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Campaign 76—Production of ^{252}Cf and the Recovery of Curium Feed Material at the Radiochemical Engineering Development Center

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Isotopes and Fuel Cycle Technology Division

CAMPAIGN 76—PRODUCTION OF ^{252}CF AND THE RECOVERY OF CURIUM FEED MATERIAL AT THE RADIOCHEMICAL ENGINEERING DEVELOPMENT CENTER

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ACRONYMS

AHIB	alpha-hydroxyisobutyrate (complexing agent)
AIX	anion exchange
BX	Berkex extraction
CB	cut bottles
CF	californium nut column loading
CL	Cleanex extraction
CX	AHIB cation exchange
DBF	deep-bed filter
DS	dissolution
FDV	flow diversion valve
HCl	hydrochloric acid
HCV	hand control valve
IX	ion exchange
LLLW	liquid low-level waste
MDV	manual diversion valve
MUA	make-up area
OH	hydroxide precipitation
OX	oxalate precipitation
RE	rare earth
SX	solvent extraction

1. REDC FACILITY AND CAMPAIGN 76 PROCESSING OVERVIEW

Californium-252 is a strong neutron emitter with several specialized uses. It is produced in the High Flux Isotope Reactor (HFIR) by irradiating curium targets followed by several steps, collectively called a processing *campaign*. These steps involve chemical processing at the Radiochemical Engineering Development Center (REDC) to recover and purify Cf and Cm, and to fabricate new targets using recovered Cm. The ultimate goal of a campaign is to replenish the Cf stock for the Californium Production Program. This report describes the processing and results of Campaign 76.

1.1 REDC FACILITY

REDC (formerly known as the Transuranium Processing Plant) is a multipurpose, Hazard Category II nuclear facility [1], which began operation during 1966 in conjunction with HFIR. The HFIR–REDC complex was built following Glen Seaborg's vision of a unified national effort to irradiate ^{239}Pu to produce hundred-gram quantities of the isotope ^{244}Cm and then to re-irradiate the material in a very high flux nuclear reactor to support the nation's heavy isotope research and development. The REDC has two main buildings, Buildings 7920 and 7930, in which remote operations are conducted. Californium-252 recovery campaigns are conducted in Building 7920, which is the focus of this report. (Californium-252 bulk wire is fabricated as part of the Californium Production Program in Building 7930.) In addition to heavy isotope production, REDC focuses on medical and research isotope production and dispensing. The center also has a strong commitment to programs in fuel cycle development and national nuclear security.

Facilities in Building 7920 include two small stand-alone shielded hot cells designated as "caves," alpha glovebox labs, radiological labs, cold labs, and a bank of nine heavily shielded hot cells numbered 1 through 9 from east to west. The hot cells have corresponding enclosures in the front designated as cubicles 1–9 (Cubs 1–9) and pits below their cubicles. Hot cells 1 through 7 also have individual tank pits located behind their respective pits under the cubicles. In addition, there is a large tank pit behind hot cells 8 and 9 that contains large vessels for collection and treatment of process waste solutions. Each of the nine cubicles—where campaign process operations are performed—has 4.5 ft thick, high-density concrete walls, a multi-layered leaded-glass window, and a pair of through-wall manipulators with which equipment in the cubicles and radioactive material can be remotely manipulated with little-to-no exposure to staff. A conveyor system allows transferring materials between the cubicles via a chain-driven dolly that carries a canister along a track in a duct-like tunnel running underneath the cubicles along the front wall. Additionally, small pass-through ports allow direct passage between cubicles in adjacent cells. Irradiated curium targets from HFIR are lowered into the cell bank through a 5-inch diameter slug shoot at Cub 7. As they are processed, product material typically moves through the hot cells toward the east. Chemical processing and separations to recover Cf and Cm from target materials are performed in Cub 7, Cub 6, and Cub 4. Cub 3, Cub 2, and Cub 1 are used in the production of curium targets after chemical separations have taken place and the curium has been converted back to an oxide. As a final step, curium targets are inspected in Cub 1. Cub 8 is used for radioanalytical sample preparations and some types of sample analysis. Cub 9 is used mainly for solid waste collection, size reduction, and storage of bulk material unassociated with the curium targets. A cutaway rendering of the hot cells and its support areas can be seen in Figure 1.

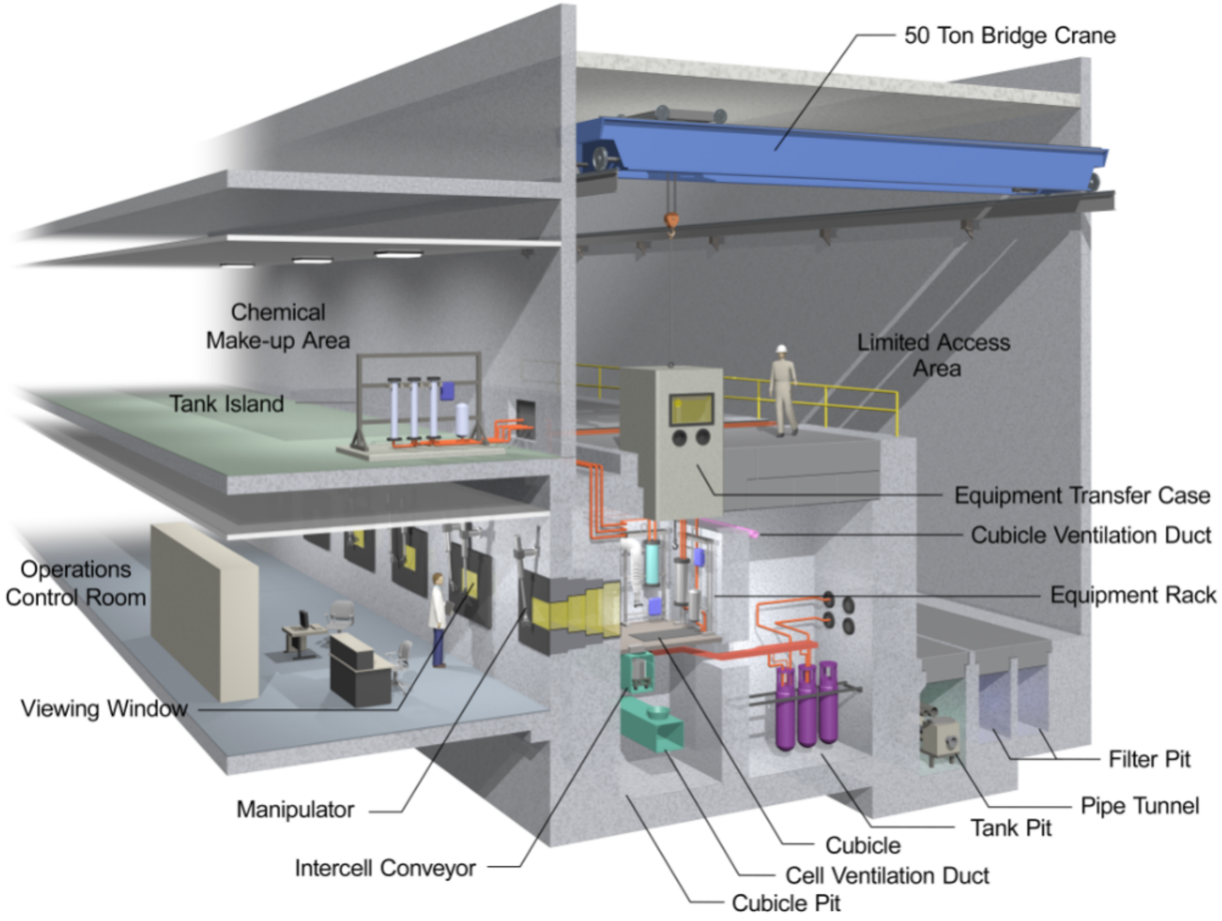

Figure 1. Cross-sectional view of the hot cell bank.

A network of tanks in the tank pits and cubicles facilitates chemical processing and waste collection. There also is a system of tanks in the make-up area (MUA) on the upper level where cold reagents can be prepared and fed to in-cell process lines and tanks. Tanks with a "T-" designation are process tanks within the tank pits and cubicles. Tanks designated with an "M-" are tanks in the MUA. Tanks with an "F-" designation were originally waste collection tanks. For this campaign, F-115 and T-23 were used to collect rework solutions for further processing. Tank T-23 is used for collecting rework material that is of higher activity than the material collected in F-115. For instance, a by-product solution may go to T-23 for collection; however, the water used to flush out the tank after emptying would be collected in F-115. In addition to the tanks, ion exchange columns are used during the chemical processing of a curium target campaign. These columns are positioned on racks in the cubicles and have a "C-" designation. Glass and polypropylene (poly) bottles (typically labeled with sequential numbers appended as Feed/Product #1, Feed/Raffinate, Cut #1, R-1, or as a circled number, mentioned herein as circle 1, etc.) also are used as convenient means to introduce reagents and to receive effluents for some in-cell processes.

1.2 CALIFORNIUM CAMPAIGN—GENERAL OVERVIEW

A campaign begins when several curium targets are irradiated in the "flux trap" of the HFIR—an unfueled region at the center of the annular core where fast neutrons are moderated and produce a region of very high thermal-neutron flux. Americium is included in the targets for transmutation to curium for replenishing the supply of curium target material; hence, americium and curium are sometimes mentioned together (i.e., Am/Cm) in describing a process or its feed/product as they behave similarly in most chemical processes. Targets irradiated in one campaign typically will have been fabricated at the REDC at the end of the previous campaign by blending actinide oxides (a ceramic) with Al metal—both in powder form—which, when pressed into a pellet form, binds the oxide forming the target *matrix*. The pellets are encased (or clad) in an Al jacket—which, together, comprise the *target*. An outer Al shroud is added to facilitate coolant flow around the jacket.

Targets are dissolved to recover transmuted products of irradiation and unconverted target materials (i.e., Cm). Before the matrix and clad are dissolved (designated as "DS" runs), Al coolant-flow shrouds are cut away from the targets and are dissolved using the caustic dissolution step in a separate operation serving two purposes: (1) test the dissolution system's operability and (2) provide a convenient means for disposing highly activated Al (sometimes referred herein as "scrap," as is the clad and matrix Al), which then may be sent to low-level liquid waste (LLLW). Target dissolution is a two-step process that separates actinides from Al target material and dissolves the actinides for further processing. In the first step, irradiated targets—along with unirradiated rework oxides and pellets from previous campaign(s)—are charged to the dissolver for a second caustic dissolution during which Al jackets and binders are dissolved, leaving behind actinide oxide solids. The second step is an acid dissolution of actinide oxides that yields a product in nitric acid solution.

The campaign continues with purification of dissolved transplutonium elements from assorted metallic impurities, as well as from much of the lanthanides and plutonium, using a liquid-liquid batch extraction version of the Cleanex process (designated as "CL" runs) [2]. The dissolver product—a nitrate solution that is prone to hydrolyzing actinides as precipitates—first must be converted to chloride form by evaporation followed by the addition of 12 M HCl, which will dissolve any hydrolyzed materials. Solution acidity is adjusted to 0.1 to 0.4 N by adding acid or sodium hydroxide (NaOH) as needed. NaOCl (or LiOCl) is added as an oxidant to ensure that all Mo in the feed is oxidized to the extractable Mo(VI) form. Molybdenum extraction avoids severe emulsion problems which can occur in the low-acid extraction step if any molybdenum is present. The organic extractant pulls trivalent actinide and lanthanide elements from the dilute, acidic aqueous phase into the organic phase, achieving up to 99% recovery with only one contact stage (although a second contact is common). The aqueous raffinate phase (with the assorted metallic impurities [mostly Na from feed adjustment] and much of the plutonium) is sent to LLLW. Scrubbing of the pregnant organic solution (solute) with dilute hydrochloric acid (HCl) helps remove undesired lanthanide and transplutonium elements dissolved in the organic phase. The solute is back-extracted—or stripped of higher actinides—with more concentrated HCL to prepare them for further purification and separation.

The main products—Cf for neutron sources and Cm for a future campaign's target material—are purified initially by ion exchange (IX) processing, designated as "CM" runs. First, Am/Cm and transcurium elements are loaded on a small column filled with a strong-base anion exchange resin that sorbs anionic actinide complexes formed in a concentrated LiCl solution. This chemistry yields a lanthanide effluent that is free of actinides, which can be disposed as waste. Elution of sorbed complexes provides an Am/Cm product free of lanthanides, and the transcurium elements are separated from residual lanthanide elements using a chromatographic separation based on the LiCl anion exchange (AIX) process. [3]. Routing of effluent solutions is determined by monitoring and tracking the ^{252}Cf neutron profile in the column. One or more primary IX runs provide an Am/Cm product free of lanthanides and transcurium elements while also preparing a lanthanide stream that is free of actinides for disposal. A secondary run further purifies the transcurium elements separated during the primary run(s). As a final step, purified transcurium elements from the secondary run are precipitated with LiOH to remove any residual LiCl and convert the transcurium product from a chloride solution to a small-volume nitrate (NO_3^-) solution with little free acid, suitable as feed for cation exchange separations.

Purified transcurium elements are separated by cation exchange ("CX") chromatography. The actinides first are loaded onto cation resin in a short loading column; unloaded resin sites are converted to ammonium form by treatment with NH_4NO_3 to ensure proper column chemistry during pH-sensitive elution. Subsequently, the elements are eluted and separated by pumping a complexing agent—alpha-hydroxyisobutyrate (AHIB)—at varying pHs through the column and then through a longer heated elution (or separation) column also filled with cation resin. Complexing strengths are different for each of the elements, so they move through the system at different rates, allowing them to form distinct bands as they progress through the longer elution column [4]. Band separations—identified using both the neutron probe (indicating the hourly position of Cf on both columns) and a flow-through alpha detector—facilitate their individual element collections in small, numbered bottles. The effluents are acidified, sampled, and analyzed individually to confirm separation and to facilitate their disposition as products, rework or waste. Products include cuts of pure Cf and mixtures of berkelium (Bk) and Cf. Bk is separated from the Cf to minimize the dilution of the ^{252}Cf isotope from in-growth of berkelium's ^{249}Cf decay daughter in those cuts.

The ^{249}Bk/^{249}Cf mixture is discarded if there is no suitable customer for purified Bk. If there is a suitable customer for ^{249}Bk, then the Bk is separated from residual Cf by a batch solvent extraction process called Berkex ("BX"). Before Berkex separation, the Bk–Cf mixture must be cleansed of any residual complexing agents used for its elution during the CX run; this is accomplished by cation exchange—using fresh resin in the same column employed during the CX runs—and results in an acidic nitrate solution amenable to the Berkex separation.

Pure Cf cuts are loaded into numbered "nut" packages for later processing into bulk wire, which can be cut conveniently to a selected length for a specified neutron-source strength. Each nut package consists of a Pt cylinder with fritted ends retaining cation exchange resin. For loading, each Pt cylinder is placed in a Swagelok™ fitting assembly—called a "nut package"—and loaded with up to 40 mg ^{252}Cf by pressurized transfer through the package. After displacing interstitial liquid with N_2 and venting to relieve pressure, the capsule is removed from its nut package, inserted into a firing wand, cured for 24 hours, and fired at a programmed temperature profile up to 700°C with vacuum-induced air flow. After cooling, the capsule is placed back into its numbered nut package for later pneumatic-line transfer to Building 7930 and further processing into bulk wire. Effluents from each primary loading run are collected in poly bottles that are monitored by neutron readings. Some californium in the feed typically resists loading, in which case, it is collected for a recycle cation exchange run (CX-3).

