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CALIFORNIUM PRODUCTION AT THE TRANSURANIUM PROCESSING PLANT*

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ABSTRACT

The Transuranium Processing Plant (TRU) at the Oak Ridge National Laboratory is the production, storage, and distribution center for the ERDA heavy element research program. Since it began operation in 1966, TRU has been the only source in the United States of significant quantities of californium, berkelium, einsteinium, and fermium. A total of 2.6 g of ^{252}Cf has been recovered and purified from plutonium and curium targets that were irradiated in the High Flux Isotope Reactor (HFIR) at ORNL or in the reactors at the Savannah River Plant (SRP). Seventy-five neutron sources have been fabricated at TRU, many of them in special configurations requested by researchers. TRU is currently producing about 0.5 g of ^{252}Cf per year. Plant capacity is estimated to be 1.5-2.0 g per year.

TRU is a hot-cell, chemical processing facility of advanced design. New concepts have been incorporated into the facility for absolute containment, remote operation, remote equipment installation, and remote maintenance. The facilities include a battery of nine heavily shielded process cells served by master-slave manipulators and eight laboratories, four on each of two floors.

Processing includes chemical dissolution of the targets followed by a series of solvent extraction, ion exchange, and precipitation steps to separate and purify the transuranium elements. The transcurium elements berkelium, californium, einsteinium, and fermium are distributed to users. Remote

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techniques are used to fabricate the americium and curium into target rods for reirradiation in the HFIR. Californium-252 that is in excess of the needs of the heavy element research program and the californium sales program is stored at TRU and processed repeatedly to recover the daughter product ^{248}Cm, which is a highly desirable research material.

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The Transuranium Processing Plant (TRU) and the High Flux Isotope Reactor (HFIR) were built at Oak Ridge National Laboratory (ORNL) to provide quantities of the transuranium elements for research.[1] At TRU, target rods irradiated in the HFIR are processed for the separation, recovery, and purification of the heavy actinide elements, which, in turn, are distributed to laboratories throughout the country for research work with these elements. TRU is the production, storage, and distribution center for the heavy-element research program in the United States. Since it began operation in 1966, TRU has been the only source of significant quantities of berkelium, californium, einsteinium, and fermium in the United States. It functions integrally with the researchers and not merely as a supplier. Products are usually highly purified prior to shipment and are frequently provided in special chemical forms and/or in special devices required by the experimenter. In addition to the normal production functions, various programs are pursued in cooperative ventures with other research laboratories to assist them in their work. Some special isotopes are provided upon request, and hot-cell space can be made available for some experimental programs with large quantities of material when such an approach is feasible.

Production History

The complete production history of key isotopes is given in Table I. From 1966 to 1970, processed materials were obtained from irradiations made for the TRU program at the Savannah River Plant (SRP) and at the HFIR,

TABLE I. KEY ISOTOPE PRODUCTION AT TRU

Fiscal Year	Isotopes						
	^{242}Pu (g)	^{243}Am (g)	^{244}Cm (g)	^{249}Bk (mg)	^{252}Cf (mg)	^{253}Es (ug)	^{257}Fm (pg)
67	87	25	134	0.34	5.6	14	0
68	0	188	212	0.05	0.5	0	0
69	15	5	57	2.2	15	98	0.07
70	8	13	72	7.6	52	369	0.19
71	10	3	439	37	284	714	0.72
72	16	3	350	66	513	945	0.85
73	5	4	240	49	428	1551	1.25
74	0	2	87	39	386	2169	1.5
75	0	3	104	75	717	3750	1.9
76	0	2	50	29	277	1700	0.7
Totals	141	248	174.	305	2678	11310	7.2

and all transuranium element products were distributed to researchers. In the period from 1970 through 1973, operations at TRU were expanded to include the processing of larger amounts of materials that had been irradiated at SRP as part of the Californium-I campaign. This was an irradiation and processing campaign designed to obtain ^{252}Cf for use in a program to evaluate the commercial market for ^{252}Cf,[2] and was the base for all of the current ERDA ^{252}Cf utilization effort. Then, in 1974, we went back to processing only HFIR targets. However, production rates have been maintained at a high level because we are now using some of the heavy curium we recovered from the Californium-I program as feed to make HFIR targets.

Curium Composition and Californium Production

The rate at which californium and other transuranium elements is produced during the neutron irradiation of curium, depends upon the isotopic composition of the curium. Figure 1 is a diagram of the primary isotope path that is followed in transmuting curium into berkelium, californium, and einsteinium by neutron irradiation. As neutrons are absorbed, ^{244}Cm is transmuted successively into curium 245, 246, 247, 248, and then to ^{249}Cm which rapidly beta decays to ^{249}Bk. The ^{249}Bk is converted to ^{250}Cf by either of the paths shown which involve both a neutron absorption and a beta decay in one order or the other. Then, more neutron absorption forms ^{253}Cf which beta decays to ^{253}Es.

The arrows pointing down to the right indicate that a significant fraction of the atoms of the isotope fission when they absorb a neutron instead of going on to the heavier isotope. That produces fission products instead of desired products and consumes curium nonproductively.

