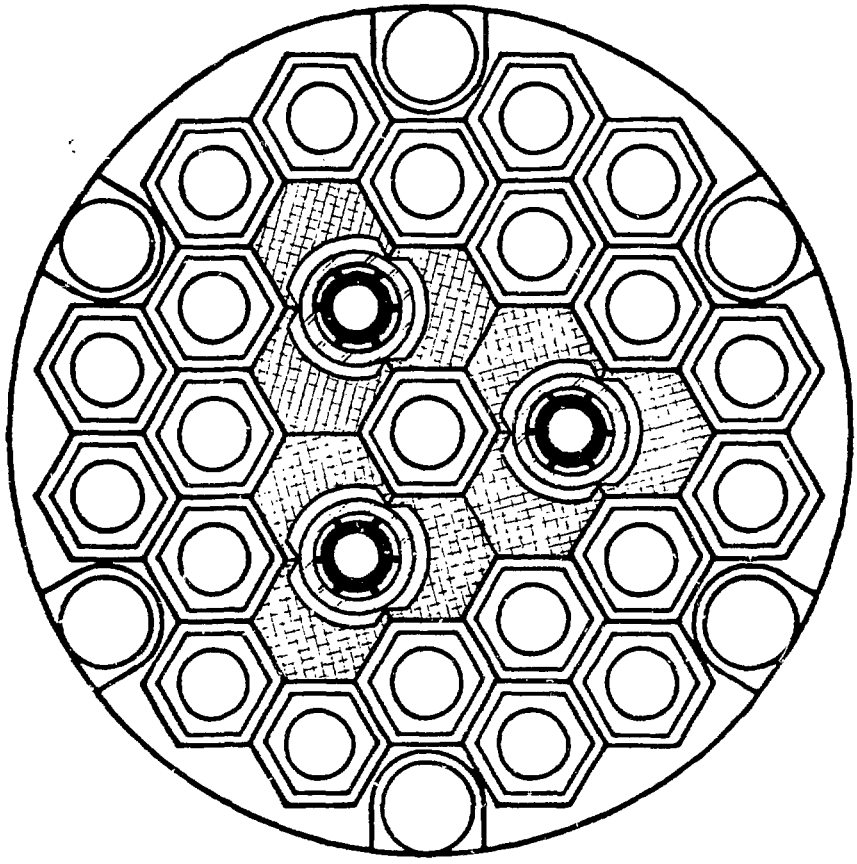

Fig. 9. Location of LEAP target assemblies in HFIR target island.

flux monitors. These assemblies will be irradiated at full power until the reactor has shut down or for a maximum of about 10 days. At this point, the assemblies will be withdrawn from the target island and a demonstration will be made of changing the cadmium filters remotely.

Information expected to be gained by a one-cycle test irradiation includes: (1) confirmation of the thermal-hydraulic design of the cadmium filters and the trefoil holders, (2) direct measurement of the perturbed fuel cycle length, (3) direct measurement of the Δk_{eff} of the cadmium filters, (4) demonstration of the capability to replace the cadmium filters in the trefoil holders, (5) additional check on the burnup of ^{113}Cd in the cadmium filters, (6) measurement of flux perturbations within the cadmium filters and in regions close to the LEAP target assembly, (7) an upper bound on the ^{253}Cf fission resonance integral, and (8) confirmation of the validity and accuracy of the einsteinium production model. In addition, two other pertinent factors that will be determined are the flux depression in the reflector and beam holes of HFIR and the extra reactor downtime required for core changes during the cadmium-filtered irradiation cycles and during subsequent burning of the residual short-life HFIR cores accumulated during the production program.

5. Conclusions

Reevaluation of neutron cross sections in the 253 and 254 mass chains has led to optimism for the possible production of an ~40-μg sample of the 276-day ^{254}Es for the LEAP project. A production scheme has been identified involving a carefully tailored irradiation of ~1 g of ^{252}Cf in specially designed targets in the HFIR at ORNL. Detailed neutron transport calculations are being made in order to confirm the assumptions used in the production scheme. The reprocessing of the irradiated targets in TRU will be accomplished by existing technology.

The production of such a large sample of ^{254}Es will require cadmium filters at certain stages of the irradiation. A prototype LEAP target assembly that can accommodate these filters has been designed and fabricated. A test irradiation which is planned later this year should yield valuable data as to the feasibility of producing the LEAP sample.

6. References

1. G. Friedlander (Chairman), Opportunities and Challenges in Research with Transplutonium Elements, Report of a Workshop, National Academy Press, Washington, DC (1983).

2. J. B. Wilhelmy, "LEAP Science," paper presented at the Symposium on Heavy Element Research, AIChE National Meeting, Seattle, WA (1985).

3. R. M. Harbour and K. W. MacMurdo, "Thermal Neutron Capture Cross Sections and Capture Resonance Integrals of ^{253}Es," J. Inorg. Nucl. Chem. 35, 1821-25 (1973).

4. K. W. MacMurdo and R. M. Harbour, J. Inorg. Nucl. Chem. 34, 449 (1972).

5. J. E. Bigelow, B. L. Corbett, L. J. King, S. C. McGuire, and T. M. Sims, "Production of Transplutonium Elements in the High Flux Isotope Reactor," Transplutonium Elements - Production and Recovery, J. D. Navratil (Ed.).

6. J. Halperin, R. L. Walker, G. D. O'Kelley, J. H. Oliver, J. E. Bigelow, J. T. Wiggins, J. B. Knauer, E. B. Cagle, and L. A Byrd, "The Thermal Neutron Capture Cross Section and Resonance Integral of ^{253}Es," Radiochim. Acta. 37, 57-61 (1984).

7. J. Halperin, G. D. O'Kelley, J. H. Oliver, J. E. Bigelow, and J. T. Wiggins, "Neutron Capture and Fission in ^{254g}Es," Nucl. Sci. Eng. 90, 298-303 (1985).

8. L. J. King, "Status of Transuranium Element Production," paper presented at the Symposium on Heavy Element Research, AIChE National Meeting, Seattle, WA (1985).

9. D. E. Benker, F. R. Chattin, E. D. Collins, J. B. Knauer, P. B. Orr, R. G. Ross, and J. T. Wiggins, "Chromatographic Cation Exchange Separation of Decigram Quantities of Californium and Other Transplutonium Elements," ACS Symposium Series, No. 161, pp. 161-71; Transplutonium Elements - Production and Recovery, J. D. Navratil and W. W. Schulz (Eds.) 1981.

10. L. M. Petrie and N. F. Landers, KENO V.a: An Improved Monte Carlo Criticality Program with Supergrouping, ORNL/NUREG/CSD-2/VI/R2 (1984).

11. N. M. Greene and L. M. Petrie, XSDRNPM-S: A One-Dimensional Discrete-Ordinates Code for Transport Analysis, ORNL/NUREG/CSD-2/VI/R2 (1984).

12. J. A. Conlin, ORNL, personal communication to J. E. Bigelow (1984).

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.