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Cf-252: PROPERTIES, PRODUCTION, SOURCE FABRICATION, AND PROCUREMENT

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ABSTRACT

Californium-252 is currently produced in the High Flux Isotope Reactor (HFIR) and processed in the Radiochemical Engineering Development Center (REDC) at the Oak Ridge National Laboratory. Medical sources are fabricated at the Savannah River Site and all other sources are fabricated at the REDC. The production of ^{252}Cf, properties, neutron source fabrication, and source procurement are described.

INTRODUCTION

The element californium was first discovered in 1950 from the bombardment of helium ions on a ^{242}Cm target at the Berkeley Crocker Laboratory 60-in. cyclotron. The reaction product was first thought to be ^{244}Cf but was later identified as ^{245}Cf.[1] However, it was not until 1952 that the isotope ^{252}Cf was discovered in the debris from uranium that had been subjected to an instantaneous and very intense neutron irradiation.[2] Early investigation of its properties revealed a half-life between two and three years and a significant branching fraction for decay by spontaneous fission, making ^{252}Cf an especially compact source of neutrons.

These desirable properties led to a sustained (and still continuing) national effort to produce and recover macroscopic quantities of ^{252}Cf. It started in late 1952 with the irradiation of multigram quantities of ^{239}Pu in the Materials Test Reactor (MTR) at the Idaho National Engineering Laboratory and eventual recovery of purified microgram quantities of ^{252}Cf in 1958 at Lawrence Berkeley Laboratory.

Scientific interest in ^{252}Cf increased to such a level that demand for the isotope quickly exceeded the supply. As a result, in late 1958, a National Transplutonium Element Production Program was undertaken to produce large quantities of ^{252}Cf and other transplutonium isotopes for the research community.

This new program led to a large-scale market evaluation program at the Savannah River Plant (SRP) and a smaller research effort at Oak Ridge National Laboratory (ORNL) beginning in the late 1960s. Since 1973, though, the entire supply of ^{252}Cf in the western world has been produced in the High Flux Isotope Reactor (HFIR) and recovered at the Radiochemical Engineering Development Center (REDC), formerly known as the TRU Facility, at ORNL.

PRODUCTION OF ^{252}Cf AT ORNL

Initially, ^{252}Cf was produced at HFIR/REDC from the neutron irradiation of ^{242}Pu. Yields of ^{252}Cf were enhanced relative to earlier ^{252}Cf production at the MTR because of the very high, steady-state thermal neutron flux (2-3 $\times$ 10^{15} n/cm^2-sec) available at the HFIR. In addition, the use of ^{242}Pu as target or feed material, rather than ^{239}Pu, enhanced the yield because it required the absorption of fewer neutrons to produce ^{252}Cf and losses due to ^{239}Pu and ^{241}Pu fission were eliminated. By 1968, most of the ^{242}Pu had been irradiated with a substantial fraction being transmuted to ^{244}Cm. This ^{244}Cm, along with ^{244}Cm produced at SRP, became the target material for subsequent ^{252}Cf production.

Continual irradiation and recovery of the ^{244}Cm target material has resulted in a significant and advantageous increase in the heavier curium isotopes. The percentage of ^{244}Cm in the target material has decreased from ~90% to 30-40%. The decrease in ^{244}Cm and subsequent buildup of, primarily, ^{246}Cm and ^{248}Cm has greatly enhanced the yield of ^{252}Cf. This is due, primarily, to reducing the loss of target material to ^{245}Cm fission and the requirement for fewer neutrons to produce ^{252}Cf. Figure 1 shows the path of neutron capture and beta decay for producing ^{252}Cf and losses incurred due to fission. A current, typical charge composition and discharge californium composition for a single transplutonium target are given in Tables I and II.

A typical transplutonium campaign involves the irradiation of 11-13 targets for 12-15 months in HFIR followed by processing and recovery in remotely-operated and remotely-maintained hot cells at the REDC. Processing of the targets and recovery of the transplutonium isotopes takes approximately four months to complete. Typical annual yields of ^{252}Cf are 400 to 500 mg from 110 to 120 g of curium feed material. Production and recovery of ^{252}Cf could probably be increased to ~1.5 g/year if demand and mission merited such a program.

SOURCE FABRICATION

The californium recovered from irradiated HFIR targets is purified as much as possible from fission products, cationic and anionic impurities, and other actinides in the hot cell facilities of REDC Building 7920. Final purification from residual curium isotopes, especially ^{244}Cm, is necessarily performed in the hot cells of the ORNL Californium Facility, REDC Building 7930, where cross contamination by ^{244}Cm can be very closely controlled. All subsequent californium handling and purification operations, including all neutron source fabrications, are done in the Californium Facility.

The original Californium Facility was located entirely within Cell G of Building 7930 and since 1970 was operated through the DOE Research Program to supply various users with bulk californium, ORNL-designed neutron sources, and high-isotopic-purity ^{248}Cm, the alpha-decay daughter of ^{252}Cf. In 1983, the Office of Nuclear Materials Production decided to move the Californium Industrial Sales/University Loan Program to the ORNL Californium Facility from the Savannah River Laboratory (SRL) where program activities were being reduced and the temporary facilities needed replacement. This program, originated in the late 1960s, supplied ^{252}Cf in the form of intense neutron sources to users in the commercial, university, and medical sectors. In order to accommodate the activities of the Sales/Loan Program, including fabrication of Savannah River-designed neutron sources, while retaining full production capabilities for the Research Program, the Californium Facility was expanded both within the existing Cell G area and into the Cell C and Cell B areas and the old fuel storage pool as shown in Figure 2. A pneumatic transfer system was also installed to permit rapid source transfers between the various hot cell workstations and the converted californium storage pool.

The processing equipment used to purify californium, recover high-isotopic-purity ^{248}Cm, and fabricate the various neutron source designs is located at the four workstations of Cell G. The equipment used for pressurized ion exchange purifications of ^{252}Cf and ^{248}Cm and for preparation of the chemical and physical forms of californium required for neutron source fabrication are shown in Figure 3, the interior of the workstation 2 cubicle. All purification and fabrication operations that involve high levels of contamination are carried out in the Cell G workstation cubicles. This includes assembly of all source configurations through primary encapsulation, helium-leak testing and decontamination of the primary capsules, and preliminary assay of the ^{252}Cf content. Secondary source capsule fabrication, helium-leak testing, decontamination, final ^{252}Cf assay, and packaging for shipment are all completed within the clean areas of Cell C and Cell B.