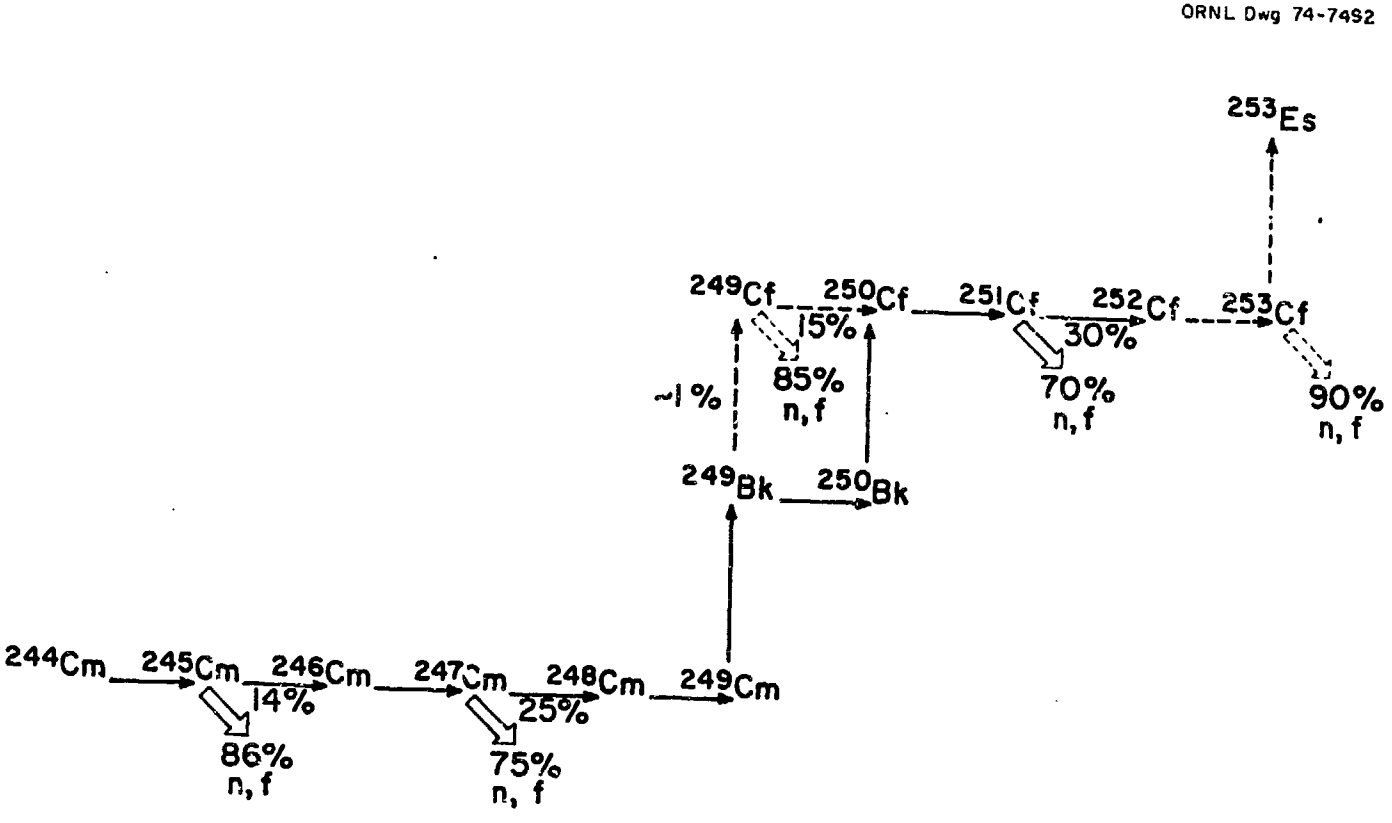

Fig. 1. Transmutation Path from ^{244}Cm to ^{253}Es.

In the curium part of the growth chain only about 14% of the ^{245}Cm atoms go on to ^{246}Cm — the other 86% are destroyed in fission — and only 25% of the ^{247}Cm atoms are converted to ^{248}Cm.

Only about 10 grams of actinides can be put into a HFIR target because of heat removal limitations during irradiation. If the 10 g of curium in a HFIR target contains a larger fraction of ^{246}Cm and ^{248}Cm, transcurium elements will be produced faster than if the curium is predominantly ^{244}Cm. The ^{245}Cm and ^{247}Cm have such high fission cross sections that these isotopes constitute only a small percentage of the curium we handle.

Figure 2 shows the buildup of ^{252}Cf in two targets; one in which the curium contains about 95% ^{244}Cm and one containing heavy curium (about 62% ^{244}Cm, 30% ^{246}Cm, and 3.2% ^{248}Cm). This heavy curium composition is about the average composition of the curium we recovered from the Californium-I campaign. The X's indicate the irradiation periods that have been used prior to removal of the targets from the HFIR for processing. Irradiation of curium containing 30% ^{246}Cm and 3.2% ^{248}Cm for only one-third as long as the same amount of total curium containing 95% ^{244}Cm produces three times as much ^{252}Cf. That is the reason we are producing so much of the transcurium elements today, even though we are actually processing fewer HFIR targets.

There is nothing basically different between the Californium-I curium and the curium produced in the HFIR. Curium gets enriched in the heavier isotopes as it is irradiated. Longer irradiations such as those given the Californium-I curium at Savannah River or cycles of irradiations such as we give curium in HFIR targets result in heavier curium. The important thing about the Californium-I heavy curium is simply that there is so much of it.