A campaign typically includes the conversion of Am/Cm to oxide and production of new targets for use in the subsequent irradiation and processing campaign. Californium-252 is dispensed in a bulk-wire form that is fabricated in a separate process performed at REDC but is *not* considered part of a californium processing campaign, *per se,* and is not included in californium-252-production campaign reporting.

The campaign of this report is summarized in the next section of this overview, including a top-level flow sheet depicting chemical processing of the targets and a tabulation of principal product recoveries. This is followed by a description of the report's organization and includes a brief discussion of run and sample nomenclature. The overview is followed by individual sections describing each process of the campaign.

1.3 CAMPAIGN 76—SUMMARY

The overall campaign is designated as C76, and individual process runs are designated using the campaign number (*sans* the leading "C") followed by a two-letter process designation and run number. For example, 76DS-1 is the first run of the dissolution process for Campaign 76.

Campaign 76 (C76) was launched with the irradiation of five Cm targets in the flux trap of HFIR. These targets were fabricated during the C75 campaign. Targets S-42, S-44, and S-46 were irradiated in HFIR cycles 454–456 (6/17/14–11/2/14) for a total of 6,538 MWD. Targets S-43 and S-45 were irradiated in cycles 455 and 456 (8/6/14–11/2/14) for a total of 4,363 MWD. The targets were retained in the HFIR pool for approximately 3 months following irradiation to allow ^{131}I to decay below acceptable transportation and facility-release thresholds. The five targets were moved into Building 7920 of REDC on January 7, 2015, and then moved into Cub 7 on February 18, 2015.

The campaign continued with chemical processing beginning February 23, 2015, with the dissolution of those targets and some rework material from C75. Processing continued with purification of the desired Cf product for sources and Cm for future target material—ending January 8, 2017, including an ~18 month pause from June 2015 to December 2016 (when delayed oxalate precipitations were conducted). Campaigns normally include the production of new targets to be used in subsequent irradiation and processing campaigns, as was the case for the targets irradiated and processed in C76 (which were fabricated as part of C75). However, no targets were fabricated during C76; Cm recovered during C76 was retained and added to Cm recovered during Campaign 77 (C77).

A top-level flow sheet for the C76 is shown in Figure 2 and depicts chemical processing of the targets by indicating major process steps with blocks; the blocks are connected by bold lines and arrows color-coded to the major constituents in that path. (Not shown in Figure 2 are all sources of rework solution from the chemical processes eventually collected in a single batch.)

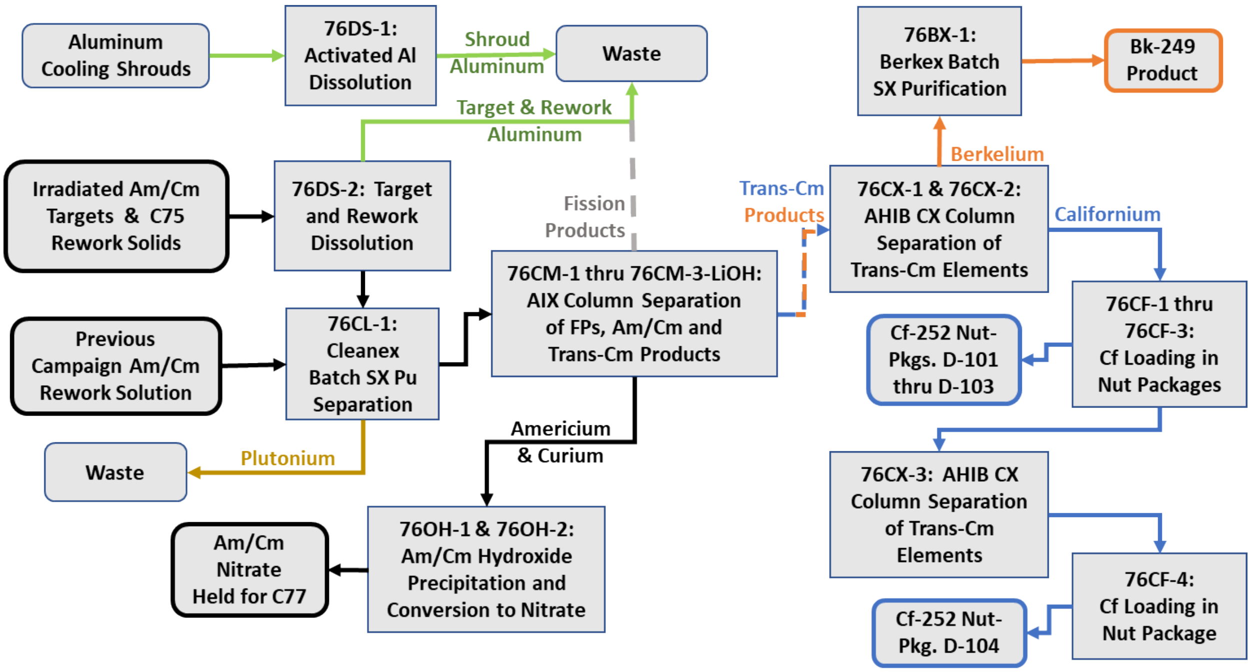

Figure 2. Top-level flow sheet of C76. Process acronyms: AIX, anion exchange; AHIB, alpha-hydroxyisobutyrate (complexing agent); BX, Berkex extraction; CF, californium nut package loading; CL, Cleanex extraction; CM, LiCl AIX; AHIB CXcation exchange; DS, dissolution; OH, hydroxide precipitation; SX, solvent extraction.

Campaign 76 produced ~100 mg of ^{252}Cf, which was transferred to Building 7930 hot cells for bulk wire fabrication. Approximately 16 mg of ^{249}Bk was recovered after further purification of the berkelium product in the alpha glovebox labs. Approximately 45 g of curium was recovered for later pressing into pellets, with ~11 g attributed to ^{244}Cm. No Cm oxide was produced for pellet pressing during C76. The curium product of the 76OX runs joined the feed for 77CL-1 in the next campaign.

According to target loading, irradiation modeling (blue background; from the C76 Plan [Appendix A], and calculations [Section 13]) and material recovery results, Table 1 depicts the overall inputs and recoveries of principal products for the C76 processing campaign (not necessarily in the order they may be presented in the more detailed discussion sections, which loosely follows the chronology).

Table 1. Summary of principal product recoveries* for C76

Sample Code	Process Component	Sample Date	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk (mg)
C76 Plan †	**C76 Input**		**3.9**	**0.8**	**46.5**	**10.0**	**110**	**24**
Planned	**Recovery**		**2**		**35**		**80**	**15**
CLAF-890	CL Feed	3/5/15	**3.25**	**0.71**	**44.9**	**10.6**	**102**	
BXBK-081	BX Product	5/12/15						**16**
CXCF-808	CX Cf Product / Nut Pkg Feed	5/19/15			0.02		**84**	
Neutron Assay	Nut Pkg Loads	5/16/15–5/27/15			0.002		**86**	
CMAP-308	OH Product	6/27/15	**3.58**	**0.78**	**44.1**	**10.4**	0.2	
OXDP-755	Recovered Rework	1/7/17	**0.6**	**0.12**	**4.3**	**1.0**	**6.8**	

* Average or median values are shown for the indicated sample groups—at lower precisions than indicated in more-detailed sections describing individual runs—mainly to track principal products of Campaign 76, and do not include waste materials (i.e., plutonium, structural metals, lanthanides, etc.). Parentheses indicate a value extracted from related samples (not the sample indicated). Short-lived isotopes (e.g., ^{249}Bk and ^{252}Cf) have been decayed to dates when the samples were acquired. Values are *not* included for outlier results or for results below limits of detection or for constituents with no related analyses.

† C76 Plan (Appendix A) values are a combination of calculated target contents (based on target loadings and irradiation conditions) and measured rework material from C75 (target scrap [pellets and powder] and chemical processing recycle [CY11T23CL-l]]). The value for Am is the sum of ^{241}Am and ^{243}Am in the plan.

1.4 CAMPAIGN 76—REPORT ORGANIZATION AND NOMENCLATURE

This report provides a brief description of chemical processes comprising this campaign but avoids much of the minutiae required for the evolution. The level of detail may vary in an effort to capture relevant details for understanding each process while avoiding redundancies when certain processes are repeated. Details indicative of off-normal operations, including some maintenance items, also are included to provide insight into difficulties encountered by staff during operations and the challenges in overcoming them—using only a pair of two-fingered manipulators—which are imposed by the hostile hot-cell/cubicle environment.

The balance of this report describes individual processes of the campaign. Processes are presented in a logical order in each of the following sections and may deviate from a purely chronological perspective. For processes with multiple runs, individual runs for the process are presented in subsections and are described in chronological order but may overlap in time with other runs involving the same process (i.e., preparations for one run may proceed while another run is being processed).

Each process also is depicted graphically in one or more run flow sheets that show operations as blocks, product streams as solid bold lines, rework and waste streams as solid fine lines, and sample streams as dashed lines.

Sample results are tabulated below the flow sheets to characterize the feed, product, rework, and waste for the process. A list of sample codes is provided in Appendix B. In some cases, sample results from product of a preceding run or process may be used to characterize the feed of a succeeding run or process. Samples shown in the flow sheets and in the tabulation are those deemed significant to understanding the process but do not necessarily represent all samples acquired during the operation. (Values are not included for outlier results or results below limits of detection.) Dates in the sample results tables typically represent the date the sample was taken but may reflect a date to which results were decayed.

A summary of C76 is provided as the last section (Section 13), before the appendices.

The nomenclature used in a campaign helps identify the process or material discussed:

- The chemical processes in a target campaign are designated by the campaign number (*sans* the leading "C" for Campaign), a two-letter process identifier and a sequence number. For instance, 76CL-1 is Campaign 76 Cleanex process number 1. If an additional Cleanex process run is needed, it would be designated as 76CL-2. Generically, dissolutions are designated by 76DS-n, curium recovery are designated by 76CM-n, etc. A complete list of process designations used in this report can be gleaned from the Table of Contents.

- Process samples typically are designated by four letters: the first two identify the chemical process, as above (e.g., DS, CL, CM, etc.), and the last two designate the function of the sampled solution in the process—for example, DP for dissolver product (DSDP), AF for A-column feed (CLAF; A-column is a historical designation for an extraction step), SR for scrub recycle (CLSR), etc. The four-letter designations are followed by a dash and "nnn," a sequential, three-digit number distinguishing the particular sampling of process solution. Replicate samples of a solution are appended by a capital letter, beginning with "A" (no dash). Resampling is assigned a new three-digit sequential value. Miscellaneous samples are designated by three letters; most are designated MSA-nnn for miscellaneous aqueous samples.

- Rework collections are recovered continually during a protracted Cleanex run throughout the campaign. They are designated by calendar year ("CYyy") collection began, collection tank (Tanks F-115 and T-23 are used as collection tanks and can accommodate batch extractions), process (Cleanex or CL), a sequence number, and a letter denoting the aqueous extraction step. For example, CY15T23CL-1A began collection during calendar year 2015, in tank T-23, as part of the first Cleanex process in that calendar year (number 1); the "A" denotes the first series of collections in the tank. Throughout the campaign, miscellaneous aqueous solutions are added to one of the two rework collection tanks (which are preloaded with organic extractant) and given a sequential-letter suffix each time the tank is subjected to a batch extraction to recover product constituents from the newly added aqueous (i.e., sparged to mix the two phases, settled to allow phases to separate, and decanted of the aqueous phase).

Most process details are excerpted from the Non-Nuclear Facilities Division (NNFD) 7920 Facility logbooks, which are detailed records of hot-cell control-room activities. The dates indicate the beginning of the 12 hour shift during which specific activities were performed, but the dates do not necessarily indicate the date that the action occurred (i.e., in cases when the action occurs after midnight on the night shift). All sample results are provided by the Chemical Sciences Division. Further calculations on sample results have been performed as necessary, and results reported herein typically indicate values that, for short-lived isotopes (e.g., ^{249}Bk and ^{252}Cf), have been decayed to the dates when the samples were acquired. Many (but not all) tank flushes and rinses are indicated; all such flushes and rinses are mixed in the tank by air sparging, typically for 10–30 minutes, but usually are not mentioned in the description.

2. 76DS—ALUMINUM SHROUD AND TARGET DISSOLUTION

Aluminum shrouds, cut previously from the targets, were dissolved in a caustic solution during 76DS-1. The five full target rods and 67 reject pellets and reject Am/Cm powder from Campaign 75 were dissolved in two-steps as 76DS-2. Rework solutions were collected in Tank F-115 as part of CY15F115CL-1B. Highly activated Al scrap (dissolved shroud, jacket, and matrix from both caustic dissolution steps) was sent to LLLW.

2.1 76DS-1—DISSOLUTION OF ALUMINUM COOLING SHROUDS (SCRAP)

2/20/15–2/25/15

The preparation and check-out of processing equipment in Cub 7 for the dissolution of the Al cooling shrouds was performed between 2/20/15 and 2/23/15. The inspection of the lid O-ring from dissolution vessel T-70 revealed that a replacement was needed, which was done. Concurrently, process solutions were prepared in the chemical MUA beginning with 7.2 L of a 2.25 M $NaNO_3$ solution in tank M-2 after the tank was flushed with water. In addition, 4.24 L of a 10 M NaOH solution was prepared in a separate 25 L poly container labeled MUA-P25-017, and a sample was submitted to Chemical Sciences Division with sample identification DSJD-228 to be analyzed for hydroxide concentration. Sample results showed the solution to be in the acceptable range for use. Dissolution process preparations continued in the MUA with the installation of new poly lines for metering in solutions into T-70. Preparations in Cub 7 continued with installations of new poly blocks at several positions, a new hand valve (HV) at HV-T737-1, new ferrule flat gaskets at F-70, and a polyvinyl chloride (PVC) ball valve on the P72010 GN. Following the installations, the T-70 to F-111 route was set up, and the correct configuration was confirmed by testing with water. During route testing, several routing problems were encountered for the transfer of water through T-737. The transfer of water was slow or nonexistent from T-70. There were multiple attempts to troubleshoot the apparent blockage in the system, and an alternate route through F-70 to F-111 was established and determined to be effective. Final preparations concluded by confirming instrument operation, NaOH addition routes to T-70, and the T-70 heating/cooling water loop.

The cooling shrouds were cut away from the Al targets on 2/20/15 using the target shroud cutter in Cub 7. On 2/23/15, the shrouds were added to T-70 after it was charged with ~7.2 L of 2.25 $NaNO_3$ (mentioned above) and a 0.514 L water flush from M-2; T-70 then was set to heat to 92°C. Once the temperature reached 92°C, 10 M NaOH was metered into T-70 from M-573 at a flowrate of 1–1.2 L/h to affect scrap dissolution. After ~5.2 L of NaOH and subsequent water flushes (2 L) were added to T-70, the temperature in T-70 was increased to 103°C, maintained at 103°C for 30 minutes, and then cooled to 60°C.