The general processing flowsheet shown in Figure 4 is initiated as soon as the californium is transferred into the Californium Facility. First, residual ^{244}Cm is removed by ion exchange. Most of the purified californium is loaded into platinum storage/shipping packages which are then held for $\sim$2 years while the ^{252}Cf decays to ^{248}Cm. The californium is then reprocessed to recover the ^{248}Cm. At this point, the californium can be reloaded into platinum storage/shipping packages and held for another decay period or can be set aside for preparation of "bulk" shipments or for the fabrication of neutron sources.

Separate processing flows have been shown for the Industrial Sales/Loan and Research Programs because the chemical form and fabrication techniques are very different even though the resulting neutron source capsules appear similar. Also, prior to 1986 the californium for the Sales/Loan Program was transferred from the ORNL Californium Facility to SRS for source fabrication. Now all source forms and capsule

designs are fabricated in the Californium Facility with the exception of the medical source designs.

The differences between the Research Program source designs, ORNL-type, and those of the Sales/Loan Program, SR-Cf-Series Sources, are reflected in the chemical procedures and fabrication techniques that are required to prepare them. The basic solid form for the ORNL-type sources is californium oxysulfate, $Cf_2O_2SO_4$, which is prepared by a resin loading/calcination technique. The oxysulfate is dispersed in aluminum powder to form a pellet, which is then singly- or doubly-encapsulated in 304L stainless steel or Zircaloy-2 to complete the source fabrication.

For the SR-Cf-Series Sources, californium oxide, Cf_2O_3, is the basic solid form. These sources are fabricated by first precipitating the californium from dilute nitric acid solution using oxalic acid. The californium oxalate slurry is filtered into a Pt-10% rhodium primary capsule, dried, and thermally decomposed to the oxide form. Following closure by seal welding, the primary capsule is assembled and seal-welded in secondary capsule components of either 304L stainless steel or Zircaloy-2.

The standard source capsules for both programs are shown in Figure 5. Even though the fabrication methods are quite different, it can be seen that the completed neutron sources are quite similar. The three capsules from the left in Figure 5 are the SR-Cf-100, SR-Cf-2000, and SR-Cf-3000 series source designs. All three are doubly-encapsulated and have the same diameter, 0.371 in. The SR-Cf-100, which is 1.48 in. in length, is the most often fabricated capsule configuration and is designed to contain up to 10 mg of ^{252}Cf. The SR-Cf-2000 and the SR-Cf-3000 are identical to the SR-Cf-100 with the exception of length. Lengthening the source capsule permits containment of larger quantities of ^{252}Cf, up to 35 mg for the SR-Cf-2000 and up to 50 mg for the SR-Cf-3000.

The two capsules to the right are the standard singly- and doubly-encapsulated ORNL-type sources designs, which have been fabricated with up to approximately 30 mg of ^{252}Cf. A larger version of the doubly-encapsulated ORNL-type source was fabricated with 57 mg of ^{252}Cf, the largest neutron source fabricated with current designs. All these source designs contain relatively large amounts of ^{252}Cf in a minimum volume and thus provide very compact, very intense neutron sources with neutron emission rates on the order of 10^{11} neutrons per second.

Another basic form of californium used in commercial source fabrication, also developed at SRS, consists of a palladium-californium oxide composite material, which is shaped into either wires or pellets. To prepare a Pd-Cf_2O_3 composite source, the californium oxalate is precipitated as usual. Tetrammine palladous nitrate is added to the oxalate slurry. The ionic palladium (Pd^{+2}) is then reduced to palladium metal by the addition of hydrazine hydrate. During the reduction process, the californium becomes dispersed uniformly in the mixture as the palladium metal deposits on the surface of the finely divided oxalate precipitate. Following decantation of the supernate, the precipitated mixture is washed, dried, and calcined at 450° C to convert the californium oxalate in the palladium matrix to the oxide form. The calcined mixture is then transferred to a die and green-pressed to form a pellet.

The green-pressed pellet is transferred to a carbon crucible and either sintered at 1300° C to form Pd-Cf_2O_3 composite cermet or melted at 1600° C to form Pd-Cf_2O_3 alloy. A sintered pellet may be coined and then encapsulated as an SR-Cf-100 Series Source. Sintered or alloy pellets may also be processed through the successive grooves of a jewelry rolling mill to form wires ~6 in. long with a nearly square cross-section of ~0.045 in. on a side.

Pd-Cf_2O_3 composite wires, as shown in Figure 6, are normally used for sources that are small in terms of ^{252}Cf content. The wires are fabricated with nominal ^{252}Cf concentrations in three ranges: 500 μg/in., 50 μg/in., and 5 μg/in. Because the concentration is very uniform over the entire wire length, required source strengths may be obtained extremely accurately by cutting a wire of the specified concentration to the appropriate length. Wire segments can then be encapsulated into standard source configurations or packaged and shipped to commercial encapsulators.

Palladium-californium oxide composite contained within a core wire is the basic component common to all ^{252}Cf medical sources. Fabrication and encapsulation of the core wires, however, require very specialized equipment and techniques. The capabilities required to fabricate the various medical source configurations have been retained at SRL. However, the ^{252}Cf medical source billet necessary to fabricate the core wires will be prepared in the Californium Facility at ORNL and then packaged and shipped to SRL.

Fabrication of a medical source billet, as shown in Figure 7, requires the preparation of a sintered pellet of Pd-Cf_2O_3 composite. This is accomplished by the procedures described earlier. The sintered pellet, containing the required amount of ^{252}Cf, is inserted into a Pt-10% iridium billet. To encapsulate the pellet, the billet is either closed by a seal-welded end plug or by gold brazing. The sealed billet will next be transferred to SRS where it will undergo multiple swaging and drawing operations to form a core wire having a diameter of 0.0125 in. The swaging/drawing operation uniformly distributes and effectively immobilizes the Pd-Cf_2O_3 composite containing ^{252}Cf within the clad core wire. The wire is then cropped into segments of the proper lengths for each of the medical source capsule designs, which are illustrated in Figures 8, 9, and 10. The ^{252}Cf is in essentially unleachable form in the core wires except for a few particles of the ^{252}Cf

composite that are possibly exposed at the cropped ends. In each design, the core wires are doubly-encapsulated in Pt-10% iridium primary (inner) and secondary (outer) capsules, which are individually sealed by fusion welds. The applicator tube (AT), the afterloading cell (ALC), and the short afterloading cell (SALC) are capable of containing 20, 5, and 2.5 μg of ^{252}Cf, respectively.