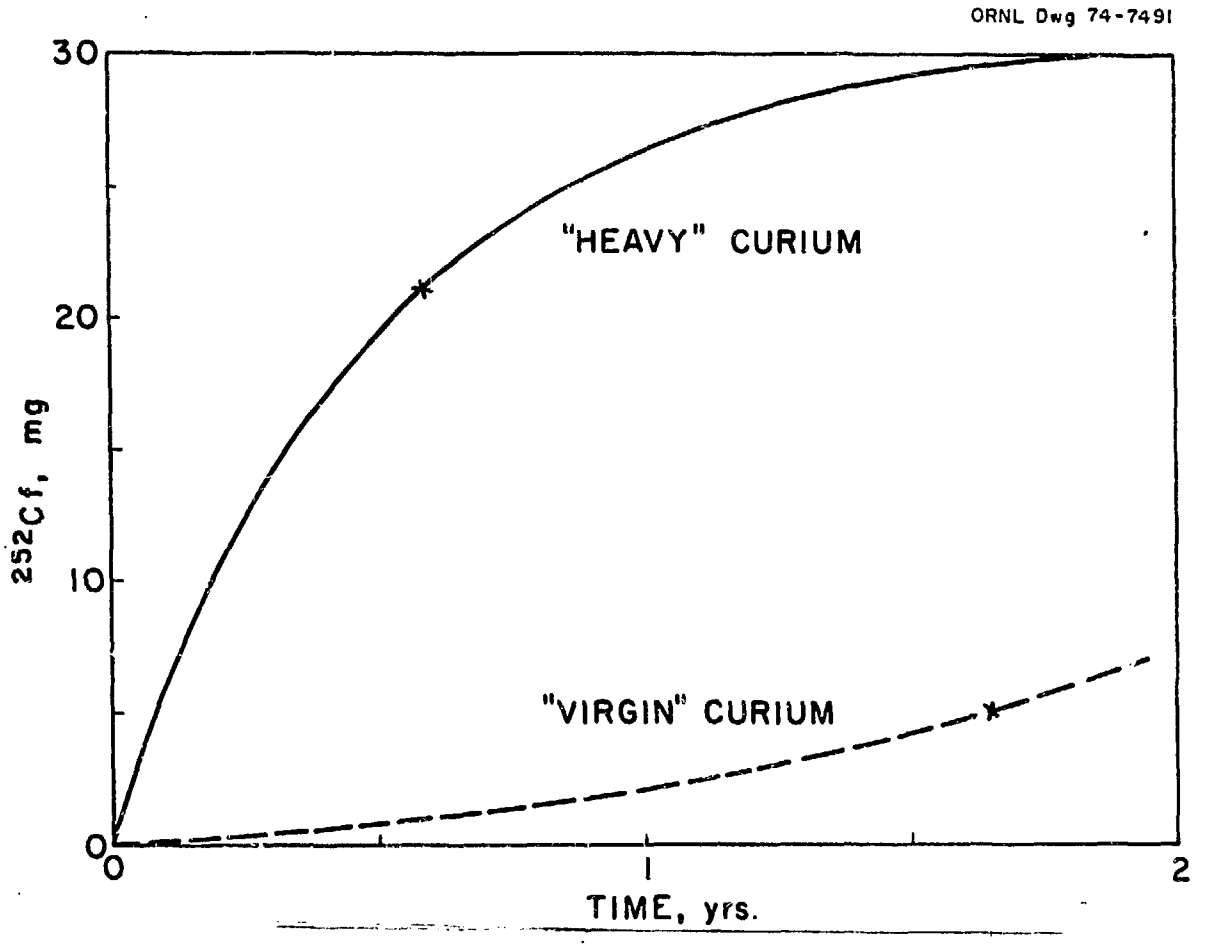

Fig. 2. Heavy Curium Makes ^{252}Cf Faster.

Annual production rates resulting from irradiating and processing about 25 HFIR targets are about 500 mg of ^{252}Cf, 50 mg of ^{249}Bk, 2.5 mg of ^{253}Es, and 1 pg of ^{257}Fm. These amounts fulfill the current needs of researchers for berkelium, einsteinium, and fermium. The ^{252}Cf supplies both the research needs and commercial needs and permits us to maintain enough inventory to recover 100 mg per year of the daughter product, ^{248}Cm.

Facilities and Equipment

The facilities at TRU[3-6] for handling radioactive materials consist of nine heavily shielded cells served by master-slave manipulators and eight laboratories, four on each of two floors. The first floor plan of the two-story building is given in Figure 3. Of the nine cells, which are shielded by 54 in. (1.4 m) of high-density concrete, four contain chemical processing equipment for dissolution, solvent extraction, ion exchange, and precipitation. Three contain equipment for the preparation and inspection of HFIR targets, and two cells are used for analytical chemistry operations. Figure 4 shows a cross section of a typical cell and the surrounding building areas. The top and back of the cell line is enclosed by a high-bay area (the "limited access area") that is equipped with a bridge crane. Removable top plugs provide access to the cells. The front face of the cell line, which is provided with windows and master-slave manipulators, makes up one wall of the operating area. Essentially all process and building service instrumentation is located in the operating area. The second floor immediately over the operating area is a chemical makeup area for process-reagent head tanks, uncontaminated pumps, etc. Transmitters for process and service instrumentation are located there.

Within the shielded cells, process equipment is enclosed in a cell cubicle formed on the front and sides by the cell walls, and of epoxy resin-coated

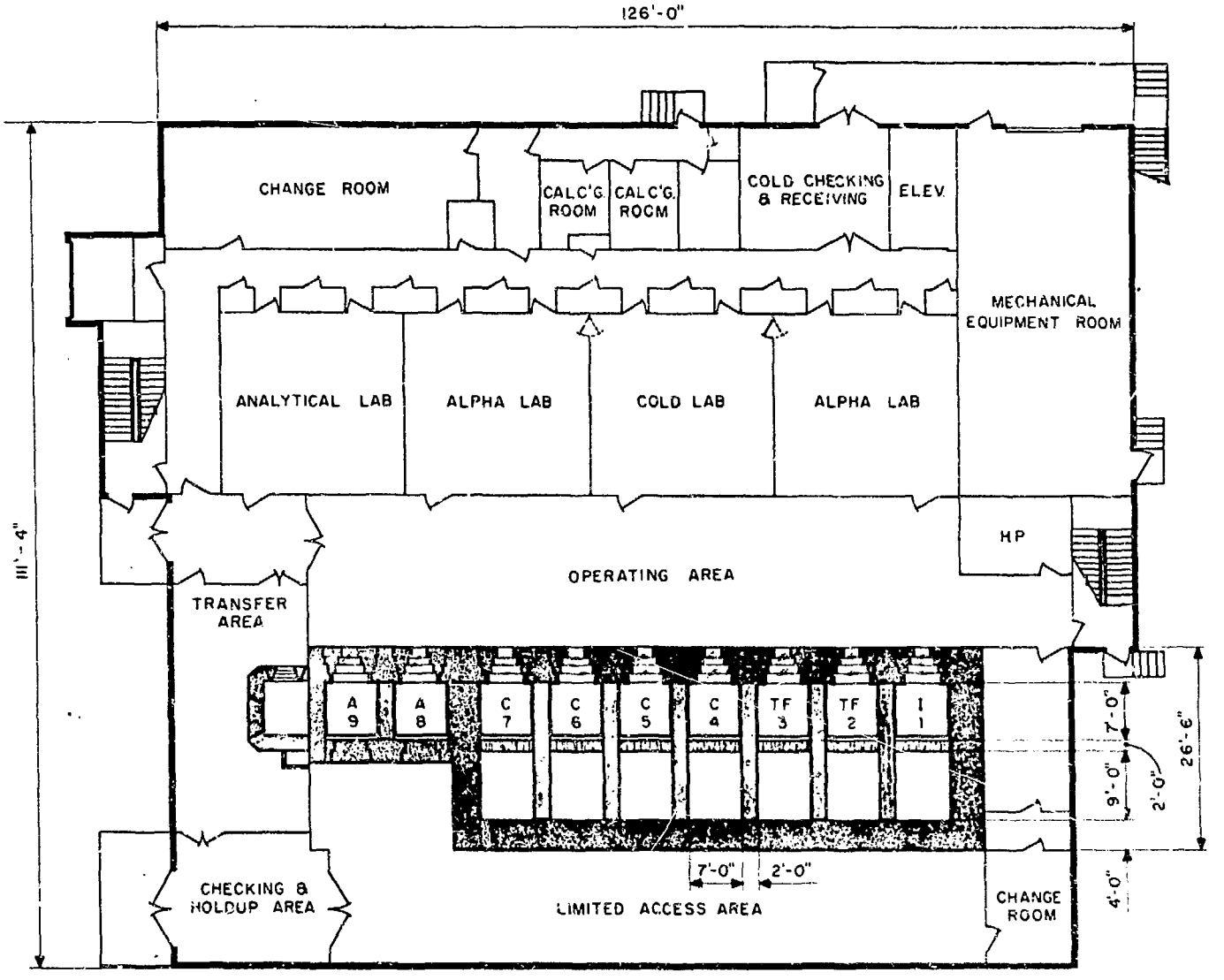

Fig. 3. First-Floor Plan, Transuranium Processing Plant.