Transfer of the dissolved scrap Al solution from T-70 to F-111 required three transfers of approximately 6 L each via the T-737 and F-70 systems. F-70 was back flushed, and a series of flushes from M-704 were routed through systems leading to T-70, collected in 5.5 L batches, and sparged in T-70 before transfer to F-111. Three repetitions of these flushes were also routed to F-111.

2.2 76DS-2—DISSOLUTION OF TARGET

2/24/15–3/11/15

Step 1. Since the same equipment setup used for the dissolution of the shrouds also was used for the target jacket and Al matrix, only new reagents had to be prepared. This involved making up 12.35 L of 2.25 M $NaNO_3$ solution in tank M-2 and 6.56 L of 10 M NaOH in a 25 L carboy labeled “P25-017.” The

NaOH solution was sampled as DSJD-229 for hydroxide concentration and was found to be in the acceptable range.

On 2/25/15, the 12.35 L of 2.25 *M* $NaNO_3$ were transferred to T-70 and followed with ~0.9 L of water, and T-70 was heated to 60°C. Also added to T-70 were the five target rods (S-42 through S-46) along with reject pellets from C75 (i.e., the contents of Twist Lock Container J (containing six heavy-Cm[1] pellets and heavy-Cm oxide powder for six more pellets), Twist Lock Container V containing 27 heavy-Cm pellets from C75CO-7), Twist Lock Container Y ; (containing 25 heavy-Cm pellets from C75CO-8), and Twist Lock Container FF ; containing 1 light-Cm pellet from C75CO-9) and Pipe Nipple 28 (containing 8 light-Cm pellets). All input materials were verified using the two-person verification protocol, added to T-70, and verified to be in T-70 (as opposed to being hung up in transfer lines or supports). The temperature in T-70 then was set to 92°C and monitored while the 6.56 L of 10 *M* NaOH was transferred to M-573. Once T-70 reached 92°C on 2/26/15, 10 *M* of NaOH was metered from M-573 to T-70 at a flow rate of 1–1.2 L/h. Temperature and pressure readings of T-70 were recorded every 30 minutes. The addition was finished after the 6.56 L of 10 *M* NaOH was added to T-70, at which point the air purge in T-70 was stopped and M-573 was flushed with 2 L of water to T-70. The digest was started when the T-70 water loop reached 103°C. The digestion was completed after 30 minutes, and T-70 was set to cool to 60°C.

The transfer of the dissolved Al waste solution from T-70 to F-111 via F-70 began on 2/26/15 and was completed on 2/27/15. The solution was dark gray in color with no solids evident. A series of three caustic flushes of T-70 was carried out during that span, which included a 2 × 1.5 L of 1 *M* NaOH back flush of F-70 to T-70 along with additions of 1 *M* NaOH solution into T-70, similar to those in 76DS-1. Those solutions were sparged, heated to ~90°C, held at 90°C for 2 hours, and cooled to <60°C. The cooled solution was flushed from T-70 to F-111 via F-70. During the second caustic flush, a valve stem of hand control valve– (HCV-) P70-1 in Cub 7 broke off. In the attempt to remove and replace HCV-P70-1, HCV-P70-2 valve stem also broke. So, two new valves and gaskets were slugged[2] into Cub 7, and both valves were replaced. The third and final caustic flush was performed after the repairs were completed. Water flushes then were performed by back flushing F-70 to T-70, followed by addition of 2 × 1.5 L of water to T-70 from M-704. T-70 was then sparged for 10 minutes at a high rate, allowed to settle, and then the water was transferred from T-70 to F-111. The water flushing operations were completed four times followed by five back flushes from F-70 to T-70 with a total of 5 L of water. After all dissolutions and additions to F-111 were complete, F-111 was sparged via the F-111 sample position (due to inoperable tank sparger) and sampled on 2/28/15 as DSJW-379A and DSJW-379B. The solution was slightly cloudy, but no solids were evident. The initial results were deemed skeptical due to possible issues with the sample, so F-111 was sparged for more than 4 hours and resampled in triplicate as DSJW-380A, DSJW-380B, and DSJW-380C. The results indicated the solutions were acceptable for disposal and were subsequently sent to waste collection tank F-126.

Step 2. Before dissolving the actinide oxides on 2/27/15, the ORNL Shift Superintendent and HFIR control room were notified of pending dissolution and to expect increased readings on the 7911 stack instrumentation. The dissolution then began with three 2.5 L additions of concentrated (15.7 *M*) HNO_3 to

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[1] In the context of americium/curium target and pellet materials, heavy curium is a nominal distinction for curium with a majority of its mass (typically greater than ~70%) comprised of heavier isotopes, ^{246}Cm and ^{248}Cm. Conversely, light curium is a nominal distinction for curium with a majority of its mass (typically greater than ~70%) comprised of lighter isotopes, ^{243}Cm, ^{244}Cm, and ^{245}Cm. (Note, the ~70% delimiter itself is a nominal value such that heavy-curium or light-curium can be found where a ⅔ or ¾ delimiter was used.)

[2] Slugging is a relatively convenient method for safely inserting a bagged group of items into a cubicle. It involves bagging the items compactly (to a maximum 5” diameter by 36” length, so the package will fit in a chute) and lowering the package by a string affixed to the top according to a procedure that ensures maintaining cell confinement.

T-70 via the F-70 back-flush route with 15 minutes allowed between additions. These were followed by three 1.5 L additions of water to T-70, also via the F-70 back-flush route. A low sparge was initiated and T-70 was set to heat to 103°C and held at 103°C for 3 hours. T-70 was cooled to 40°C, the sparge was terminated, and the dissolver solution was sampled as DSDP-560A, DSDP-560B, and DSDP-560C on 2/28/15. On 3/2/15, several 8 *M* HNO_3 flushes of F-70 to T-70 were done to ensure that all the dissolved actinides were in T-70. Additional samples, DSDP-561A and DSDP-561B, were taken of T-70 after the back flushes, following the original samples (DSDP-560A/B/C) taken of T-70 immediately after the dissolution.

On 3/3/15, the transfer route from T-70, through F-720 (a 10 µm filter), to T-72 was connected and leak tested; the transfer was made of the dissolved actinide solution to T-72. T-70 was flushed twice with 3 L of 4 *M* HNO_3 and twice with 3 L of 1.0 *M* HNO_3 to T-72 via F-720—sparging each flush in T-70 for 15 minutes. Three liters of water was added to T-70, sparged 30 minutes, and sampled as DSDF-293. The first water flush was sent to T-72, and another 3 L was added to T-70, sparged and sampled as DSDF-294 and sent to T-72. T-72 heated to concentrate solution to ~30 L—ended with 30.65 L on 3/4/15.

The filter used in the T-70 to T-72 transfer, F-720, was flushed twice with 5 L of 1 *M* HNO_3 from Cub 7 solution addition line and each addition was sent to T-72. Filter, F-720, then was flushed nine times, each with 5 L 1 *M* HNO_3 and allowing the acid to reside on the filter 5 minutes for each flush before transferring to T-79, and finished with a 5 L flush of water, also sent to T-79. Upon completion, T-79 was sampled as PVC-177, and the contents of the tank were transferred to the F-115 rework tank and collected as part of CY15F115CL-1B.

F-70 was back flushed with 4 × 3 L of 8 *M* HNO_3, letting a full F-70 volume of acid sit for 15 minutes on the filter on each flush before sending it to T-70. This was followed by 3 × 2 L of water. Next, T-70 was sparged slowly, heated to 100°C for 6 hours, cooled to <50°C, and sampled as DSDP-562.

Figure 3 illustrates the processes used to dissolve the aluminum cooling shrouds (76DS-1) and the target rods (76DS-2). Table 2 lists the sample results for 76DS-1 and 76DS-2.

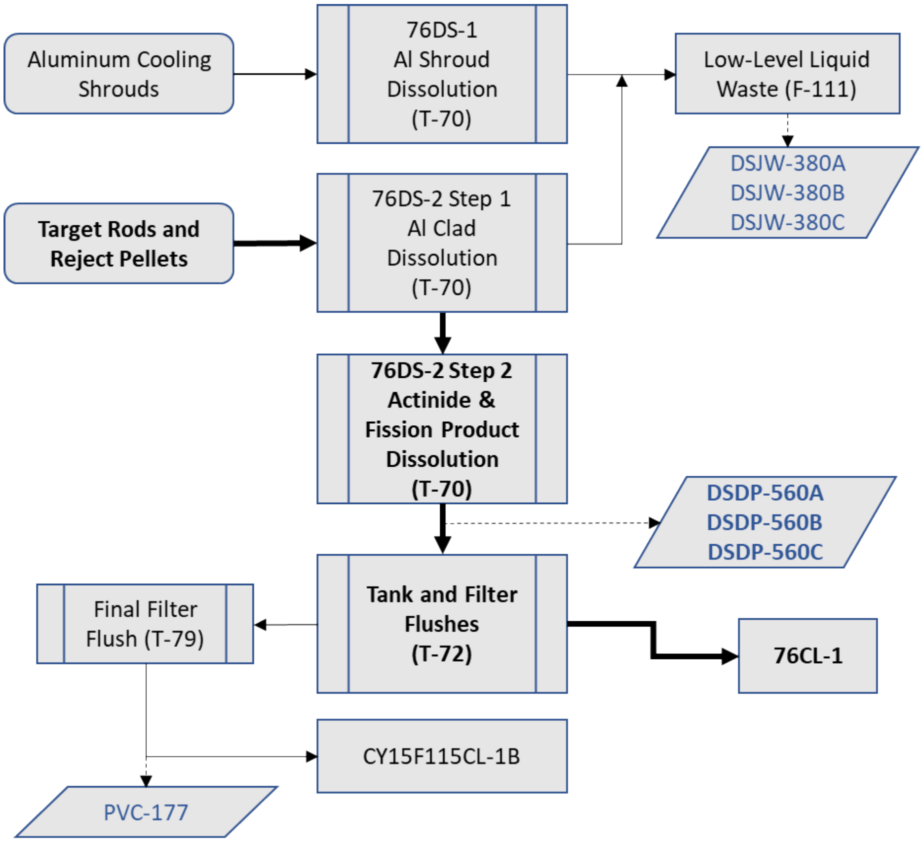


Figure 3. Target and rework material dissolution flow sheet—76DS-1 and 76DS-2.

Table 2. Sample results* for 76DS-1 and 76DS-2

Sample Code	Date	Tank No.	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk† (mg)	^{90}Sr† (Ci)	^{137}Cs (Ci)	^{144}Ce (Ci)	^{154}Eu (Ci)
DSJW-380A	2/28/15	F-111	0.030	0.000		0.006	0.001			32.5	5.77	0.206	
DSJW-380B	2/28/15	F-111	0.036	0.000		0.006	0.001			33.3	5.36	0.136	
DSJW-380C	2/28/15	F-111	0.023	0.000		0.006	0.001			30.8	5.36	0.119	
DSDP-560A	2/28/15	T-70	1.475	0.703	0.084	40.836	9.497	90.627	21.727		3.08	141.6	0.874
DSDP-560B	2/28/15	T-70	1.665	0.741	0.088	43.035	10.008	94.064	24.007		3.21	120.7	1.041
DSDP-560C	2/28/15	T-70	1.452	0.520	0.062	39.382	9.159	90.635	21.963		3.16	104.1	0.999
PVC-177	3/5/15	T-79	0.091	0.007	0.001	0.027	0.006	0.033			0.001	0.002	0.001

* Values are *not* included for outlier results or results below limits of detection. Zero values indicate rounding *down*, nominally from the next digit (i.e., 0.0001 < value < 0.0005 would be rounded down to 0.000).

† Berkelium-249 results shown for DSDP-561A and DSDP-561B were included from the prior samples, DSDP-560B and DSDP-560C. (DSDP-560A was considered an outlier result at 13.217 mg.)
Strontium-90 results for samples DSJW-380A and DSJW-380B were included from the prior sampling (DSJW-379A and DSJW-379B) because strontium-90 was not identified for any of the DSJW-380A/B/C sample set.

3. 76CL—CLEANEX BATCH SOLVENT EXTRACTION FOR TRANSPLUTONIUM PURIFICATION

Dissolved transplutonium elements (C76) and rework solution from C75 were purified from assorted metallic impurities, as well as much of the plutonium, using the Cleanex batch solvent extraction process [2]. All organic-aqueous contacts were performed in one tank, T-72.

Feed for the Cleanex process (76CL-1) was prepared by boiling off excess nitric acid in the dissolver product—to achieve a dilute acid feed conducive to extracting transplutonium elements—and then adding HCl to convert the actinides from a nitrate form to a chloride form for further downstream processing. The adjusted feed was contacted by a batch of extractant—di-(2-ethylhexyl) phosphate (HDEHP) at 1 *M* HDEHP in a diluent of Exxsol D60 (a normal paraffinic hydrocarbon) to remove trivalent actinide and lanthanide elements from the dilute acid/salt solution. Typical extraction contacts involved ~60 minutes of sparging followed by ~10 minutes of quiescence. The raffinate samples indicated that the raffinate contained 224 mg of ^{244}Cm, which was above the desired ≤150 mg target for disposal; consequently, it was retained along with the scrub solution.

Scrubbing the pregnant organic solution with dilute (0.03 *M*) HCl helped to flush impurities (entrained from the extraction step) into the aqueous remnants below the organic layer. Since sodium is included in relatively large amounts from feed adjustment, scrubbing the organic layer also helped remove sodium that may have extracted into the HDEHP. Typical scrub contacts involved ~30 minutes of sparging followed by 10 minutes of quiescence. The solute was stripped of higher actinides with more concentrated (6.7 *M*) HCL to prepare them for further purification and separation.

3.1 76CL-1—CLEANEX FEED ADJUSTMENT

3/2/15–3/7/15

Before starting the solvent extraction process, all material to be recovered was collected in the tantalum-lined evaporator tank, T-72. First, a rework solution from Campaign 75 (C75; CY11T23CL-l) was added to T-72, evaporated down to 15.67 L, and sampled as CLAF-889. Then, material from 76DS-2 was transferred to T-72 as described in Section 2.2, adding to the rework material already in the tank. Then, tank T-72 was sparged and sampled in triplicate as CLAF-890A, CLAF-890B, and CLAF-890C.

To remove excess acid contained in the feed, T-72 was set to evaporate down to a volume that was 3–4% above the instrument zero (actually 1%). Once the volume in T-72 had reached ~5%, the tank level was maintained with the continued evaporation of T-72 while 10 L of water were metered in at about 1 L/h. This continued the removal of excess acid in the feed. Evaporation was concluded after the last of the water was added, and the acid digest to redissolve any hydrolyzed materials was initiated by adding 1.25 L of concentrated (12 *M*) HCl to T-72. T-72 was heated and sparged before 9 L of water were added to bring the total volume to 14 L for acid analysis (CLAF-891). The results from CLAF-891 indicated the acid was at 1.97 *M* (27.6 mol total). To adjust the solution acidity to the range needed (0.1–0.4 *M*) an acid adjustment was made by adding 4.28 L of 5.3 *M* NaOH flushed with 0.5 L of water to T-72. The tank was sparged and resampled as CLAF-892, which indicated an acidity of 0.33 *M*.