PROPERTIES

Californium-252, due to its spontaneous fission decay mode and its availability in macroscopic quantities, has been one of the most extensively studied transplutonium isotopes. Most of the effort has been directed at understanding the spontaneous fission properties. Some of these fission properties are summarized in Tables III through VI. These properties should be taken as "typical" or "representative" of ^{252}Cf spontaneous fission and not necessarily the "best" or "refereed" values.

Table III shows the decay properties of californium isotopes that are routinely produced during the production of ^{252}Cf. Californium-253 has a short half-life, 17.81 days, and will not be present in any of the fabricated sources. Californium-254 decays almost entirely by spontaneous fission. However, it is only produced in minute amounts (due to the short half-life of its precursor ^{253}Cf), and its contribution to the overall neutron source strength is generally negligible. All the other listed californium isotopes will be present. The exact isotopic mixture is dependent on the conditions of irradiation and subsequent period of decay.

Table IV gives some of the gross spontaneous fission properties of ^{252}Cf. The neutron emission rate of 3.768 n/s/fission ($\sim 2.31 \times 10^{12}$ n/s/g of ^{252}Cf) does include the fission product delayed neutrons.

The ^{252}Cf neutron energy spectrum is given in Table V. The NBS evaluated spectrum in Table V compensates for the deviations from an ideal Maxwellian spectrum with the use of energy-dependent adjustment functions. The relative uncertainties in the NBS neutron energy spectrum are small with the exceptions of the relative uncertainties in the 0-0.25 MeV and 8.0-12.0 MeV energy groups. However, the number of neutrons emitted in these two energy groups is small compared to the total neutron emission rate.

An excellent treatment on prompt fission neutron spectra is given in Ref. 12 and, in particular, for ^{252}Cf in Ref. 13. A comparison of four representations of the prompt neutron spectrum for ^{252}Cf is given in Ref. 14.

The prompt gamma spectrum and equilibrium fission product gamma spectrum are given in Table VI. X-rays from decay, which are relatively abundant, are not included in either spectrum.

Table I

Typical Transplutonium
Charge Composition

Isotope	Quantity (g/target)
^{238}Pu	8.10-4
^{239}Pu	1.00-3
^{240}Pu	2.30-1
^{241}Pu	1.40-4
^{242}Pu	8.70-4
^{241}Am	1.46-1
^{243}Am	3.24-1
^{242}Cm	7.00-5
^{243}Cm	1.60-3
^{244}Cm	3.83+0
^{245}Cm	5.36-2
^{246}Cm	3.60+0
^{247}Cm	1.02-1
^{248}Cm	6.30-1

Table II

Typical Discharge
Cf Composition

Cf Isotope	Quantity (g/target)
^{249}Cf	~2.27-4[a]
^{250}Cf	3.19-3
^{251}Cf	8.82-4
^{252}Cf	4.06-2
^{253}Cf	5.55-4

[a]Quantity is dependent on period between discharge and Bk-Cf separations.

Table III

Decay Properties of Selected Cf Isotopes[6]

Isotope	Half-life	Specific Activity (Ci/g)	Decay Mode	Branching Fraction (%)
^{249}Cf	350.6 y	4.095	α SF	~100 5.2×10^{-7}
^{250}Cf	13.08 y	109.3	α SF	99.923 0.077
^{251}Cf	898 y	1.59	α	100
^{252}Cf	2.645 y	536.3	α SF	96.908 3.092
^{253}Cf	17.81 d	2.898×10^{4}	β^- α	99.69 0.31
^{254}Cf	60.5 d	8497	SF α	99.690 0.310

Table IV

^{252}Cf Spontaneous Fission Properties

SF branching fraction	3.092%[6]
Neutron emission rate	3.768 n/sec/fission[7]
Mean fission neutron spectrum energy	2.13-2.15 MeV[8-10]
Prompt γ-ray multiplicity (mean)	~10/fission[11]
Average prompt γ-ray energy	0.7-0.9 MeV[11]
Total prompt γ-ray energy	6.7-9.0 MeV[11]

Table V

NBS Evaluation of the ^{252}Cf Neutron Spectrum[8]

$$X_{Cf}(E) = [0.6672 \sqrt{E} \exp(-E/1.42)] \cdot \mu(E)$$
(E is in MeV)

Energy Interval (MeV)	$\mu(E)$
0 - 0.25	1 + 1.20E - 0.237
0.25 - 0.8	1 - 0.14E + 0.098
0.8 - 1.5	1 + 0.024E - 0.0332
1.5 - 6.0	1 - 0.00062E + 0.0037
6.0 - 20	1.0 exp [-0.03 (E - 6.0)]

Energy Interval (MeV)	Relative Uncertainty (1σ) (%)
0 - 0.25	±13
0.25 - 0.8	±1.1
0.8 - 1.5	±1.8
1.5 - 2.3	±1.0
2.3 - 3.7	±2.0
3.7 - 8.0	±2.1
8.0 - 12.0	±8.5

Table VI

Gamma Rays from Spontaneous Fission of ^{252}Cf[15]

Energy (MeV)	Photons/(sec) (g of ^{252}Cf) Prompt Gammas	Equilibrium Fission Product Gammas	Total
0 - 0.5	3.3×10^{12}	1.3×10^{12}	4.6×10^{12}
0.5 - 1.0	1.7×10^{12}	4.0×10^{12}	5.7×10^{12}
1.0 - 1.5	7.7×10^{11}	9.1×10^{11}	1.7×10^{12}
1.5 - 2.0	4.2×10^{11}	3.5×10^{11}	7.7×10^{11}
2.0 - 2.5	2.2×10^{11}		2.2×10^{11}
2.5 - 3.0	1.1×10^{11}		1.1×10^{11}
3.0 - 3.5	5.6×10^{10}		5.6×10^{10}
3.5 - 4.0	3.0×10^{10}		3.0×10^{10}
4.0 - 4.5	1.7×10^{10}		1.7×10^{10}
4.5 - 5.0	8.2×10^{9}		8.2×10^{9}
5.0 - 5.5	4.9×10^{9}		4.9×10^{9}
5.5 - 6.0	1.8×10^{9}		1.8×10^{9}
6.0 - 6.5	1.0×10^{9}		1.0×10^{9}