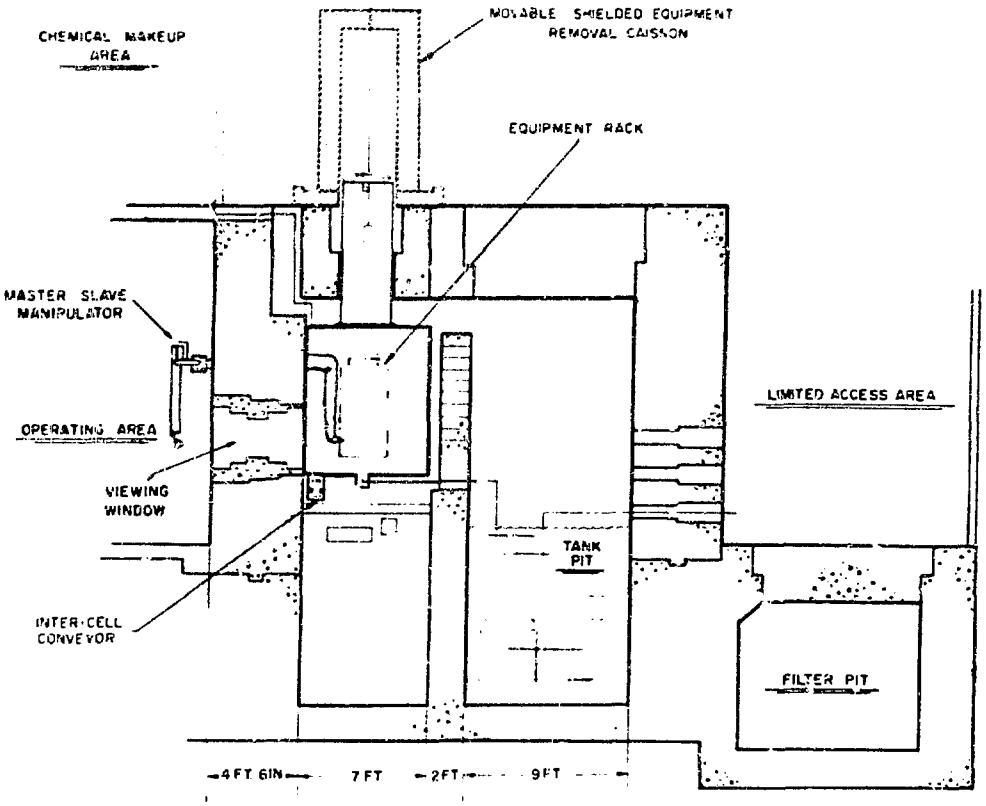

Fig. 4. Cross Section of a Typical Cell at TRU.

stainless-steel plate on the rear wall and top; the bottom is of Hastelloy C to withstand the corrosion of accidentally spilled chloride solutions. A tank pit for housing waste collection equipment and process and storage tanks is behind and below each cell cubicle and shielded from it by a concrete wall.

Service lines enter through removable plugs in the back and top of the cell. Shielded pits in the floor behind the cells house off-gas filters and a pipe tunnel for process lines.

Contaminated equipment can be removed from the cell bank either through an inter-cell conveyor to a shielded carrier at a loading station at one end of the cell bank or through the top of the cell to a shielded caisson (transfer case) designed to maintain shielding and contamination control during the transfer.

Process vessels in the pits behind the cubicles are serviced and maintained with a combination of remote and underwater maintenance techniques.

Figure 5 is a photograph of the main operating control room showing the cubicle viewing windows and the panelboard for containment, safety, and process controls.

Smaller items of chemical processing equipment, such as valves, pumps, ion exchange columns, and equipment for withdrawing solution samples from the tanks in the cell pits, are mounted on racks in the cubicles. This equipment can be installed or removed remotely by using manipulators and air-operated impact wrenches. The equipment rack for the lithium chloride-based anion exchange step is shown in Fig. 6. This rack is about 16 in. (0.4 m) x 36 in. (0.9 m) x 72 in. (1.8 m), which is nearly the maximum size that can be handled in the equipment transfer case.