An oxidant solution of NaOCL (2.5 L) was added to T-72, flushed in using two rinses of 0.25 L of water. The tank was sparged and set to heat to 75°C–85°C. After the desired temperature was reached, the solution was held at 75°C–85°C for 30–40 minutes. The completion of the oxidation and digestion of the feed concludes the feed adjustment step. The feed then was ready for extraction.

3.2 76CL-1—CLEANEX EXTRACTION

3/7/15–3/10/15

The 76CL-1 run was started on 3/8/15 by adding 30 L of 1.1 *M* HDEHP extractant prepared in tank M-3 via M-71 to the feed solution in T-72. The addition of the extractant was followed by two separate flushes of 0.5 L water. Mixing of the aqueous and organic phases in T-72 was achieved by sparging air through the sampling line. (While preparing for the transfer, about 4 L of unknown solution was observed in M-71; this was collected in a carboy [MUA-G19-004] and stored in the MUA hood.) M-71 was rinsed with alcohol (2 × 250 mL), which was disposed. Upon transfer from M-3, only 24.65 L was found in M-71 (not the expected 30 L). This was made up by returning the extractant to M-3 and augmenting it with a 3.33 L of HDEHP and 2 × 3.33 L of Exxsol D60. After mixing, extractant was sampled as CLEX-402 and analyzed to be 1.1 *M* HDEHP, which was deemed acceptable for use.)

After 30 minutes of sparging T-72, 750 mL of 5.3 *M* NaOH was added to lower the acidity of the aqueous solution from a calculated value of 0.28 *N* H^+ to about 0.1 *N* H^+. Keeping the free acid low was important to help drive extraction of actinides to the organic phase, and to avoid potential hydrolysis, precipitation of unextracted actinide constituents, or emulsification in the mixture. The tank was sparged again to affect extraction. A sample of the aqueous solution was taken as CLER-300 and found to have a free acid concentration of 0.11 *N*; however, 440 mg of ^{244}Cm remaining in the aqueous phase was well above the goal of ≤150 mg to declare the extraction process complete. Therefore, another addition of 5.3 *M* NaOH (215 mL) was made to T-72 and the tank was sparged again and sampled as CLER-301. Results showed an acid concentration of 0.11 *N* after fixing the titrator, and a ^{244}Cm value of 298 mg, which was still above the 150 mg goal. Further adjustments were advised by research personnel and the Ops Manager toward an acidity of 0.07 *N*. This was attempted by adding 174 mL of 5.3 *M* NaOH to T-72 with two flushes of 0.25 L of water and sparging T-72. A sample was taken of the aqueous solution as CLER-302 and revealed an acid concentration of 0.08 *N* and 204 mg ^{244}Cm. A decision was made to perform only one final addition of 5.3 *M* NaOH (136 mL) to T-72. That was followed by the usual sparging and settling with a sample taken as CLER-303 before the extraction step was declared complete.

Before the sample results from CLER-303 were received from Chemical Sciences Division, the aqueous solution in T-72 was transferred to T-23. This was done after an initial attempt was not successful because of a bad flow diversion valve (FDV-T-70-2). The valve was replaced, and a clean phase separation was made at ~28% in T-72. Transfer systems to both T-23 and T-72 were flushed with water, and the remaining organic solution in T-72 was set up for scrubbing. CLER-303 revealed that the final ^{244}Cm value in the aqueous phase was 224 mg.

3.3 76CL-1—CLEANEX SCRUBBING

3/10/15–3/12/15

Scrubbing of the pregnant organic solution was completed to flush impurities entrained from the extraction step into the aqueous remnants below the organic layer. Since sodium was added in relatively large amounts, scrubbing the organic layer also helped to remove any amount of sodium that may have been extracted by the HDEHP.

A scrubbing solution of ~7.5 L of 0.03 *M* HCl (7.5 L H_2O and 25 mL of concentrated HCl) was added to T-72 and flushed in with 2.5 L of water. T-72 was then sparged with air for 30 minutes via the sample line and allowed a 10 minute settling period before the aqueous phase was sampled as CLSR-924. The results indicated an acid concentration of 0.056 *N* H+ and ^{244}Cm as 200 mg. These values met the requirements for disposition for future rework, so the first scrub in T-72 was transferred to T-23 and the transfer system was flushed both ways (to T-23 and to T-72) with ~1 L of water each way.

A second addition of the scrubbing solution (~7.5 L of 0.03 *M* HCl followed by 2.5 L of water) was added from M-704 to T-72 and subsequently sparged for 30 minutes. The tank was allowed to settle for at least 10 minutes, and then it was sampled as CLSR-925. Results indicated an acid concentration of 0.057 *N* H+ and 163 mg of ^{244}Cm. Like the first scrub, transfer of T-72 to T-23 was acceptable, so the transfer was completed and the transfer system flushed both ways (to T-23 and to T-72) with ~1L of water each way. Later in the campaign, T-23 was acidified with 600 mL of HCl, sparged, and then sampled as CLSR-926 to evaluate the contents of the tank.

Before the scrubbing was concluded, an organic phase modification was made. This was done by adding 5 L of 0.2 *M* Adogen to T-72 followed by two 0.5 L water flushes. The Adogen solution was a mixture containing 3.6 L of Adogen 364 and 1.4 L of Ultra-Low Naphthalene Aromatic 150 used as the diluent. This solution was used as a phase transfer catalyst to retain the Fe(III) in the organic phase and to prevent it from following the trivalent transplutonium elements through the stripping steps. The solution in T-72 was sparged for at least 15 minutes before the stripping steps commenced.

3.4 76CL-1—CLEANEX STRIPPING

3/12/15–3/18/15

Before stripping the organic phase in T-72 that contained the trivalent actinides and lanthanides, the transfer routes were set up and leak checked with water. The route from T-72 to T-604 via the T-734 system confirmed to be leak free. A leak was noted during the testing of the T-604 to T-43 transfer route via pump P-653 coming from the Jaco® fitting on the P65301 gooseneck (GN) and had to be tightened to stop its leaking. No further leaks were noted. With the system ready and T-72 already sparging, the first strip solution was added to T-72 from M-704 and followed by a 0.5 L water rinse. The strip solution was 9 L of 6.7 *M* HCl plus 500 mL of 30% hydrogen peroxide (H_2O_2). The addition of the H_2O_2ensures that the berkelium was reduced to the trivalent state so the organic could be sufficiently stripped.

Sparging of T-72 was continued after the addition of the first strip solution for an extended period because the slower-flowing sample line was used as the sparging apparatus. The solutions in the T-72 were allowed to settle for at least 30 minutes, and then the aqueous solution from the first strip was transferred to T-604 with a clean phase separation. Before the first strip was transferred to T-604, 5 L of Exxsol D60 with Red O dye was added to T-604. The Red O solution in T-604 allowed for clear identification of the organic and aqueous phases during the phase separation and transfer of the product solutions from T-604, through the PS-604 phase separator and P-653 to T-43. After the first strip was transferred from T-72 to T-604, a second strip solution containing 4.5 L of 6.7 *M* HCl and 250 mL of 30% H_2O_2 was added to T-72, and the tank was sparged and then allowed to settle. Each sparging–settling cycle was at least 30 minutes.

While the second strip of the organic in T-72 was ongoing, solution in T-604 was transferred to T-43 using P-653 and a phase separation in P-604. The transfer of the first strip from T-604 to T-43 was discontinued once T-43 reached 18 L in liquid volume. At that point, the transfer route was flushed both ways with 0.5 L of water, and T-43 was set to evaporate to provide freeboard for subsequent strip additions. Once sufficient room was achieved in T-43, the second strip was transferred to T-43 after a phase separation via T-604 and P-653.

The above sequence—stripping the organic in T-72 with 4.5 L of 6.7 *M* HCl and 250 mL of 30% H_2O_2, followed by a transfer of the aqueous strip solution to T-604, and then transferring the separated aqueous product through the P-604 phase separator and P-653 and on to T-43—was repeated six more times for a total of eight strip passes. Evaporation of T-43 was initiated whenever freeboard in the tank was needed. During the transfer of the seventh strip from T-604 to T-43, no suction could be applied to P-653, so the pump for P-653 (P-16) was changed out with the pump from P-651 (P-17), and the transfer was completed.

After the eighth strip of the organic solution in T-72, a sample of the aqueous solution was taken as CLCP-422. A sample of the remaining organic in T-72 was taken as CLWO-119 once the aqueous strip was transferred to T-604. All specified limits for ceasing stripping operations were met according to the results of the aqueous solution in T-72 (CLCP-422). Also, results from the sample of the organic in T-72 indicated that gross alpha and neutron limits were met, but process-control limits set for the 5.15 and 5.50 MeV alpha energies (to ensure meeting material recovery goals) were exceeded.

After the 8th strip was transferred from T-604 to T-43, a confirming flush of T-604 was completed by adding 4.5 L of 6.7 *M* HCl plus 250 mL of 30% H_2O_2 and sparging for at least 30 minutes. The aqueous solution in T-604 was sampled as CLCP-423 and then transferred to T-43 using the P-653 phase separator. The organic in T-604 was sampled as CLWO-120 after the aqueous transfer to T-43, and after sparging at least 30 minutes a few days later, it was resampled in the organic phase as CLWO-121, which showed a reduction in the alpha activities at 5.15 and 5.50 MeV sufficient to proceed. On 3/17/15, T-43 was sparged for longer than 1 hour and then sampled in triplicate as CLCP-424A, CLCP-424B, and CLCP-424C for accountability. Evaporation of T-43 was initiated after sampling, and it was discontinued once the tank volume reached approximately 5 L.

The Cleanex operations were completed later in the campaign (May 2015) after the disposition of the LLLW solutions generated during the Cleanex process (Figure 4). The organic solution in T-72 was transferred to T-20, followed by two 4 L flushes of Exxsol D60 and a single water flush of 10 L. Afterward, three water flushes of T-72 were completed and sent to LLLW via tank F-115. The organic in T-20 then was neutralized with 15 L of 4 *M* NaOH. After this, the contents of T-20 were sent to F-126, along with subsequent flushes of Exxsol D60 and water, after verification the solution was caustic by sampling as MSA-750. The final water flushes of T-20 were sent to F-115. Similar operations were done with T-604, as 5 L of 3 *M* NaOH were added to neutralize the contents before it was transferred to F-126. That transfer was followed by a single flush of Exxsol D60 and three flushes of water. Sample MSA-749 was taken to verify the solution in T-604 was caustic before transferring to F-126.

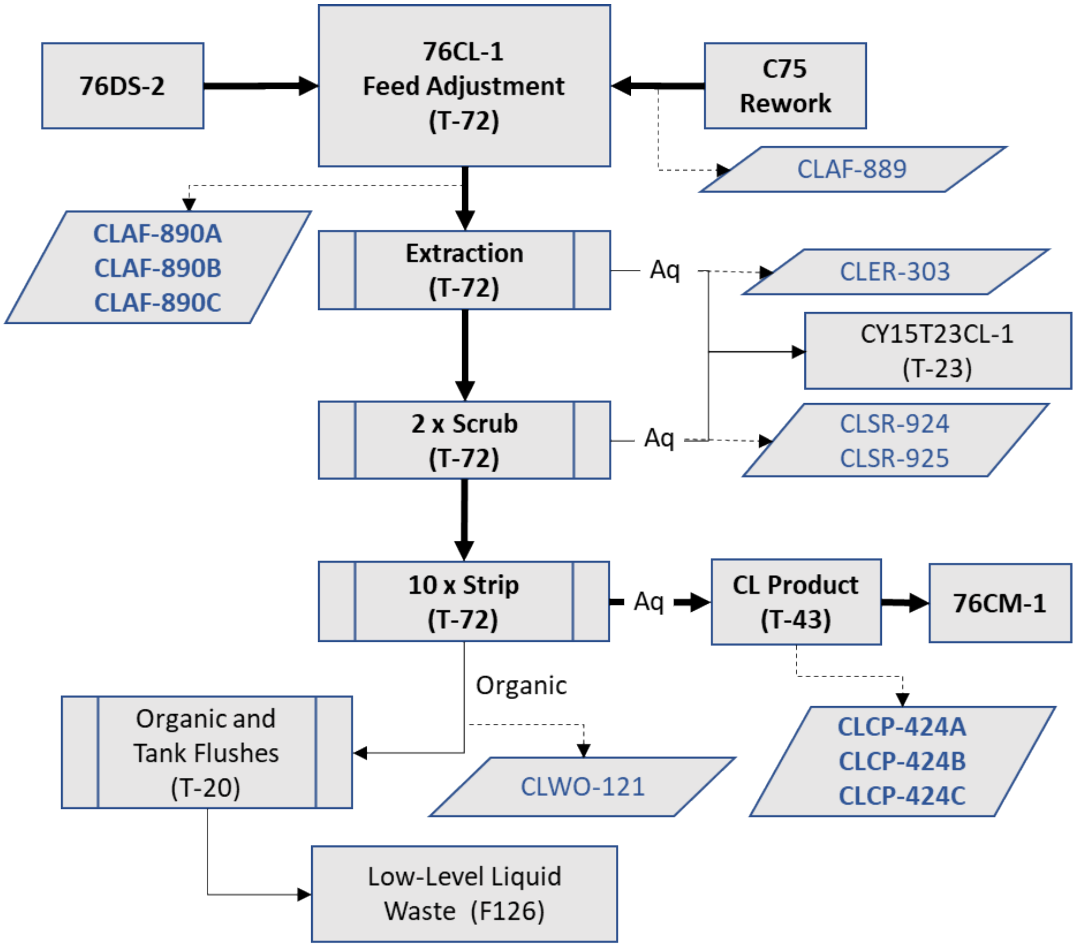


Figure 4. Cleanex batch extraction process flow sheet for separating plutonium and impurities (all organic-aqueous contacts performed in T-72)—76CL-1.

Table 3. Sample results* for 76CL-1

Sample Code	Date	Tank No.	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{137}Cs (Ci)	^{144}Ce (Ci)	^{154}Eu (Ci)
CLAF-889	3/2/15	T-72	1.483	3.000	0.650	6.435	1.519	11.713		1.116	0.906
CLAF-890A	3/5/15	T-72	1.042	3.597	0.779	44.877	10.591	105.30	3.184	127.4	1.783
CLAF-890B	3/5/15	T-72	1.727	3.083	0.668	43.621	10.295	101.79	3.057	121.0	1.656
CLAF-890C	3/5/15	T-72	0.626	3.254	0.705	45.030	10.627	101.78	3.057	126.1	1.656
CLER-303	3/10/15	T-72	0.053	0.105	0.027	0.964	0.002	0.104	2.907	1.914	0.078
CLSR-924	3/11/15	T-72	0.000	0.001	0.000	0.009	0.002	0.003	0.063	0.027	0.003
CLSR-925	3/12/15	T-72	0.001	0.001	0.000	0.007	0.165	0.002	0.008	0.019	0.001
CLCP-424A	3/17/15	T-43	0.728	3.262	0.707	48.736	11.502	109.14		143.6	1.634
CLCP-424B	3/17/15	T-43	1.342	3.262	0.707	48.739	11.503	109.14		148.5	1.683
CLCP-424C	3/17/15	T-43	0.569	3.329	0.721	45.401	10.715	106.42		143.6	1.584
CLWO-121	3/22/15	T-72	0.774			0.001	0.000	0.005		0.088	0.009

* Values are *not* included for outlier results or results below limits of detection. Zero values indicate rounding *down*, nominally from the next digit (i.e., 0.0001 < value < 0.0005 would be rounded down to 0.000).

