CONF-780159--1

MASTER

PRODUCTION OF EINSTEINIUM AND FERMIUM IN REACTORS

by

D. E. Ferguson

Oak Ridge National Laboratory*
Oak Ridge, Tennessee 37830

Presented at the Symposium Commemorating the 28th Anniversary of Elements 99 and 100 held on January 20, 1978

and published in the Proceedings

*Operated by Union Carbide Corporation under Contract W-7405-eng-26 with the U.S. Department of Energy.

Production of Einsteinium and Fermium in Reactors

D. E. Ferguson

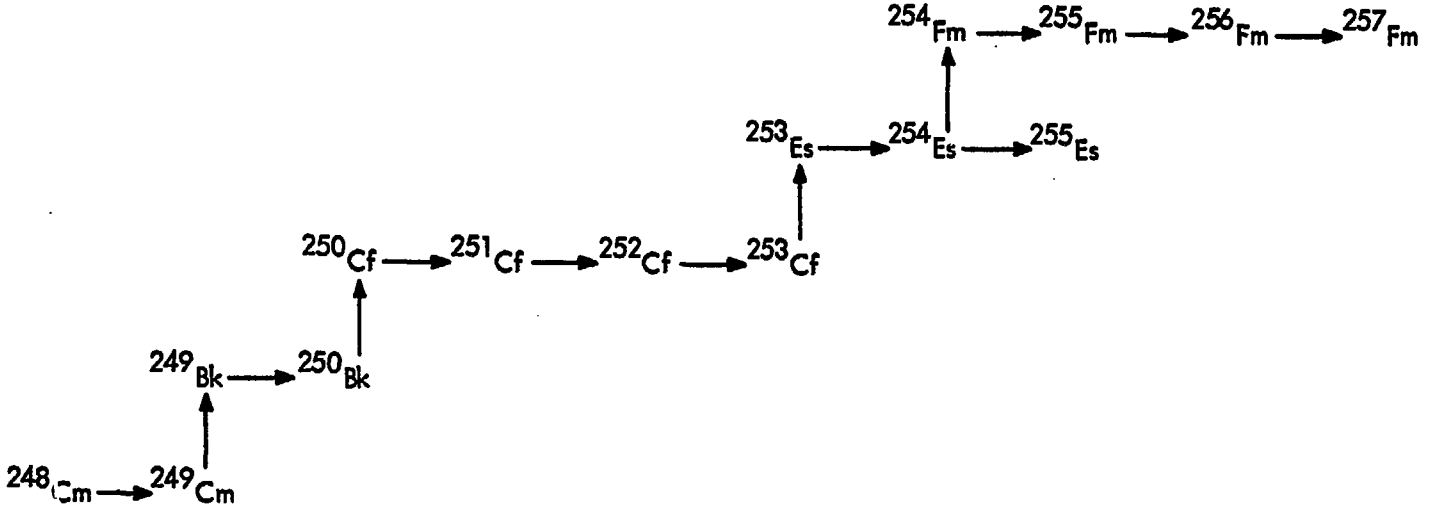
This paper will be presented in two parts. The first part will provide a historical background for the production of elements 99 and 100 in nuclear reactors, while the second will consist of a survey of the current technology used for producing these elements. In 1975, John Crandall[1] reviewed the history of the production of the heavier transuranium elements; therefore, I will only summarize and update his paper.

The production of einsteinium and fermium in reactors is accomplished by the successive capture of neutrons on a source material, as illustrated in Fig. 1. This figure traces only the primary path of the neutron capture sequence. Twenty years ago we would have started this chain at plutonium or americium, the two heaviest elements available in quantity at that time. Now, we begin it with ^{248}Cm to correspond with what currently takes place in the production program, since the feed material to the High Flux Isotope Reactor (HFIR) today is rich in ^{248}Cm.