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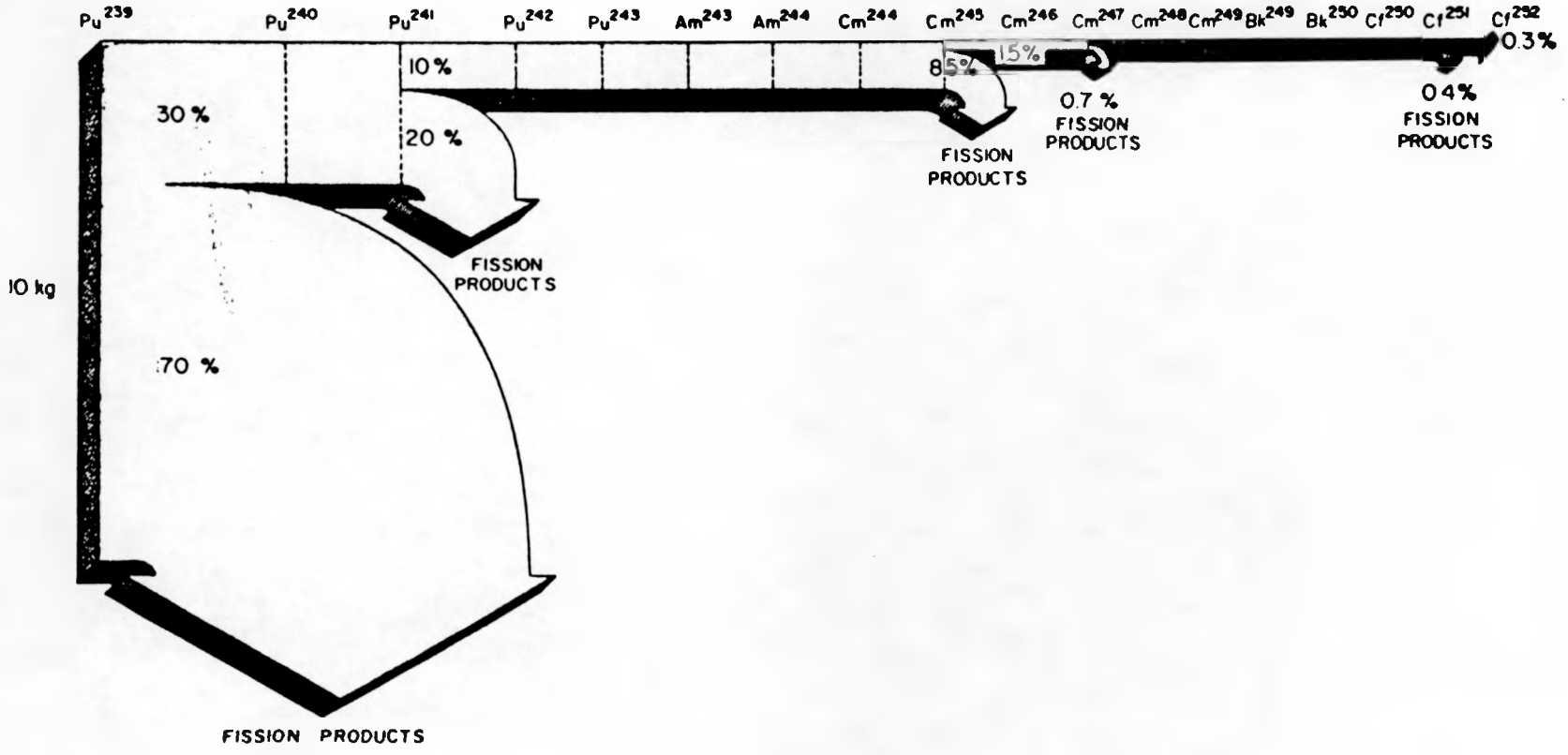