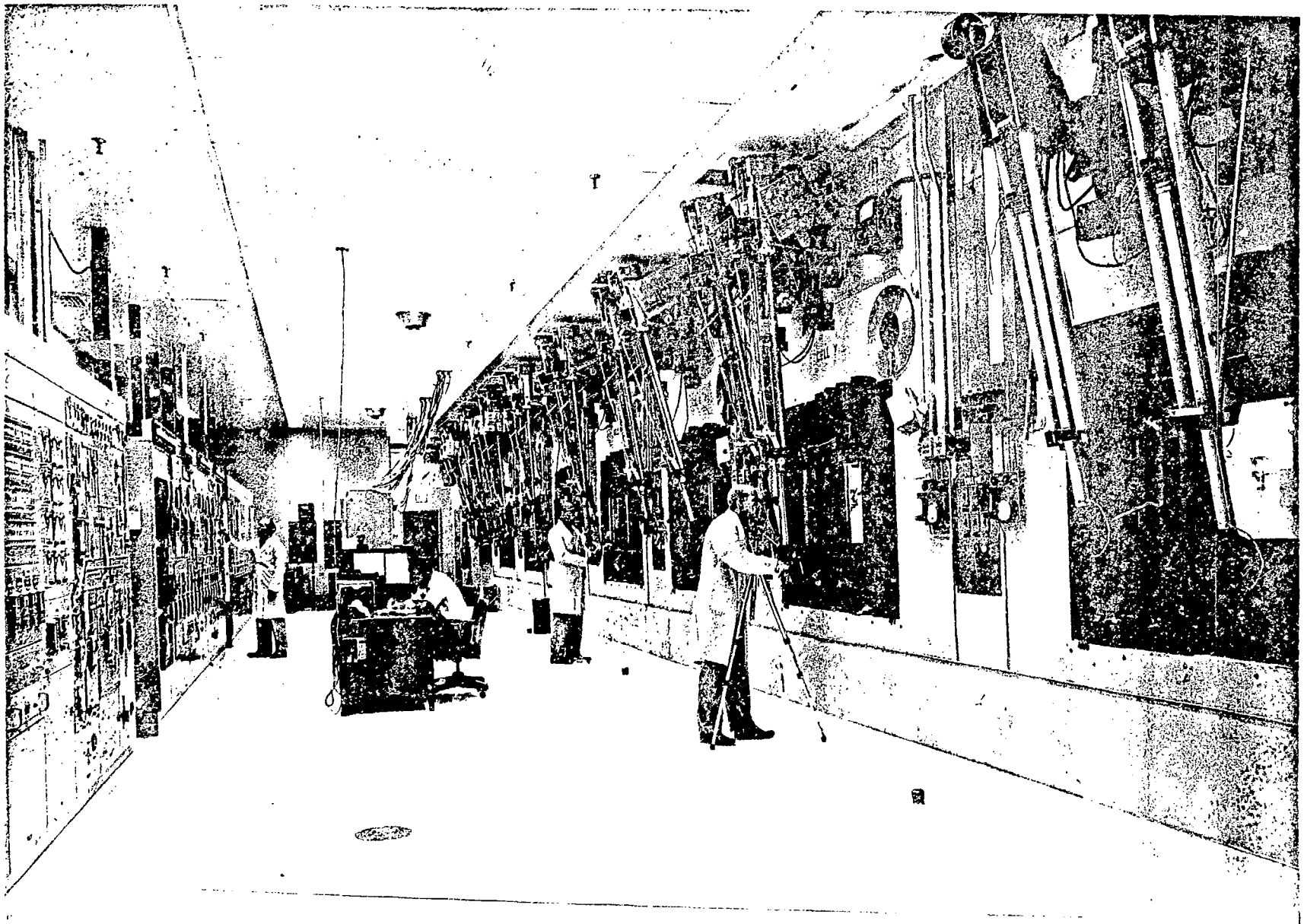

Fig. 5. Operating Control Room, Transuranium Processing Plant.

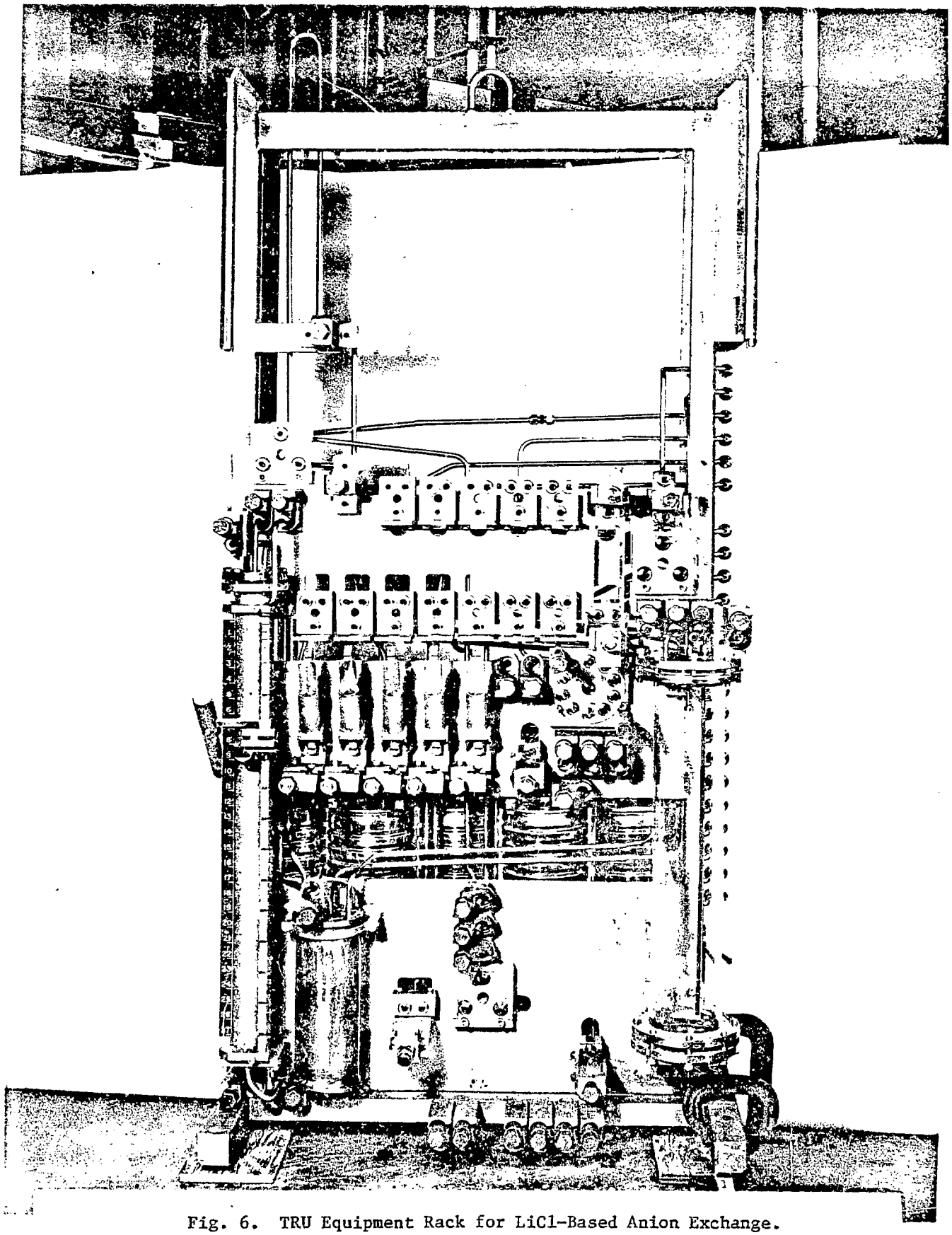

Fig. 6. TRU Equipment Rack for LiCl-Based Anion Exchange.

The ion exchange column mounted at the left side is equipped with a scale and a holder for a neutron probe that is used for scanning the profile of neutron activity on the resin column. Continual monitoring of the rate at which ^{252}Cf is being moved down the column during elution provides a valuable process control. The glass tank at the right side is the feed tank. The ribbed tanks that are partially visible in the back of the rack are tantalum collection tanks for the product fractions. At various places on the rack, groups of pipe ends may be seen which terminate in mounting blocks for various high-maintenance items such as valves. When the rack was installed, it was connected to service lines from the chemical makeup area by a row of TRU disconnect clamps[6] (easy-open mechanical joints developed for TRU) that match the row of disconnect ferrules at the right edge of the rack. Connections to process vessels in the tank pit were made at the TRU disconnects at the bottom of the rack.

The TRU process equipment is assembled using about 3000 TRU disconnect clamps in the cell cubicles and tank pits.

Target Fabrication

Figure 7 is a cutaway view of a target rod. A blend, about 18% of whose volume is actinide oxide powder and the rest aluminum powder, is cold-pressed at 15,000 lbf/in.^{2} (100 MPa) into a thin-walled aluminum sleeve capped on both ends with aluminum powder. Thirty-five of the resulting 1/4-in.-diam, 1/2-in.-long (6.35 mm-diam., 12.7 mm-long) pellets, with a density approximately 80% of theoretical, are then loaded into a finned aluminum tube. This tube is sealed by tungsten–inert-gas welding and is hydrostatically compressed onto the pellets at about 20,000 lbf/in.^{2} (140 Pa).