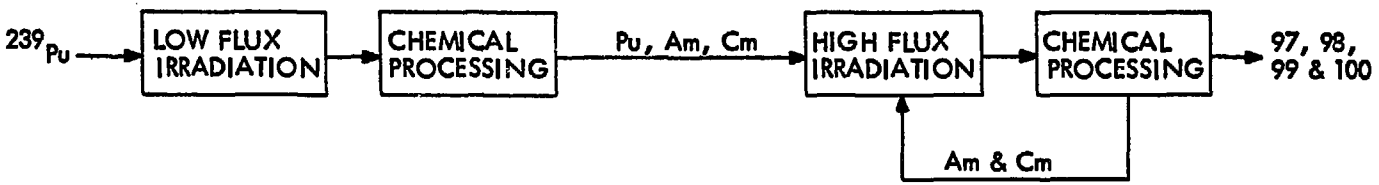
The overall scheme that was adopted in the late 1950s for the production of research quantities of the heavy transplutonium elements is shown in Fig. 2. In this scheme, tens of kilograms of ^{239}Pu would be irradiated in the Savannah River reactors at a comparatively low flux to burn up the fissionable isotopes of plutonium and produce ^{242}Pu, ^{243}Am, and ^{244}Cm. The irradiated material would then be chemically processed to isolate the plutonium and a mixture of americium and curium. These materials would subsequently be fabricated into irradiation targets for a high-flux reactor. After a suitable neutron irradiation, the targets would be processed to recover elements 97, 98, 99, and 100. Finally, the recovered americium and curium would be recycled to the high-flux reactor for further irradiation.

Fig 1

NEUTRON CAPTURE CHAIN



PRODUCTION SCHEME FOR ELEMENTS 99 & 100



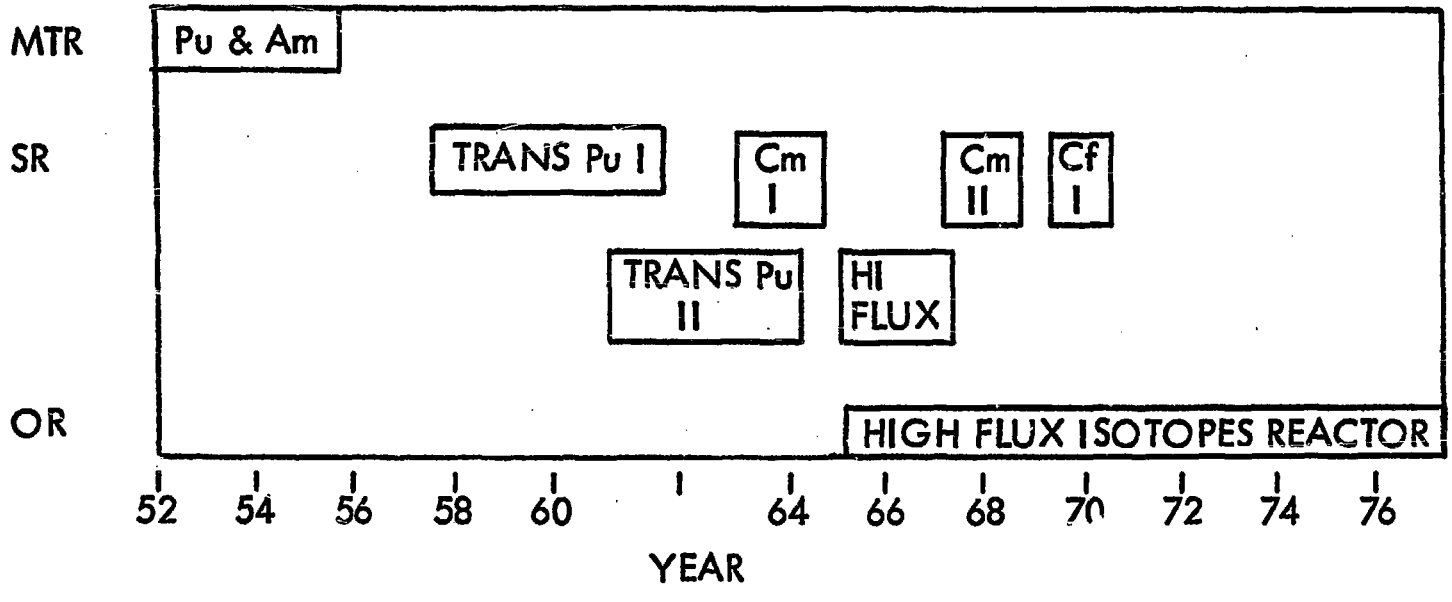
The chronology of the transplutonium processing program is detailed in Fig. 3. Small amounts of plutonium and americium were irradiated in the MTR in the early 1950s to produce transplutonium elements for use in research and chemical process development. The first 10 kg of ^{239}Pu was inserted in a Savannah River reactor in 1957, and this irradiation campaign was labeled TRANS Pu I. A similar irradiation was initiated four years later. While small amounts of elements 99 and 100 were produced in these irradiations, it was not possible to recover them because of the long cooling period required prior to processing at Savannah River.