PRODUCTION OF TRANSURANIUM ELEMENTS

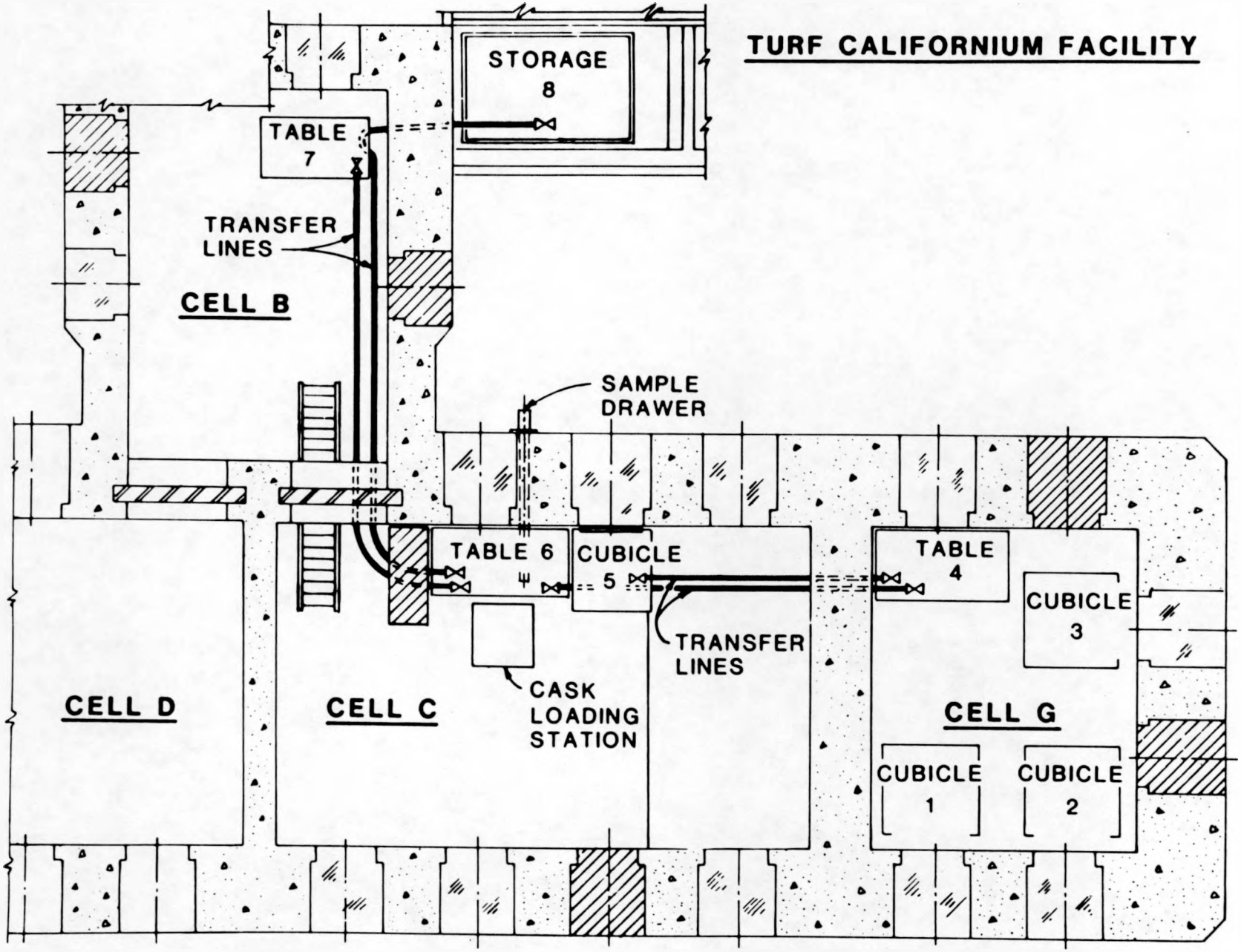


Fig. 6. Workstations of the REDC