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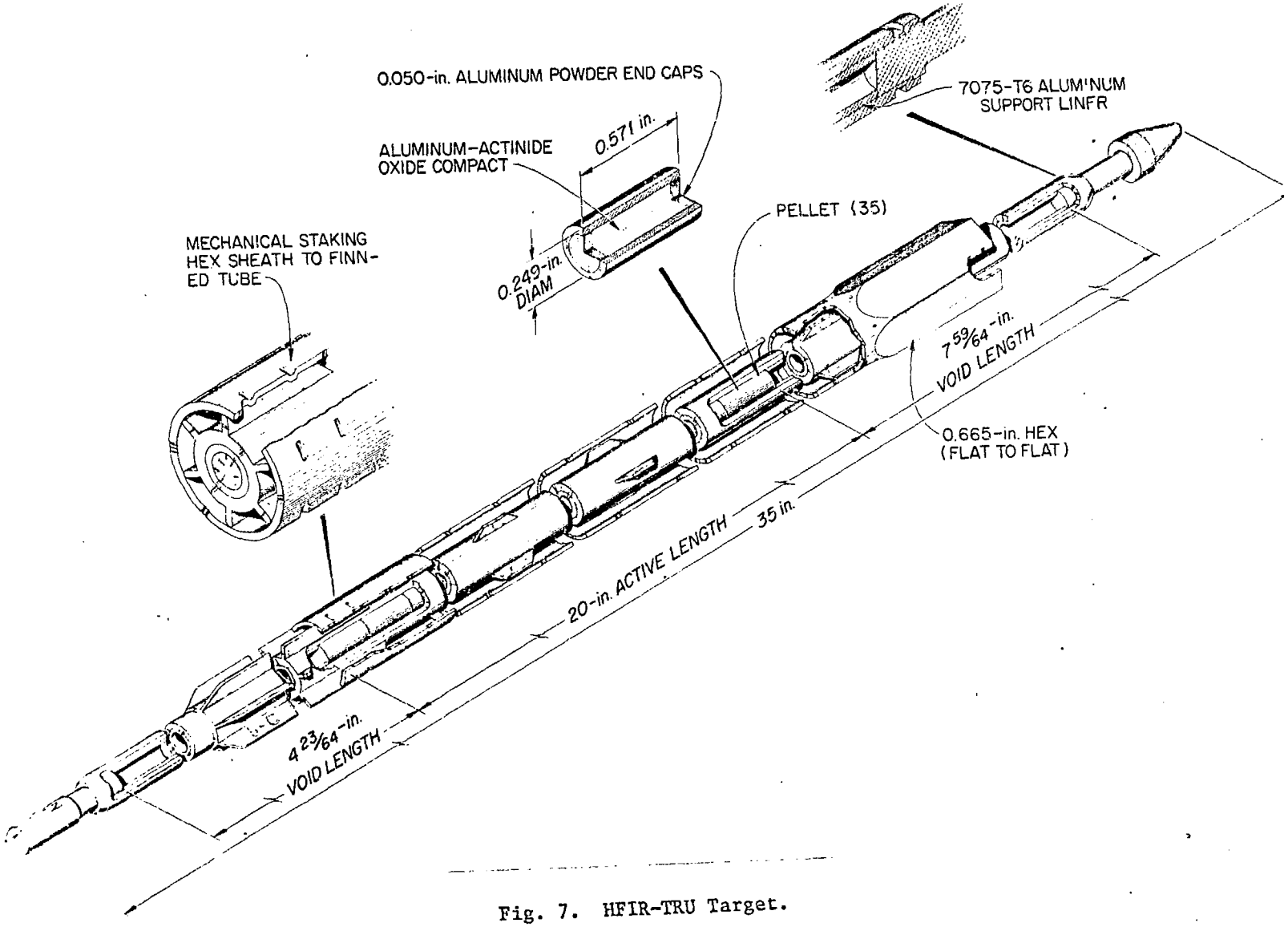

Fig. 7. HFIR-TRU Target.

The target rod is then subjected to a rigorous inspection procedure which includes helium leak check, radiography of the end-closure welds, dimensional inspection, and a contamination check. After the inspection, a coolant-flow shroud is placed over the target rod and mechanically staked at the fins prior to insertion into the HFIR.

Heavy Element Separation

Figure 8 shows the sequence of chemical processing steps now being used in the first stages of processing — those which are performed in the cell bank. When an irradiated target from HFIR arrives at TRU, it enters the main-line process, which consists of:

- dejacketing the target by dissolving the aluminum in caustic-nitrate solution and dissolution of the actinide oxide particles in nitric or hydrochloric acid;
- separating the radioiodine and the residual plutonium by batch solvent extraction into di(2-ethylhexyl)phosphoric acid (HDEHP) in aromatic diluent;
- feed conditioning and general cleanup by batch extraction of the actinides (and lanthanides) from a low-acid salt solution using HDEHP in aliphatic diluent;
- further decontamination of the actinides from fission products in a solvent extraction process using concentrated lithium chloride solution;
- final decontamination from fission products and separation of americium and curium from the transcurium actinides by lithium chloride-based anion exchange;