4. 76CM—ACTINIDE AND LANTHANIDE SEPARATION BY LICL-BASED ANION EXCHANGE

Curium, rare earth fission products, and transcurium elemental groups were separated in a series of anion exchange column runs, 76CM-1 through 76CM-3, based on a LiCl anion exchange (AIX) process [3]. This process uses a strong base, anion exchange resin—Dowex 1 × 10 packed in a tantalum column (C-636) located on the right rack of Cub-6—to sorb anionic chloride complexes formed in a concentrated (nominally 12 *M*) LiCl solution. The complexes for each element form at varying strengths in the feed, which allows each to move through the column at different rates and facilitates chromatographic separation with appropriate eluants. This chemistry yields a lanthanide effluent that is free of actinides and can be disposed as waste. Elution of sorbed complexes provides an Am/Cm product that is free of lanthanides and transcurium elements.

Two primary runs (76CM-1 and 76CM-2; each one half of the feed) were performed to accommodate the amount of Cleanex product to be purified. Chromatographic operation of the anion exchange column during the primary runs facilitated separation of fission products, Am/Cm and transcurium elements by preferentially eluting the element groups using progressively weaker concentrations of LiCl eluent solution: rare earth (RE) elements were eluted nominally at 10 *M* LiCl, Am/Cm at 9 *M* LiCl, and transcurium elements with 8 *M* HCl (i.e., with no LiCl). (Dilute amounts of other constituents—0.1 *M* HCl and hydroxylamine hydrochloride for RE and Am/Cm, and 0.1 *M* H_2O_2 for transcurium elements—were present in the elutriants.) These two primary runs also prepared a RE stream for disposal that was free of actinides and that could be, and was, sent to the LLLW system. Curium products from both primary runs were later transferred (05/17/15) to T-60 to make up feed for the 76OH runs.

A third run (76CM-3) was performed using C-636 with fresh AIX resin to further purify the transcurium elements recovered during the two primary runs. Transcurium product then was converted to a nitrate form by precipitation using LiOH followed by re-dissolution using concentrated nitric acid.

The routing of effluent solution was determined by monitoring the neutron profile of the column, once every hour, during elution and flushing to track the ^{252}Cf as it migrated down the length of the column. Data for these scans were graphed as a function of column height and are included for each run.

4.1 76CM-1—LICL-BASED ANION EXCHANGE—1

3/6/15–3/31/15

Set up and systems check began on 03/06/15 with the replacement of all HCVs, hand valves, FDVs, and MDVs on the right rack in Cub-6; a new special FDV, 7PDV Type D and two polyvinyl chloride ball valves with poly line jumpers to serve T-604; and four sample needle blocks. Fresh Dowex 1x8, 200-400 Mesh "B" Fraction resin (previously treated with HCl) was charged to Column C-636 per NNFD-7920-OP-166. The nut for SA-T634 swing clamp would not tighten and, after trying a second nut, it was suspected that the stud had a bad thread at ~2.5 turns. Log does not indicate that stud was replaced but T-634 would not sample on 4/9/15. Although no resolution to the swing clamp problem was logged, T-634 was able to be sampled later in the run (on 04/10/15 as CMAP-304).

On 03/18/15, stock LiCl, synthetic feed, pretreatment flush, elutriant, and column flush solutions were made up as needed according to procedure NNFD-7920-OP-164 and confirmed by sample analysis. The 76CM-1 feed was pretreated using 2.67 L of 7.5 *M* LiCl, heated to boiling per NNFD-7920-OP-169, and sparged at 8% until temperature reached 80°C. Evaporation continued until the boiling temperature reached 142.7°C; then it was cooled to <50°C and 190 mL of 12 *M* HCl plus a 200 mL flush of 12 *M* LiCl was added. Next, it was sparged at 8% for 15 minutes. After testing the route integrity with water, the route from T-43 was set through the 1 µm graded-density floor filter to a three-prong flask. T-43 and T-65 were flushed and sparged with 4 × 0.75 L of pretreatment flush (12.1 *M* LiCl and 0.9 *M* HCl) and T-65 was flushed using P-653. On 03/19/15, T-65 was sparged at 8% for >30 minutes and sampled as CMAF-172A, CMAF-172B, and CMAF-172C. The floor filter was back flushed with 4 L of 0.5 *M* HCl, and then 4 L of water was flushed to T-43, and 2.5 L of 12 *M* HCl and 2 × 1.25 L water was added to T-43 via the decontamination line. After evaporating T-43 down to 10 L, then cooling to <50°C, and sparging for >30 minutes, T-43 was sampled as RECF-061.

On 03/26/15, T-43 was sparged at 7% for >1 hour and sampled as CMCR-580 under NNFD-7920-OP-169. On 03/27/15, fresh Dowex 1x8, 200-400 Mesh "B" Fraction resin (previously treated with HCl) was charged to Column C-636 per NNFD-7920-OP-166 after being prepared over the previous 2 days per NNFD-7920-OP-167. On 03/28/15, LiCl AX checks and feed pump test (per NNFD-7920-OP-168) revealed that the process pump at P-452 required replacement. An available pump from Cub-4 was used after unsuccessfully trying three others already in Cub-6. Also, no buildup was indicated in T-630 after several 1 L and 2 L water transfers from M-706 with no evidence of a system leak. At the same time FC-M67 would not turn on. The operations manager allowed continuing and used level depletion in M-67 as a workaround for these system checks. A leak was found and repaired in the T-630 level instrument line, which then indicated proper buildup on the T-630 level recorder. Also, the HCV block and toggle stem at HCV-T630 were replaced as part of the system checkout.

On 03/30/15, the LiCl AX RR6 feed was adjusted in T-65 according to NNFD-7920-OP-170. Column C-636 was pretreated the next day with synthetic feed from T-636 via P-638 per NNFD-7920-OP-171, and the feed adjustment was completed. Feed was loaded onto column C-636 and collected effluent in T-61. The column was eluted per NNFD-7920-OP-172 with the following suspensions:

- RE Elutriant (5 × 0.5 L) was directed to T-61 until the last 0.3 L, which was switched to T-630, to start the first pre-curium cut.

- The first 2 × 0.5 L Cm Elutriant was directed to T-630 to continue the pre-Cm until the last 0.2 L, which was switched to T-633 to start the curium cut. (***No*** second pre-Cm cut was warranted based on little or no evidence [i.e., a "gamma-caused" rise in the alpha trace] of rare earths present in the first Cm cut).

- The other 6 × 0.5 L Cm Elutriant was routed to T-633 as the Cm cut until last 0.2 L, which was switched to T-632 for the transcurium cut.

- The first 2 × 0.5 L of Cf Elutriant (to displace Cm Elutriant heel) then 1 × 1 L of Cf Elutriant through the column and to T-632 until last 0.2 L, which was switched to T-72 to start the column flush.

During the column run, the column was scanned hourly with a neutron probe to track the ^{252}Cf as it migrated down the length of the column, beginning with scan 1. Data for these scans are shown in Figure 5 with the neutron count rate shown as a function of column height. The graph was set up to accommodate 10 scans, although only 7 (scans 1–6 and scan 10) were needed to complete the operation.

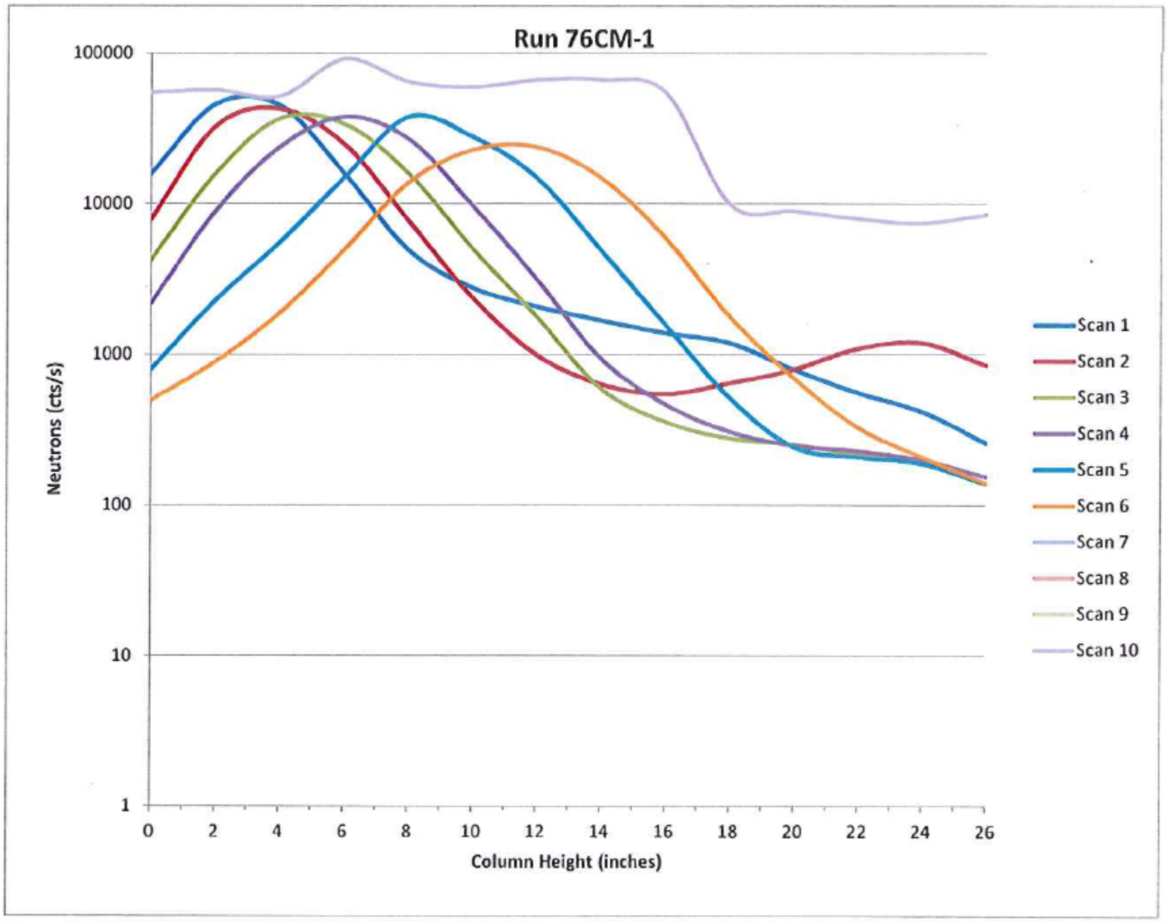


Figure 5. Neutron probe scans of the exchange column tracking material migration in 76CM-1.

76CM-1 elution samplings were performed on 3/31/15 (Figure 6). First 0.1 L of 12 *M* HCl plus 0.2 L of water was added to T-630, sparged for 30 minutes, and sampled as CMCR-581. The sample was light blue in color. After sparging for 30 minutes, T-632 was sampled as CMBP-152. While sampling T-632, the syringe sampler came apart at the ball-joint stem and had to be repaired. Next, 0.4 L of 12 *M* HCl plus 0.2 L of water was added to T-633, sparged for 30 minutes, and sampled as CMAP-303 (which was mostly clear with a slight tint). Next, 0.5 L of 12 *M* HCl followed by 2 × 0.5 L of water was added to T-61. It was sparged for 30 minutes and sampled as CMCR-583. Then, T-72 was sparged for 30 minutes and sampled as CMCR-582. Finally, T-72 was boiled down in follow-up operations to provide freeboard for collecting additional rework solution during 76CM-2, and column C-636 was left charged with resin.

Problems discovered during 76CM-1 were diagnosed and repaired between 4/1/15 and 4/6/15 in anticipation of 76CM-2.

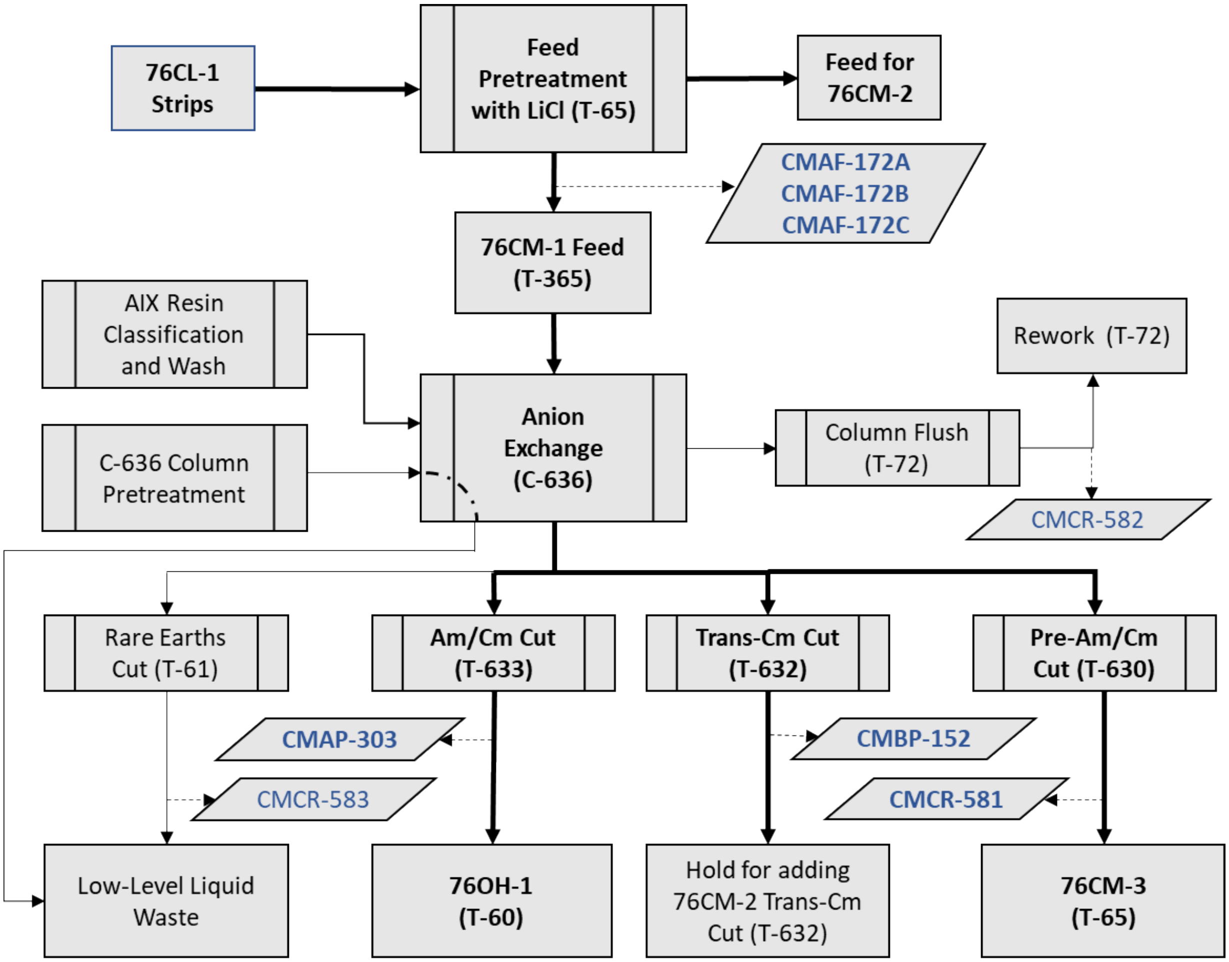


Figure 6. Flow sheet of anion exchange group separations of actinides and lanthanides—76CM-1.