Californium Facility in Building 7930.

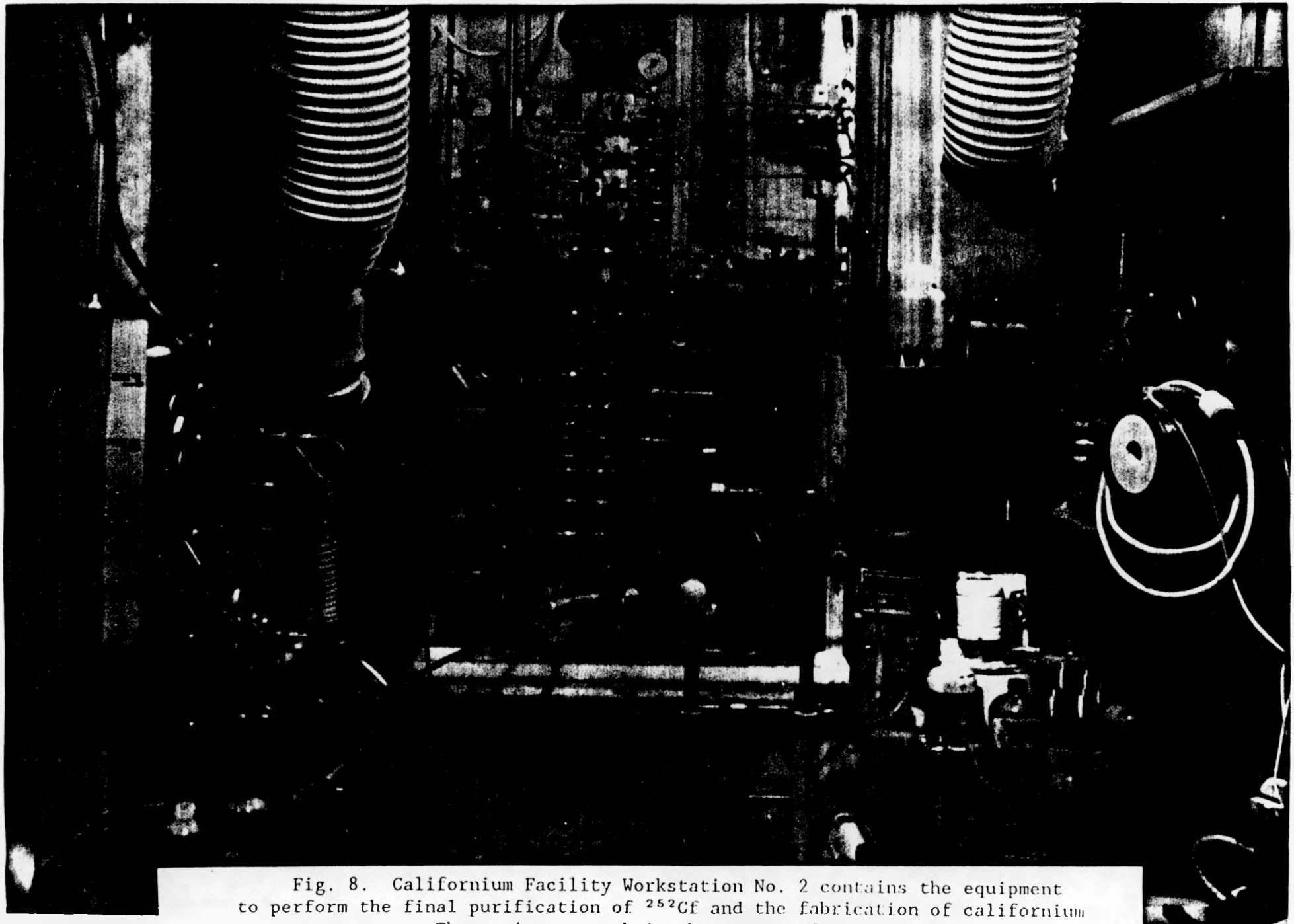
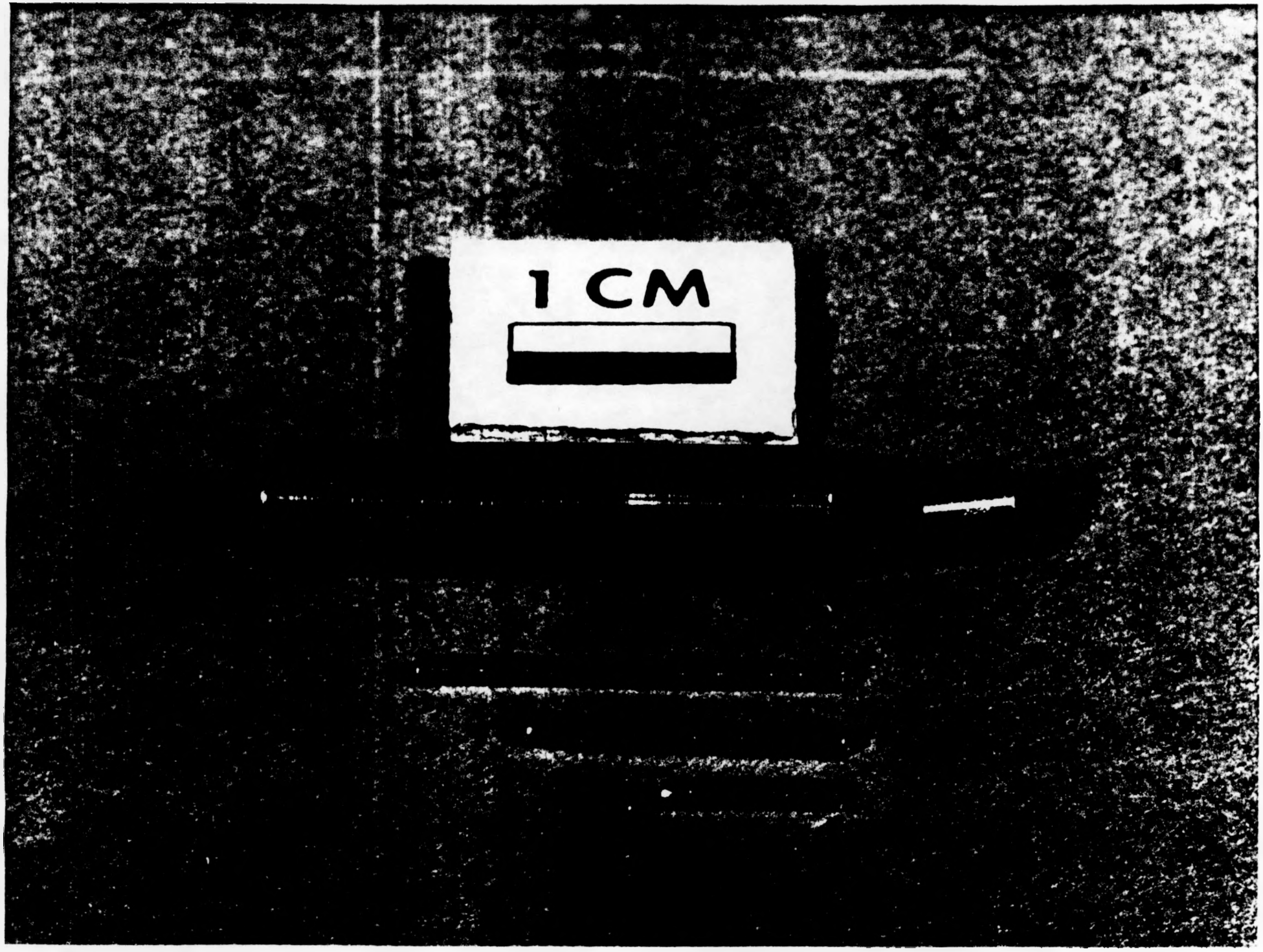