These irradiations were followed by two ^{239}Pu irradiations to produce curium for isotopic heat sources and a demonstration that a Savannah River reactor could be operated at a very high flux, greater than 5×10^5 neutrons/$cm^2 \cdot sec$. All of these irradiations produced future feed for the HFIR, which was completed in 1965 in Oak Ridge.

When the HFIR and a small transplutonium processing plant (TRU) were started up in the mid-1960s, we were on our way to obtaining significant research quantities of elements 99 and 100. In certain respects, the HFIR has been the most successful research reactor ever built. For example, it has operated at full power 95% of the time for 11 years except for two periods of maintenance.

You will note a fifth irradiation campaign at Savannah River, designated as Cf I. In this campaign, large quantities of plutonium, americium, and curium were irradiated to produce gram quantities of californium. While it was not possible to recover einsteinium and fermium from the irradiated material because of the necessary long cooling period, a supply of curium rich in the 248 isotope was produced. This curium has served as an excellent feed for HFIR.

REACTOR IRRADIATIONS



Typical results of an irradiation cycle in HFIR and a processing campaign in TRU are illustrated in Tables 1 and 2. Table 1 gives the isotopic composition of the americium-curium feed before and after the irradiation. Note that the curium is rich in the 246 and 248 isotopes, which significantly enhances its value for producing the heavier elements. Table 2 lists the products of the campaign, including the isotopic composition of the recovered californium. As an aside, I might mention that tens of milligrams of very pure ^{248}Cm are being produced each year at TRU by the decay of stored californium. The californium solution is "milked" periodically to obtain the curium daughter.

The production of elements 99 and 100 has now reached a steady state. The production totals since 1967 are given in Table 3. You should note that the values listed for 1978 are for only one-half of the year.

Finally, I will show you the effective cross sections for the heavier transplutonium elements in HFIR (Table 4). We call them the Melton Valley, Anderson County, Tennessee cross sections. They differ only slightly from those published in 1965 for the isotopes up through ^{252}Cf; but, of course, we have made significant refinement in the heavier isotope regions.

In the second part of this paper, I will review the technology presently being used in TRU[2] to isolate the heavy elements from irradiated HFIR targets. It is based on someone's realization that, instead of purifying all of the curium each time, a portion of it could be recycled to the next campaign through the use of an ingenious batch solvent extraction contactor.

CAMPAIGN 53

	INPUT (g)	OUTPUT (g)
AMERICIUM-243	4.7	<0.1
CURIUM	113.0	54.8

	%	%
^{244}Cm	57.66	33.91
^{245}Cm	0.777	0.348
^{246}Cm	36.40	55.75
^{247}Cm	1.053	1.550
^{248}Cm	4.11	8.44

Table 2

CAMPAIGN 53 PRODUCTS

INPUT		OUTPUT	
		(mg)	
BERKELIUM-249	-	52.8	
CALIFORNIUM-252	-	361	
			(%)
^{250}Cf			7.58
^{251}Cf			2.10
^{252}Cf			88.73
^{253}Cf			1.53
^{254}Cf			0.06
		(µg)	
EINSTEINIUM-253		2745	
		(pg, est.)	
FERMIUM-257		0.7	

Table 3

PRODUCTION OF ELEMENTS 99-100
IN TRU-HFIR

NUCLIDE	FISCAL YEARS						
	1967-70	1971-73	1974	1975	1976	1977	1978
^{253}Es, μg	481	3210	2170	3750	1700	2620	1960
^{257}Fm, pg	0.26	2.8	1.5	1.6	0.54	1.1	0.7

Table 4

HFIR-EFFECTIVE CROSS SECTIONS, barns

Nuclide	σ_c	σ_f
^{248}Cm	9.26	0
^{249}Bk	1267	0
^{250}Cf	2055	0
^{251}Cf	2454	3361
^{252}Cf	18.4	31.2
^{253}Cf	10.6	1090
^{253}Es	289	0
^{254m}Es	1.1	1540
^{254}Fm	64	0
^{255}Fm	22	840
^{256}Fm	38	0
^{257}Fm	8	4600

When TRU was started up, a continuous solvent extraction process, called Tramex, was used to separate the transplutonium elements from fission products and other impurities. The operational steps in this process recovered all the curium for reuse. We have since replaced the continuous solvent extraction with a batch extraction carried out in the equipment shown schematically in Fig. 4. This consists of a tank equipped to mix the phases and separate them. Thus, we extract the transplutonium elements and strip them from the solvent in a series of batch contacts.