Table 4. Sample results* for 76CM-1

Sample Code	Date	Tank No.	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk (mg)	^{144}Ce (Ci)	^{154}Eu (Ci)
CMAF-172A	3/20/15	T-65	1.176	2.954	0.640	44.043	10.394	97.622		132.3	1.441
CMAF-172B	3/20/15	T-65		3.494	0.757	42.757	10.091	91.104		134.6	1.535
CMAF-172C	3/20/15	T-65	0.882	3.049	0.661	40.956	9.666	91.115		125.2	1.394
CMAP-303	3/31/15	T-633		1.380	0.299	17.375	4.101	0.028		0.245	
CMBP-152	3/31/15	T-632		0.067	0.015	0.904	0.213	41.233			
CMCR-581	3/31/15	T-630	0.008	0.013	0.003	0.386	0.091	0.001		0.730	0.008
CMCR-582	3/31/15	T-72	0.046	0.003	0.001	0.020	0.005	0.517		0.027	
CMCR-583	3/31/15	T-61	0.003	0.000		0.002	0.001	0.005		57.94	0.655

* Values are *not* included for outlier results or results below limits of detection. Zero values indicate rounding *down*, nominally from the next digit (i.e., 0.0001 < value < 0.0005 would be rounded down to 0.000).

4.2 76CM-2—LICL ANION EXCHANGE—2

4/6/15–4/13/15

Set up and system checks began on 4/6/15 with activities paralleling those for 76CM-1 (reagent and elutriant makeups, valve, and sample block change outs, leak checks and repairs, system flushes, etc.), but *without recharging resin to Column C-636*. The alpha probe was confirmed to be operational, but the neutron probe had to be replaced. As before, the feed was adjusted per NNFD-7920-OP-170 in T-65 by boiling to a final temperature of 142.5°C (2.58 L after cooldown to <40°C) and acidified it with 253 mL of 12 *M* HCl followed by 200 mL stock LiCl. After sparging, heating was initiated to digest at 120°C, but the heating was ramped up too fast. Consequently, the steam pressure controller was set to zero to let it settle, and heating was restarted with a temperature rise set to ~1°C/min., with sparging on at 7% until 110°C was reached. Then, the temperature was at 120°C for 10 minutes. *The adjusted feed was not resampled*; 76CM-1 feed sample results were used. The final volume, 2.54 L, was transferred to T-635 and followed by 3 × 0.5 L of synthetic feed. Column C-636 was pretreated on 4/8/15 per NNFD-7920-OP-171.

Feed was loaded onto Column C-636 at 2 L/h. with effluent directed to T-61 (Figure 8) on 4/9/15 per NNFD-7920-OP-172. When the hot feed level in T-635 reached <0.2 L, 3 × 0.5 L of synthetic feed was added from T-65 to T-635 and was pumped through column C-636 to T-61. While continuing to pump from T-635, the column was eluted with the following.

1. RE elutriant (5 × 0.5 L), all but the last batch directed to T-61. Some of the last batch (<0.2 L) was switched to collect in T-630 and start the first pre-Cm cut

2. The first ~0.3 L Am/Cm elutriant to T-630 to continue the pre-Cm cut

3. The last <0.2 L Am/Cm elutriant, switched to T-634 to start the Am/Cm cut, and another 5 × 0.5 L Cm elutriant was directed to T-634, followed by 2 × 0.25 L Cm elutriant (except the last <0.2 L which was redirected from T-634 to T-632 for the transcurium cut) (***No*** second pre-curium cut was warranted based on little or no evidence [i.e., a “gamma-caused” rise in the alpha trace] of REs present in the first curium cut)

4. The first 2 × 0.5 L of Cf elutriant (to displace Cm elutriant heel) then 1 × 1.6 L of Cf elutriant through the column and to T-632 (except for last 0.2 L, which was switched to collect in T-72 as an initial column flush)

During the column run, the column was periodically scanned with a neutron probe to track the ^{252}Cf as it migrated down the length of the column, beginning with scan 1. Data for these scans are shown in Figure 7 with the neutron count rate shown as a function of column height. The graph was set up to accommodate 10 scans, although only 8 (scans 1–8) were needed to complete the operation.

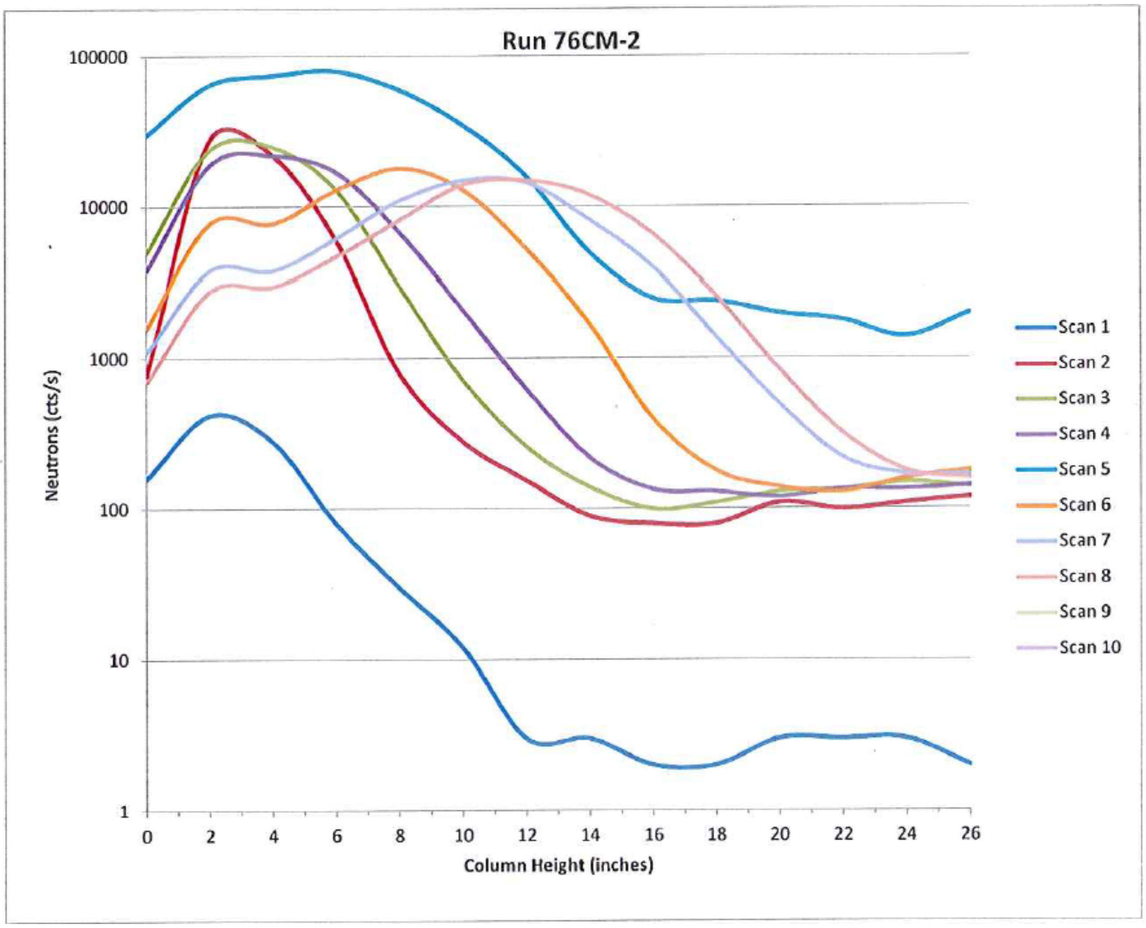


Figure 7. Neutron probe scans of the exchange column tracking material migration in 76CM-2.

On 4/9/15–4/10/15, T-635 was flushed and F-640 was back flushed to T-65 with HCl and water, sparged and sampled T-65 (heel check; post-run feed residue) as CMCF-536. Per NNFD-7920-OP-172, acidified T-61, sparged and sampled as CMCR-584; acidified T-630, sparged and sampled as CMCR-585; acidified T-634, sparged and sampled as CMAP-304; sparged and sampled T-632 as CMBP-153; sparged and sampled T-72 as CMCR-586, which was boiled down for freeboard to collect additional rework solution during 76CM-3. Due to unexpected results, re-sparged and resampled T-61 and T-630 as CMCR-587 and CMCR-588, respectively.

It appeared that plutonium contamination entered into products collected during 76CM-2 (see results for CMAP-304 and CMBP-153 in Table 5)—adding ~3 g of Pu to the ~1 g present in the feed. (It is possible that the contamination occurred during 76CM-1, but no Pu analysis was performed for products collected during that run.) Note, ^{238}Pu production operations were being conducted in the cubicles at the time leading up to 76CM-1.

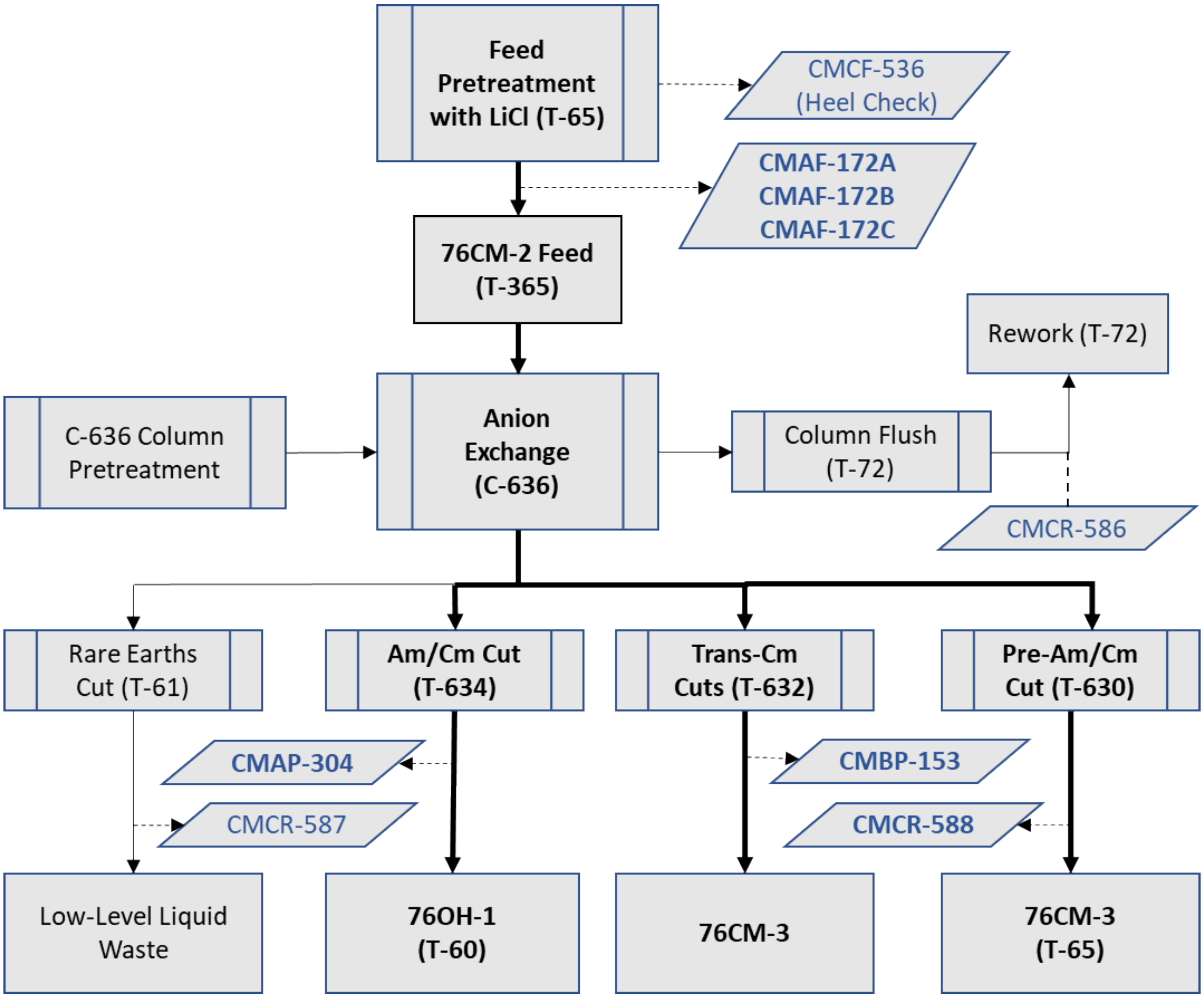


Figure 8. Flow sheet of anion exchange group separations of actinides and lanthanides—76CM-2.

Table 5. Sample results* for 76CM-2

Sample Code	Date	Tank No.	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk (mg)	^{144}Ce (Ci)	^{154}Eu (Ci)
CMAF-172A	3/20/15	T-65	1.176	2.954	0.640	44.043	10.394	97.622		132.3	1.441
CMAF-172B	3/20/15	T-65		3.494	0.757	42.757	10.091	91.104		134.6	1.535
CMAF-172C	3/20/15	T-65	0.882	3.049	0.661	40.956	9.666	91.115		125.2	1.394
CMAP-304	4/10/15	T-634	2.079	1.459	0.316	20.012	4.723	0.179	0.149	0.789	0.025
CMBP-153	4/10/15	T-632	1.904	1.344	0.291	9.330	2.202	97.196		1.831	0.225
CMCR-586	4/10/15	T-72	0.128	0.008	0.002	0.062	0.015	0.872		0.077	0.005
CMCR-587	4/10/15	T-61	0.012	0.001	0.000	0.010	0.002			138.7	1.651
CMCR-588	4/10/15	T-630	0.009	0.014	0.003	0.282	0.067			0.246	0.012
CMCF-536	4/10/15	T-65	0.004	0.002	0.000	0.029	0.007	0.058		0.069	0.001

* Values are *not* included for outlier results or results below limits of detection. Zero values indicate rounding *down*, nominally from the next digit (i.e., 0.0001 < value < 0.0005 would be rounded down to 0.000).