Fig. 8. Californium Facility Workstation No. 2 contains the equipment to perform the final purification of ^{252}Cf and the fabrication of californium neutron sources. The equipment rack in the center of the cubicle is used to carry out high pressure ion exchange separations.

0
1
2
3
CM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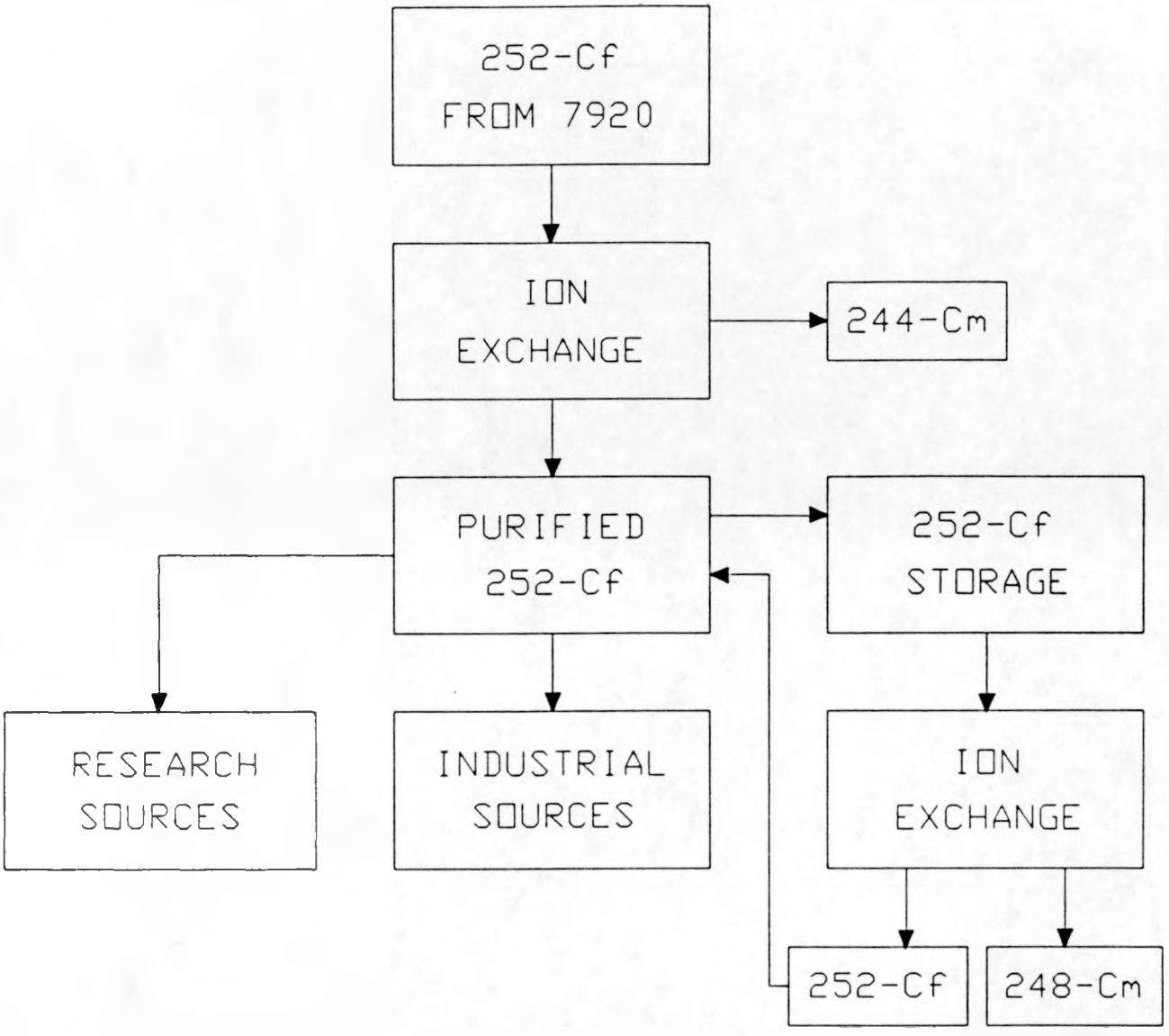
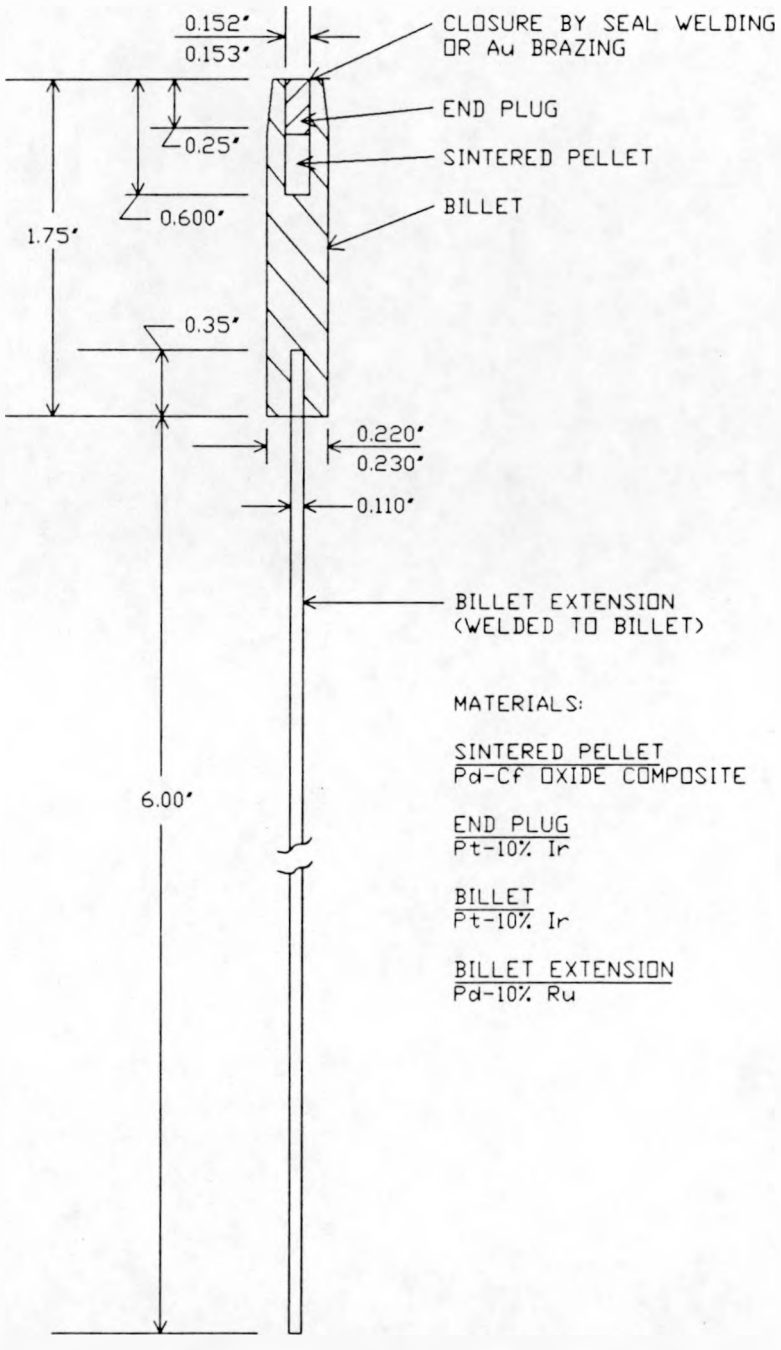
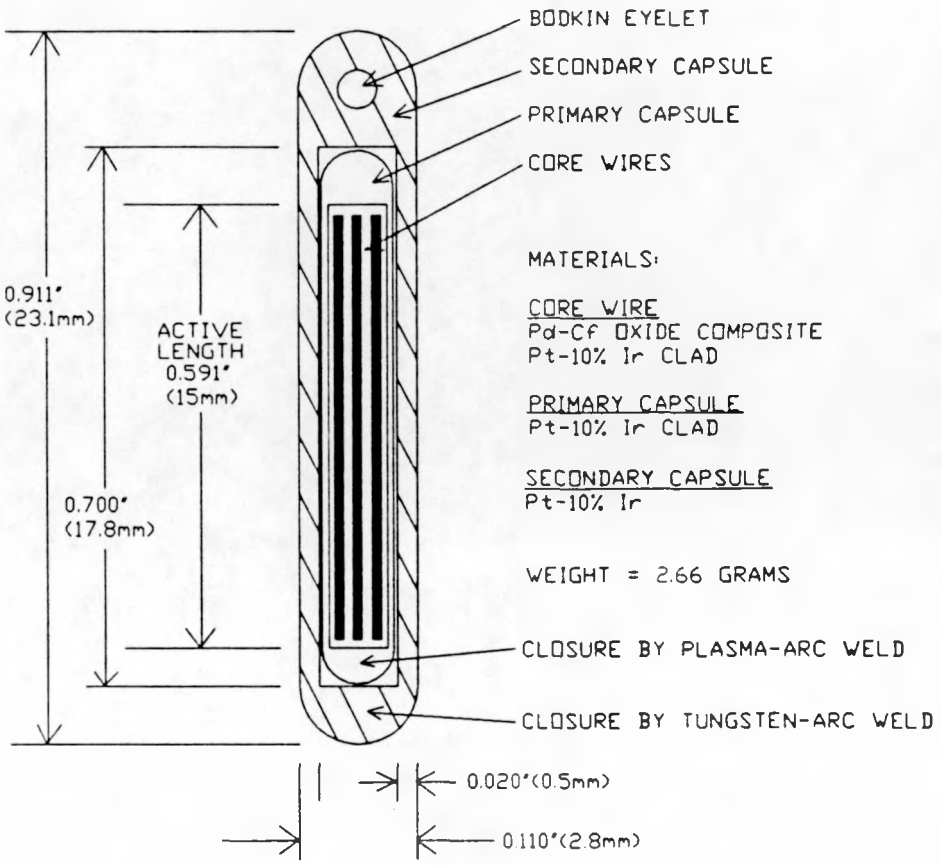