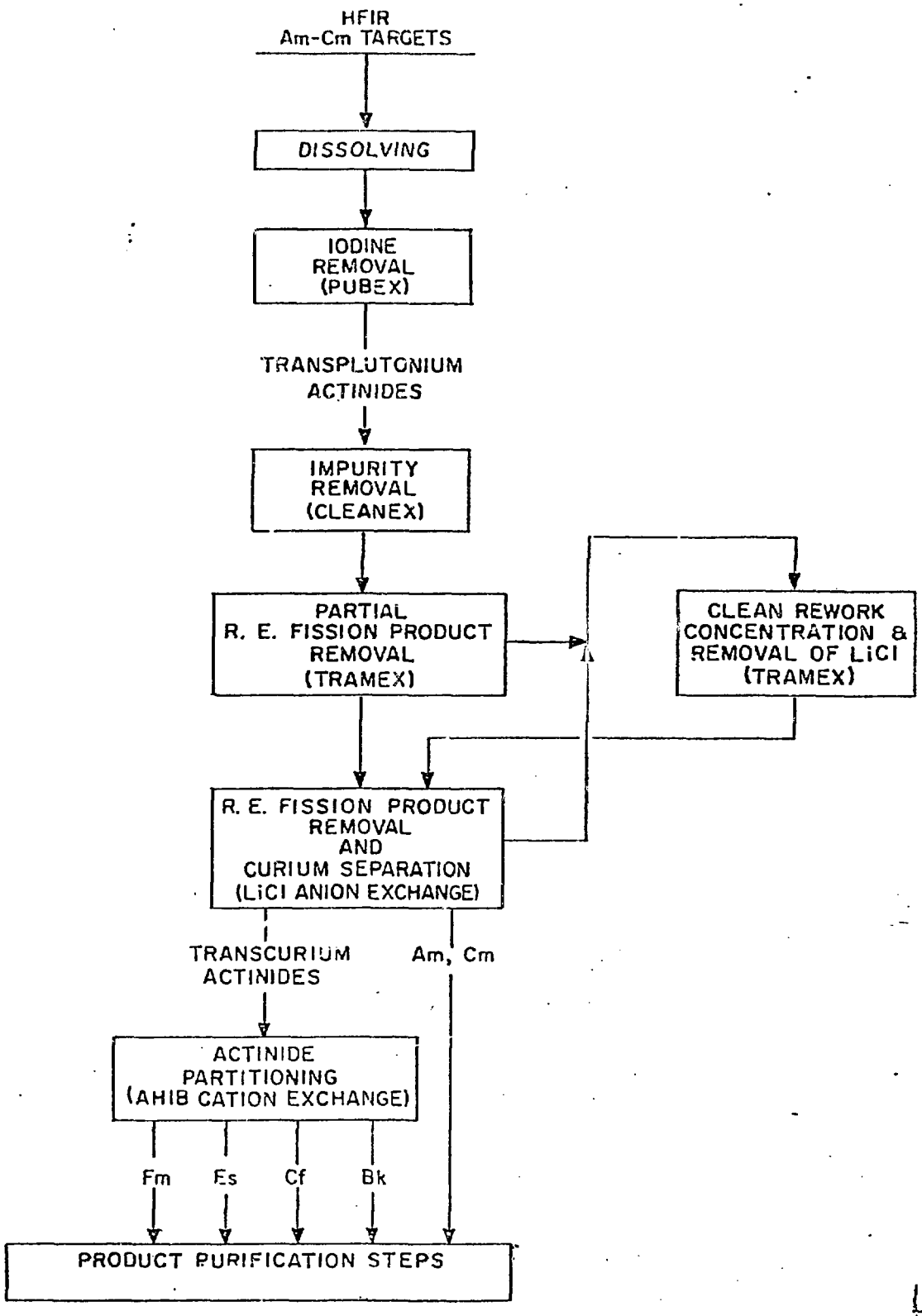

Fig. 8. Chemical Processing Steps in TRU Hot Cells.

- isolation of the berkelium, californium, einsteinium, and fermium by chromatographic elution from a cation exchange resin column using α-hydroxyisobutyrate (AHIB); and
- purification of berkelium by a batch solvent extraction technique using HDEHP in dodecane.

Final purification of the various product fractions is accomplished by the sequences of steps shown in Fig. 9. The fermium and einsteinium fractions are purified from residual californium in a shielded cave using AHIB cation exchange. Once free of californium, the products receive their final purifications in glove boxes.

The americium-curium fraction is recovered from the lithium chloride solution by batch solvent extraction using tertiary amines. Final purification is done by successive oxalate precipitations. Actinide oxide particles are produced for recycle to the target fabrication process by loading the purified americium-curium solution onto cation resin beads and burning the loaded resin to decompose the organic matrix and drive off the volatile constituents. The resulting particles have ideal properties for use in HFIR targets.

The californium in the product fraction from the AHIB ion exchange column is loaded into a shipping package and is transferred by means of a pneumatic transfer system to the TURF Californium Facility in a building next door to TRU. This special facility was built to provide an environment that is free from ^{244}Cm contamination in which extremely pure ^{248}Cm can be prepared. Curium-248, the alpha decay product of ^{252}Cf, is of special interest to researchers because it has a very low specific activity ($T_{1/2}$ = 3.39×10^5 yr) and it can be recovered in relatively pure form.