The product is further processed by LiCl anion exchange to remove residual impurities and separate 95% of the curium from the transcurium elements. The californium fraction is then transferred to another facility, and the californium, einsteinium, and fermium are separated by high-pressure ion exchange, as shown in Fig. 5.

I have not described all of the process details, but simply pointed out the major differences from a decade ago. As stated earlier, partially purified curium (about 25%) is stored and recycled to the next campaign. This permits a great simplification of what originally was a very complicated process.

In closing, I would like to pay tribute to two ORNL chemists who were instrumental in developing the technology to routinely produce elements 99 and 100. These chemists, now deceased, are A. ("Chet") Chetham-Strode, Jr., and Russel D. ("Russ") Baybarz. Chet taught us what engineers could and could not do with the transplutonium elements, and Russ was instrumental in developing all the chemical processes we have used to date.

Fig. 4

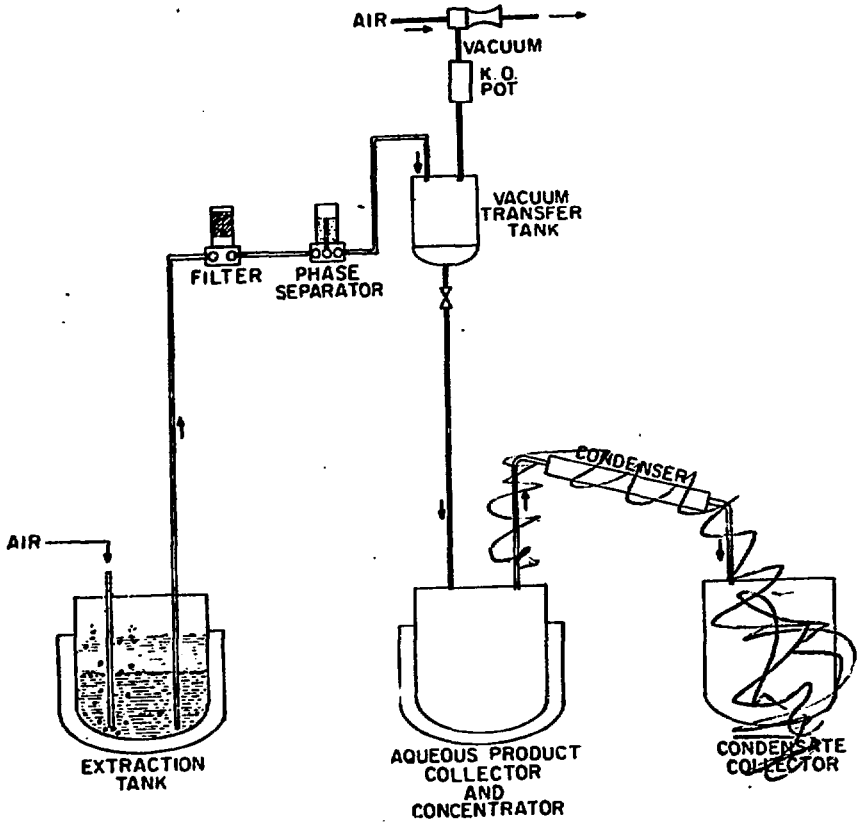
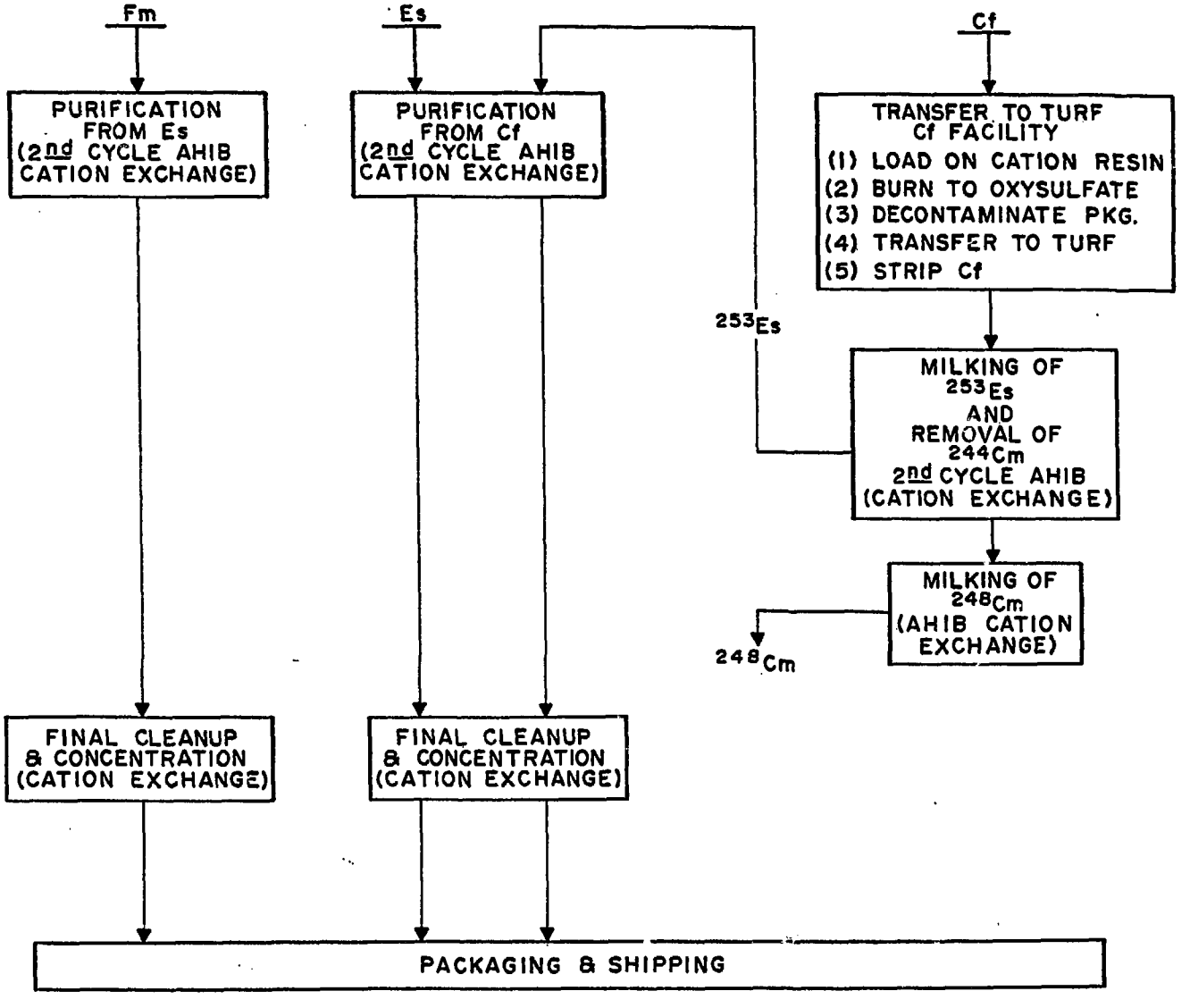