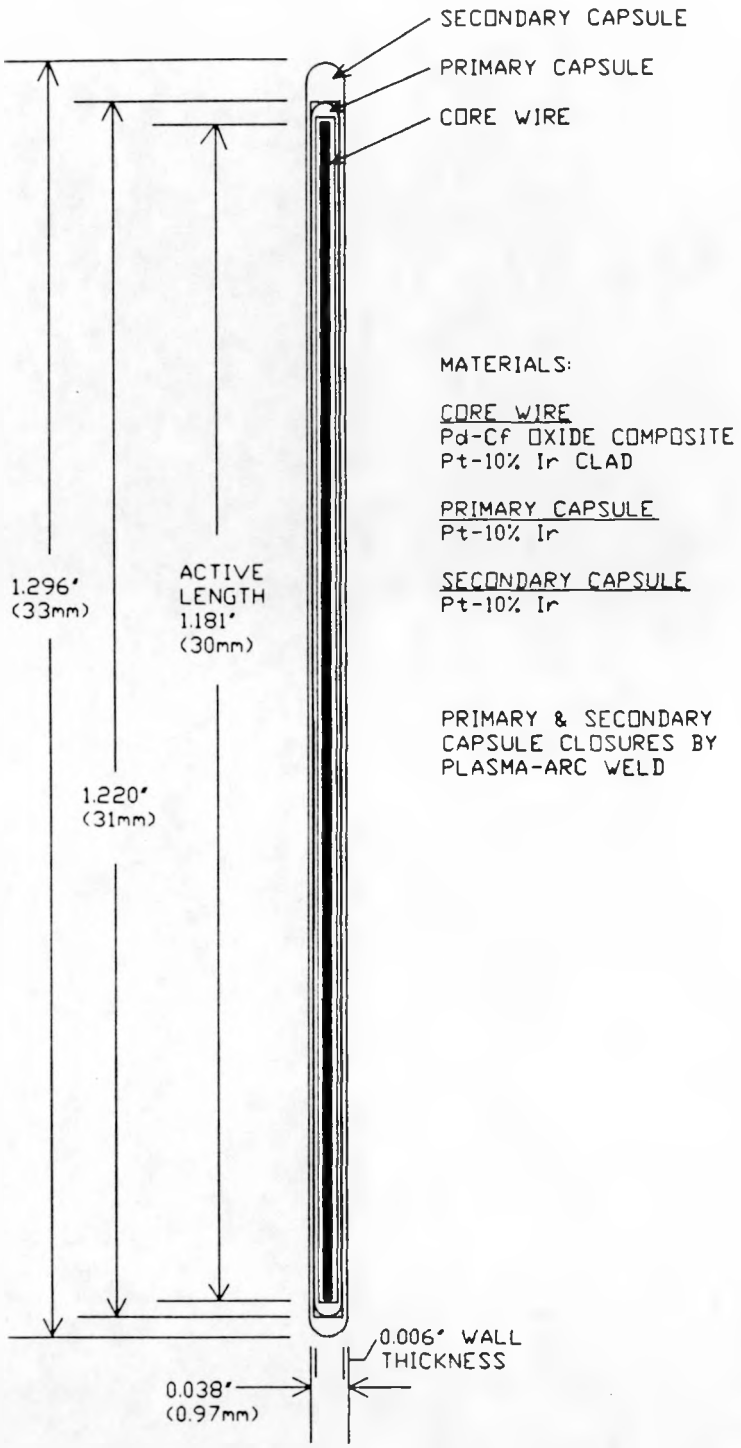
CALIFORNIUM FACILITY PROCESSING FLOWSHEET



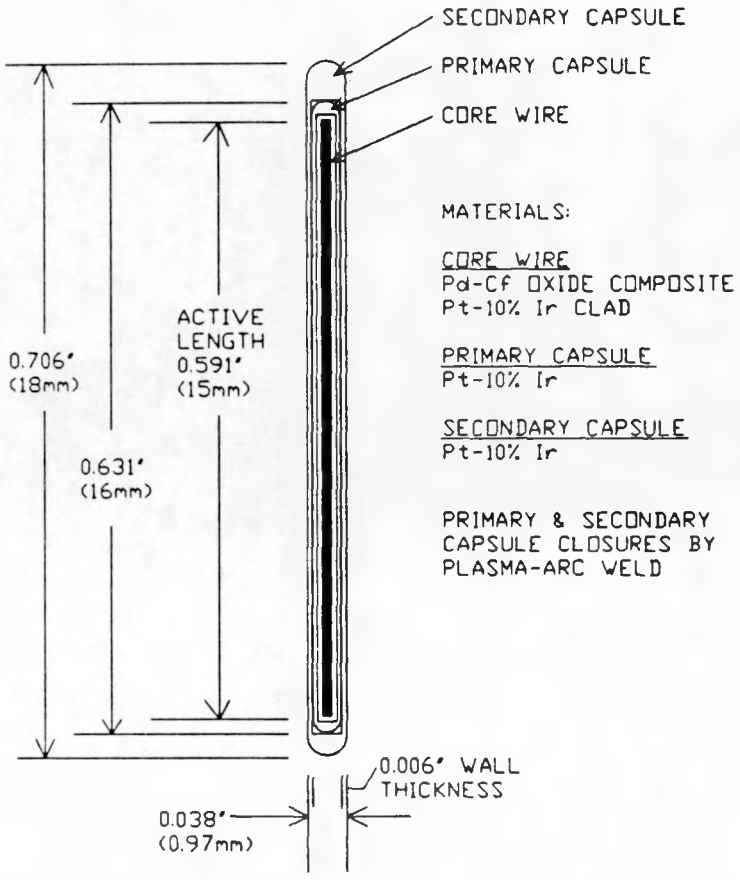
252-Cf MEDICAL SOURCE BILLET



252-Cf APPLICATOR TUBE (AT) ASSEMBLY



252-Cf AFTERLOADING CELL (ALC)



252-Cf SHORT AFTERLOADING CELL (SALC)