As part of 76CM-2 run follow-up on 4/10/15, resin was removed from C-636 to poly bottles, and the column was flushed with demineralized water; effluents were transferred from 76CM-2 pre-Cm cuts from T-632 to T-65 for the 76CM-3 recycle run. This was sequentially followed with 2 L of 4 *M* HCl, 7 L of 0.5 *M* HCl, and 2 × 1 L of water, sparging each flush with a 5 minute sparge via the instrument purge line. T-65 was evaporated to provide freeboard for adding Cm product from T-630 to T-65, which was completed 4/11/15. The final T-65 volume was 13.7 L. Effluent from the hot feed in T-61 (CMCR-584) was sent to T-79 via T-607, flushed with 3 × 4.5 L water (sparging 10 minutes between each flush), and transferred to F-115.

4.3 76CM-3—LICL ANION EXCHANGE—3

4/10/15–4/15/15

The third AIX run was a recycle run of combined pre-Am/Cm cuts (in T-630) and transcurium cuts (in T-632) from 76CM-2 (which included the cuts collected previously from 76CM-1). Set up and system checks began on 4/10/15 with activities paralleling those for 76CM-1 (reagent and elutriant makeups, valve and sample block checks, leak checks and repairs as needed, system flushes, etc.). The alpha probe read low (63,000 counts per second [cps]) on the conveyor lid; the Mylar seal also looked bad so on 4/13/15 it was swapped for a new one that read OK at >100,000 cps. The second batch of HCl-treated, Dowex 1X8, 200-400 mesh “B” Fraction Resin, was added to C-636 per NNFD-7920-OP-166 and pretreated on 4/14/15 per NNFD-7920-OP-171. Feed was adjusted beginning 4/12/15 per NNFD-7920-OP-170 in T-65 by boiling. Prior to T-65 reaching 120°C on 4/13/15, a sample was requested by management; so, cooled T-65 to <50°C, sampled as CMAF-173 at a volume of 4.23 L indicating a content of 35.11 mol LiCl. Proceeded to heat T-65 to a final temperature of 142.5°C (2.50 L after cooldown). T-65 was then adjusted to ~1M HCl by adding 245 mL of 12M HCl (flushed in with 200mL of stock LiCl) and heating T-65 back up to 120°C. T-65 was held at 120°C for 10 minutes then cooled to <50°C. On 4/14/15, T-65 contents were transferred to T-635 followed by 3 × 0.5 L of synthetic feed with 5 minutes of sparging between each flush.

On 4/14/15 per NNFD-7920-OP-172, hot feed from T-635 was loaded onto column C-636 at 2 L/h. with effluent directed to T-61 (Figure 10). A small accumulation of a leak was noticed on the cubicle floor under the jumpers. The leak was not found, *per se*, but the off-going shift had repaired a leak at HCV-C636. When the hot feed level in T-635 reached <0.2 L, 3 × 0.5 L synthetic feed from T-65 was added to T-635 while pumping continued through C-636 to T-61. The column was eluted with the following:

1. RE elutriant—3 × 0.5 L, 1 × 1 L, and part of 1 × 0.5 L to T-635→C636→T-61, then switched effluent collection to T-630 to finish RE elutriant.

2. Cm elutriant (pre-curium cut)—1 × 0.5 L and part of a second 1 × 0.5 L to T-635→C636→T-630, then switched effluent collection to T-631 to finish the second 0.5 L Cm elutriant for the curium cut.

3. Cm elutriant (curium cut)—1 × 1 L, 1 × 0.5 L, 2 × 0.25 L Am/Cm elutriant, and 1 × 0.5 L of Cf elutriant to T-635→C636→T-631.

4. Cf elutriant—Switched effluent collection to T-632 and added 2 × 0.5 L and 1 × 1 L of Cf Elutriant to T-635→C636→T-632.

5. Column flush—Switched to effluent collection T-72 and added 2 × 0.5 L, and 1 × 3 L of column flush to T-635→C636→T-72.

During the column run, the column was scanned with a neutron probe at periodic intervals to track the ^{252}Cf as it migrated down the length of the column, beginning with scan 1. Data for these scans are shown in Figure 9 with the neutron count rate shown as a function of column height. The graph was set up to accommodate 10 scans, although only 8 (scans 1–8) were needed to complete the operation.

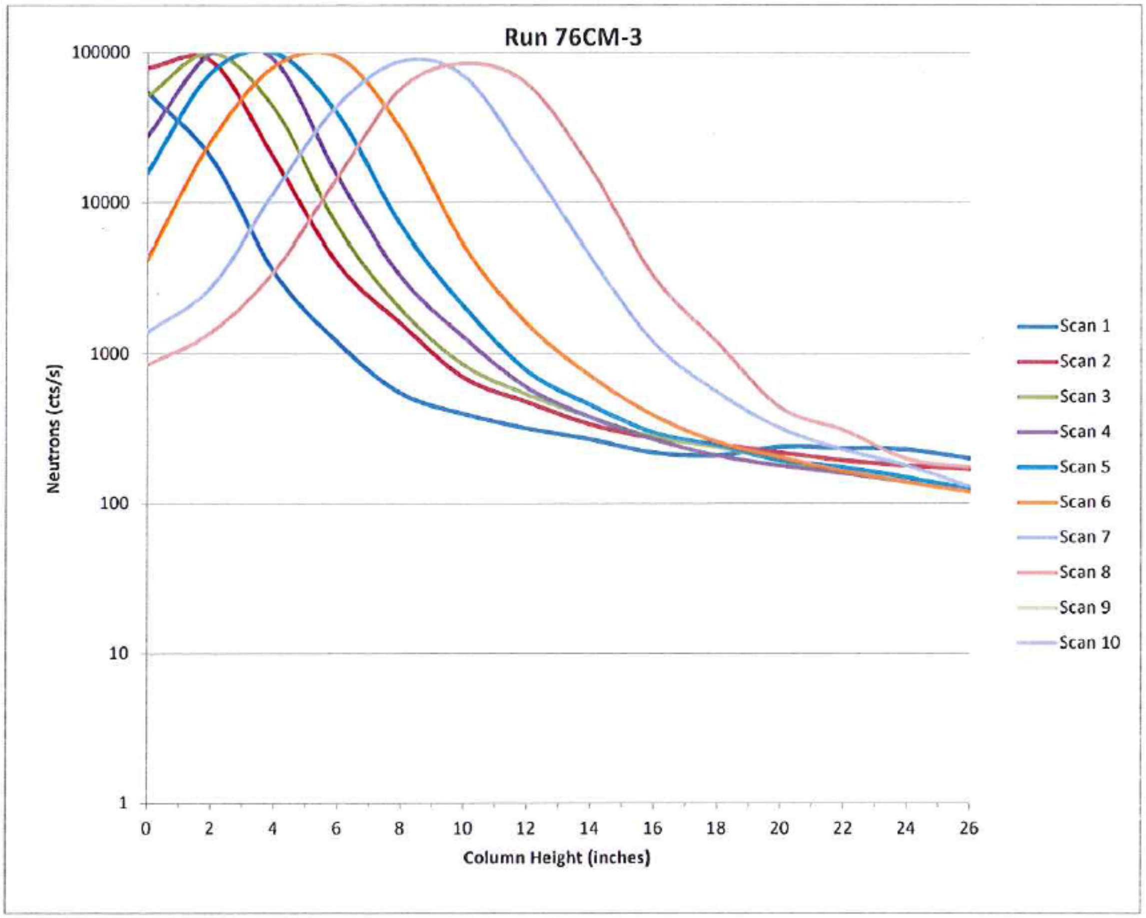


Figure 9. Neutron probe scans of the exchange column tracking material migration in 76CM-3.

Per NNFD-7920-OP-172 on 4/14/15, T-61 was acidified, sparged, and sampled as CMCR-589. T-630 was acidified, sparged, and sampled as CMCR-590. T-631 was acidified, sparged, and sampled as CMAP-305. T-632 was sparged and sampled as CMBP-154 T-72 was sparged and sampled as CMCR-591. In accordance NNFD-7920-OP-170, F-640 was back flushed to T-65 with 3 × 1 L of 0.8 *M* HCl and 1 × 1 L of water. Then T-635 was flushed to T-65 with 2 × 2 L of 0.8 *M* HCl and 1 × 1 L of water. Another 6 L of 1 *M* HCl was added to T-65 followed by 2 × 0.5 L of water, sparged >30 minutes, and sampled as CMCF-537.

Plutonium contamination noted in describing 76CM-2 appeared to be exacerbated by its increasing to more than 9 g (see results for CMAP-305 in Table 6). Tracking the plutonium will continue in 76OH-1, the destination of material represented by these results.

Curium products were consolidated into T-60 on 5/12/15 and 5/13/15 and were sampled in preparation for the 76OH runs (reported below). As part of consolidation, T-631 (a pre-curium cut in 76CM-1) seemed to have some restriction preventing transfer; this was dislodged and then flushed with HCl and water to T-60 for later processing, as part of a 76OH run.

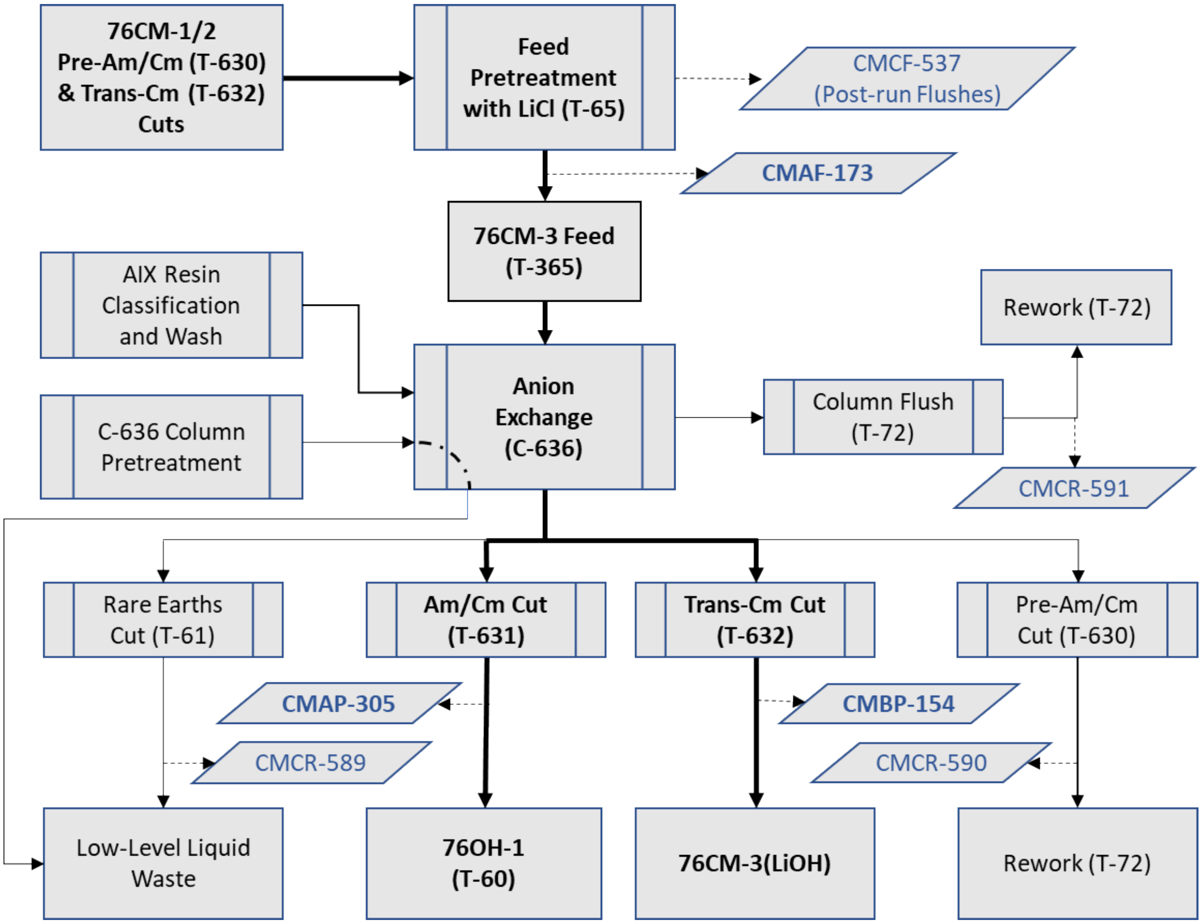


Figure 10. Flow sheet of anion exchange group separations of actinides and lanthanides—76CM-3.

Table 6. Sample results* for 76CM-3

Sample Code	Date	Tank No.	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk (mg)	^{144}Ce (Ci)	^{154}Eu (Ci)
CMAF-173	4/14/15	T-65	0.080	0.393	0.085	3.081	0.727	87.50		0.598	
CMAP-305	4/14/15	T-631	9.178	0.518	0.112	3.811	0.899	0.954	0.223	0.728	0.044
CMBP-154	4/14/15	T-632	0.123	0.068	0.015	0.161	0.038	77.31	17.369		
CMCR-589	4/15/15	T-61				0.000				0.896	0.021
CMCR-590	4/15/15	T-630		0.001	0.000	0.025	0.006			0.002	
CMCR-591	4/15/15	T-72	0.111	0.031	0.007	0.063	0.015	2.138		0.087	
CMCF-537	4/15/15	T-65	0.004	0.132	0.000	0.010	0.002	0.191		0.008	

* Values are *not* included for outlier results or results below limits of detection. Zero values indicate rounding *down*, nominally from the next digit (i.e., 0.0001 < value < 0.0005 would be rounded down to 0.000).

4.4 76CM-3(LIOH)—CONVERSION OF TRANSCURIUM ACTINIDES TO NITRATE FORM BY HYDROXIDE PRECIPITATION

4/15/15–6/8/15

As part of concluding 76CM-3, transcurium products (Bk, Cf, Es, and/or Fm) were converted to nitrate form by hydroxide precipitation, filtration, and redissolution in HNO_3 (Figure 11). This process is designated herein as 76CM-3(LiOH). Precipitation using 5 *M* LiOH was performed in the transcurium product receiving tank, T-632, and the precipitate was collected on a glass frit filter. The precipitate then was dissolved with nitric acid and collected in a glass bottle, which was transferred to another cubicle for CX runs. The goal was to remove any residual LiCl and convert the transcurium product from a chloride (Cl^-) solution to a nitrate (NO_3^-) solution suitable as feed to an AHIB CX column run. A similar strategy of precipitation followed by dissolution will be used to remove LiCl residues from 76CM Am/Cm cuts during the 76OH runs later in the campaign.