Fig. 2 Schematic arrangement of multipurpose equipment for batch solvent extraction

Fig 5


Fm
Es
Cf
PURIFICATION FROM Es (2nd CYCLE AHIB CATION EXCHANGE)
PURIFICATION FROM Cf (2nd CYCLE AHIB CATION EXCHANGE)
TRANSFER TO TURF Cf FACILITY
(1) LOAD ON CATION RESIN
(2) BURN TO OXYSULFATE
(3) DECONTAMINATE PKG.
(4) TRANSFER TO TURF
(5) STRIP Cf
253Es
MILKING OF 253Es AND REMOVAL OF 244Cm 2nd CYCLE AHIB (CATION EXCHANGE)
MILKING OF 248Cm (AHIB CATION EXCHANGE)
248Cm
FINAL CLEANUP & CONCENTRATION (CATION EXCHANGE)
FINAL CLEANUP & CONCENTRATION (CATION EXCHANGE)
PACKAGING & SHIPPING

References

(1) John L. Crandall, Proceedings of the Symposium Commemorating the 25th Anniversary of Elements 97 and 98 held on January 20, 1975, LBL-4366 (July 1976).

(2) E. D. Collins and J. E. Bigelow, "Chemical Process Engineering in the Transuranium Processing Plant," Proc. 24th Conf. Remote Syst. Technol., 130 (1976).