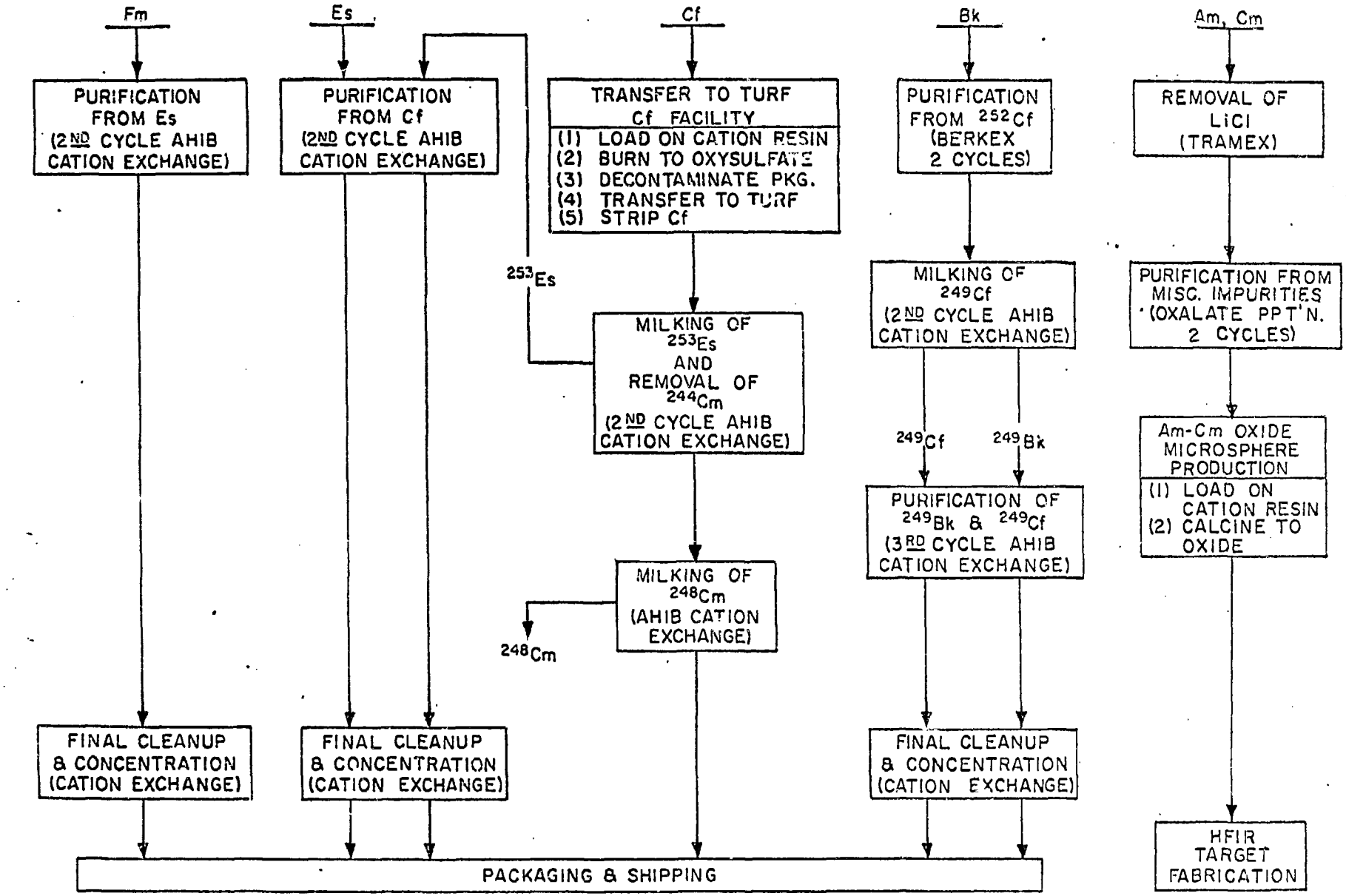

Fig. 9. Product Finishing Steps at TRU.

The shipping package is shown in Fig. 10. About 0.9 ml of cation exchange resin is loaded into the platinum column (shown at the top of the photograph) which has a porous platinum frit on each end. The platinum column is then assembled into a package formed by two metal compression fittings using plastic ferrules (as shown in the exploded display) and the californium is sorbed on the resin by pumping the solution through the small column. After the californium is sorbed on the resin, the resin column is removed from the compression fittings and calcined at 650°C which converts the californium to the oxysulfate. In this form, the californium can be stored for long periods and then be recovered easily in a few ml of HNO_3. Then, the platinum column is put into a package (bottom of photo) made from two of the fitting nuts spot welded together and two fitting plugs. Plastic ferrules may be used for storage but stainless steel ferrules are always used for shipments.

The normal processing sequence is to store the californium product about one month and then process it by AHIB cation exchange to recover isotopically pure ^{253}Es that has formed by decay of ^{253}Cf. Traces of ^{244}Cm are also removed in this step. The californium may then be loaded into a small ion exchange column and calcined either for shipment to a researcher or to the Savannah River Plant for distribution through the demonstration or sales program. Some of the californium may be fabricated into neutron sources for use in programs sponsored by ERDA or other U.S. government agencies. Californium not needed immediately is stored for the production of ^{248}Cm.

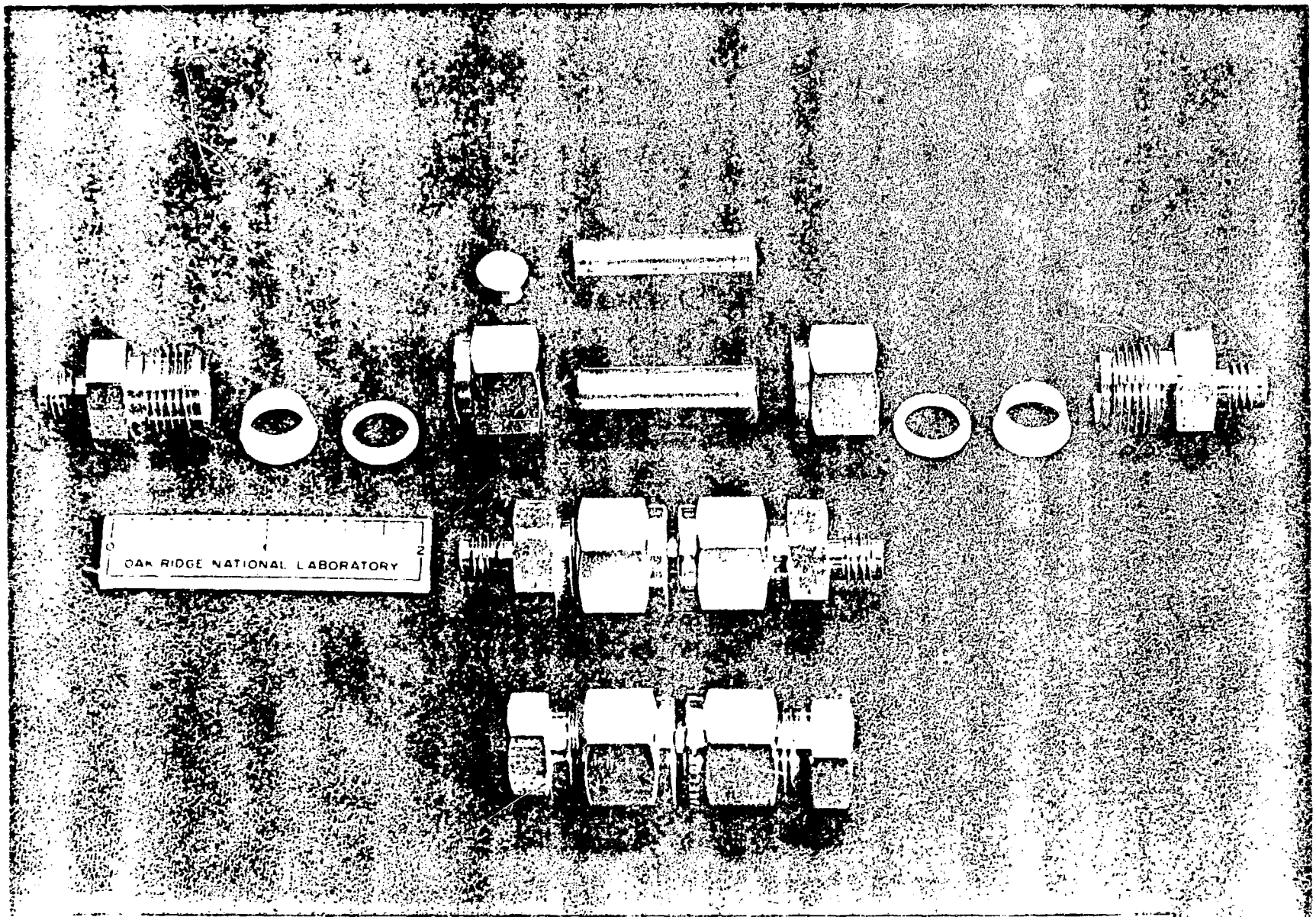

Fig. 10. Components of TRU Shipping Package for ^{252}Cf.

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