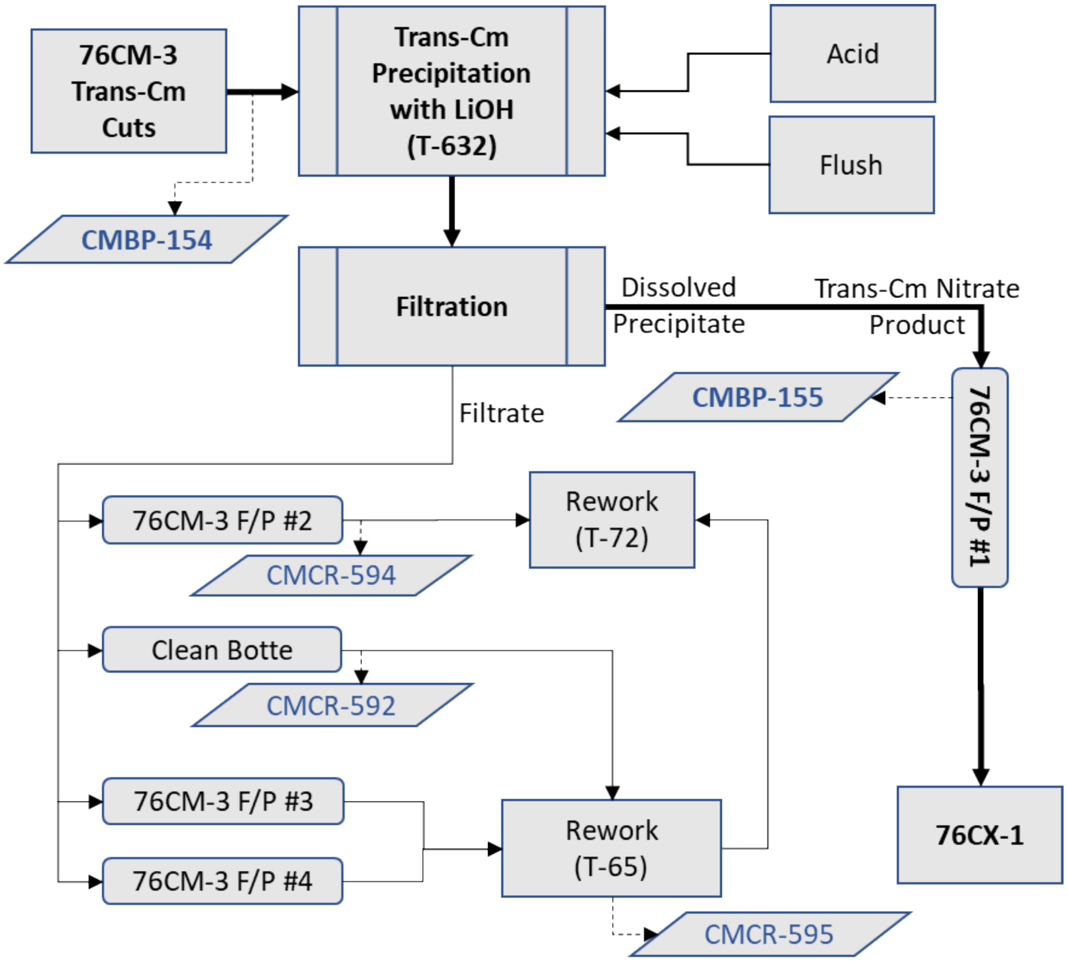


Figure 11. Flow sheet for converting transcurium actinides from chloride to nitrate form—76CM-3(LiOH).

Set up and system checks began on 4/15/15 per NNFD-7920-OP-185. During preparations, the manual diversion valve (MDV-637) stem sheared requiring valve replacement. A replacement valve was installed in a manner such that the position indicator was misaligned; this required consulting with the REDC technical assistant, so the valve could be operated in contradiction to the operator aid.

Filtration of precipitate began 4/21/15; solution transfer was “extremely fast through the filter and filtrate had an orange-ish hue.” Filtrate bottle indicated ~17,000 cps above background prompting its recycle through the filter and collection (with a bottle flush) into 76CM-3 Clean Bottle. This reduced the reading to ~1,300 cps. The second filtration batch had similar results as the first: “an orange-ish hue and was 18,000 cps above background” and was recycled and then transferred (700 mL) into a poly bottle labeled “76CM-3 Filtrate #2.” The product solution continued filtering with the third batch, reading to 2,700 cps, which was transferred (900 mL) to a poly bottle labeled “76CM-3 Filtrate #3.” A fourth product filtration resulted in filtrate reading 1,000 cps and was transferred (800 mL) to a poly bottle labeled “76CM-3 Filtrate #4.” Repeat measurements of the bottles using the portable neutron cabinet were lower, thereby affirming (and providing better values than) those taken in a higher background. The highest-dose bottle—deemed to be 76CM-3 Clean Bottle—was sampled as CMCR-592 to bound activity in the filtrate.

Product recovery began on 4/22/15 with two 200 mL water flushes of T-632 being routed through the intermediate bottle and collected in 76CM-3 Product #1. A second 50 mL water flush directly into the intermediate bottle showed a change of color (a sign of peptizing), so the transfer was diverted to 76CM-3 Filtrate #2, which was sampled for activity as CMCR-593. Neutron monitoring indicated a dose rate ~1,300 cps above background. Recovering the precipitate cake began by monitoring the glass frit filter (>100,000 cps) and then slowly adding 14 mL of concentrated nitric acid (and, after a 15-minute digestion, 10 mL of water) to dissolve the precipitate. The dissolved precipitate was transferred, followed by two dilute nitric acid flushes of the precipitation vessel (T-632), a second precipitate dissolution (with 8 *M* HNO_3), and a water flush of T-632 to 76CM-3 Product #1, which was sampled as CMBP-155. The final volume was 750 mL, and the highest neutron dose rate was 740 cps. The filter indicated 280 cps above background after re-dissolutions.

As a follow-up to the 76CM runs, filtrates and system flushes (from intermediate bottle) were transferred on 4/23/15 to T-65 followed by water flushes. On 4/29/15, tank T-65 was sparged and sampled as CMCR-595. 76CM-3 Filtrate #2 was treated with 100 mL of concentrated HCl, shaken to dissolve solids, and then resampled as CMCR-594. The heel in T-43 was checked on 5/26/15 after treating with HCl, heating, sparging, and sampling as MSA-760. T-43 was transferred to F-115 on 6/2/15.

The anion exchange resin was removed from the LiCl column (C-636) on 6/3/15 per NNFD-7920-OP-165.

Table 7. Sample results* for 76CM-3(LiOH)

Sample Code	Date	Tank No.†	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk (mg)	^{253}Es (μg)
CMBP-154	4/15/15	T-632	0.123	0.068	0.015	0.161	0.038	77.31	17.37	20.71
CMCR-592	4/21/15	F/P-Clean	0.002			0.000		0.120		0.028
CMBP-155	4/23/15	F/P#1	0.042	0.002	0.001	0.033	0.008	84.74		16.79
CMCR-594	4/23/15	F/P#2	0.002	0.000		0.001	0.000	2.822		0.369
CMCR-595	4/29/15	T-65	0.012	0.001	0.000	0.016	0.004	1.989		0.264

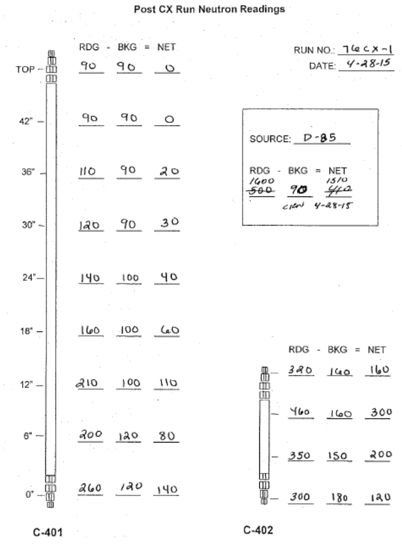
* Values are *not* included for outlier results or results below limits of detection. Zero values indicate rounding *down*, nominally from the next digit (i.e., 0.0001 < value < 0.0005 would be rounded down to 0.000).

† F/P# Tank No. represents 76CM-3 Filtrate/Product bottle # and appears with a single-digit number or a “Clean” Bottle indication.

5. 76CX—AHIB CATION EXCHANGE FOR TRANSCURIUM SEPARATION

Transcurium elements purified during the 76CM-3(LiOH) run were separated in two cation exchange column runs, 76CX-1 (~60% of the 76CM-3[LiOH] product) and 76CX-2 (the balance of 76CM-3[LiOH] product). This type of process is sometimes referred to as high pressure ion exchange due to the small granule size (30–45 µm) of the Dowex 50W-X8 resin, which imparts a substantial pressure drop as the feed is pushed through the resin bed at high flow rates. The small-granule resin facilitates resolution of the different actinides, and the high flow rate reduces retention time, thereby mitigating radiation damage and suppressing outgassing of radiolytic by-products. Resolving the actinides is particularly important to remove berkelium from the californium since ^{249}Bk decays ($t_{½}$ = 330 days) to ^{249}Cf, which dilutes the isotopic purity of ^{252}Cf and its value as a powerful neutron source.

First, the actinides were loaded onto the resin in a short loading column (C-402; with 25 mL of resin) with preference given to trivalent actinides; unloaded resin sites are converted to ammonium form by treatment with 0.3 L of 0.3 *M* NH_4NO_3. (Establishing the ammonium form of the resin is essential to maintaining complexing conditions during subsequent, chromatographic elution.) The loading column effluent then was routed to a longer column (C-401), also filled with Dowex 50W-X8 resin (145 mL). Elemental separation was accomplished during elution by pumping a complexing agent, ammonium alpha-hydroxyisobutyrate (AHIB), at varying pHs through the loading column and then through the longer elution (or separation) column, which was heated to about 70°C. Complexing strengths are different for each of the elements so that they moved through the system at different rates, forming distinct bands as they progressed through the longer elution column [4]. Band separations facilitated their individual identification—using both the neutron probe (indicating the hourly position of the ^{252}Cf band on both columns [see example of post-run readings in Figure 12] graphed as a function of column height) and an inline alpha detector at the column effluent (to monitor elution progress [see example graph for CX-1 effluent in Figure 13])—and allowed each to be collected separately (Es, Bk, and Cf) in small, numbered bottles referred to as cut bottles (CBs or cuts). (Einsteinium and berkelium are useful in super heavy element research.) The effluents were acidified, sampled and analyzed individually to confirm separation and facilitate disposition as products, rework or waste. Fresh Dowex 50W-X8 resin was used in the loading column (C-402) for each CX run; resin filling the elution column (C-401) was used for both CX runs.



Post CX Run Neutron Readings

RUN NO.: 76CX-1
DATE: 4-28-15

SOURCE: D-85

RDG	BKG	NET
~~500~~ 1600	90	~~410~~ 1510

CRW 4-28-15

C-401

	RDG	BKG	NET
TOP	90	90	0
42"	90	90	0
36"	110	90	20
30"	120	90	30
24"	140	100	40
18"	160	100	60
12"	210	100	110
6"	200	120	80
0"	260	120	140

C-402

RDG	BKG	NET
320	160	160
460	160	300
350	150	200
300	180	120

Figure 12. Typical neutron probe readings of loading column (C-402, on right) and elution column (C-401, on left) at <u>end</u> of a CX run elution. Sets of readings taken during a CX run are not retained.

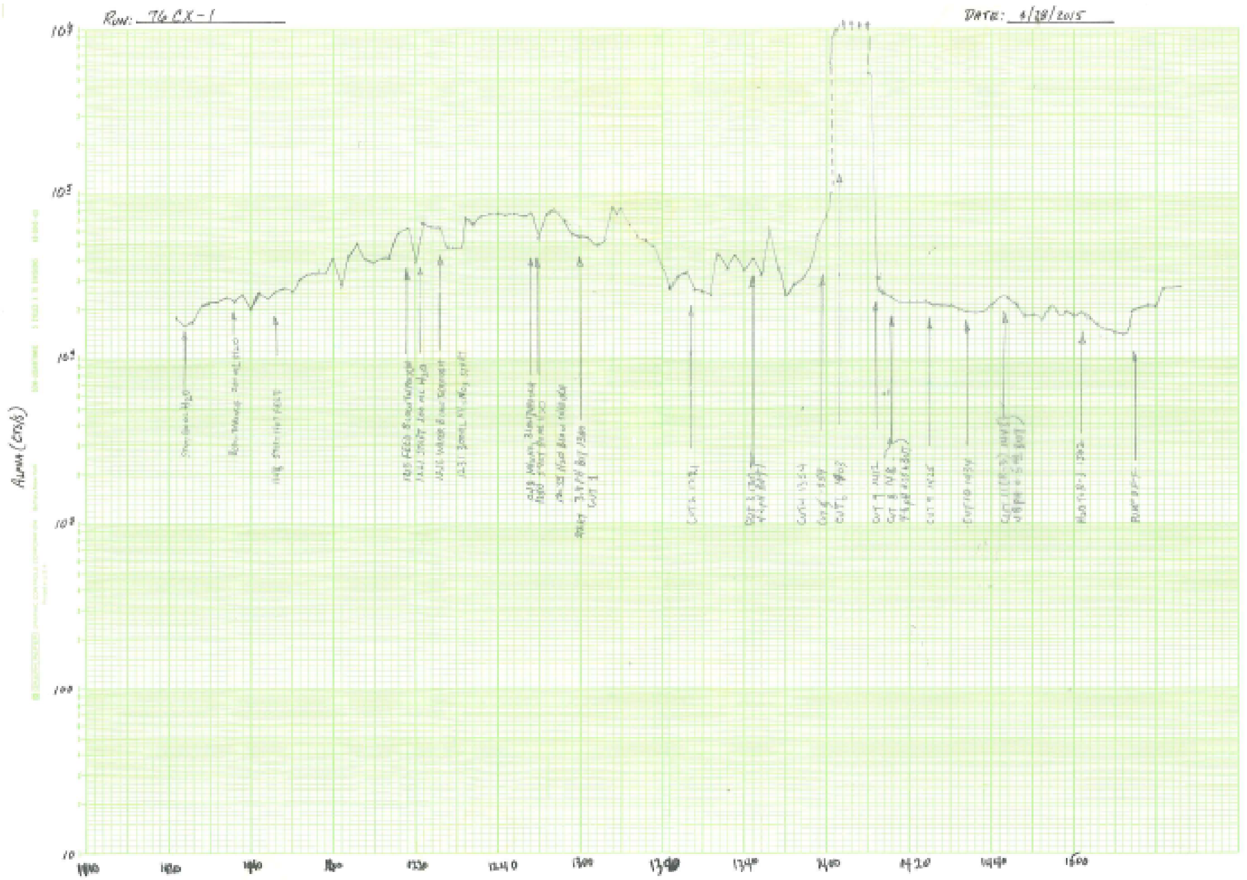


Figure 13. Typical alpha detector readings of C-401 column effluent during CX run elution.

5.1 76CX-1—AHIB CATION EXCHANGE FOR TRANSCURIUM SEPARATION

4/23/15–4/30/15

Set up and system checks began on 4/23/15 for reagent solutions (including AHIB) per NNFD-7920-OP-178 and typical pre-run maintenance on the right rack in Cub-4 (RR4) and pre-run preps per NNFD-7920-OP-177. The pre-run checks included staging bottled eluting reagents and empty bottles for recycle and product cuts in Cub-4. Fresh Dowex 50W-X8 resin was prepared and added to columns C-401 and C-402 per NNFD-7920-OP-175 and NNFD-7920-OP-176, respectively. Before loading began, the pump and column system were tested with water; the effluent was collected in a 1 L poly bottle labeled “R-2” and sampled later as CXLR-512.

Actinides from 60% of the feed were loaded onto the resin in column C-402 on 4/28/15 per NNFD-7920-OP-179, and they were eluted from C-402 and through C-401 per NNFD-7920-OP-180 the same day. Raffinate from column loading was collected for recycle in an empty poly bottle labeled “R-1” and sampled as CXLR-511. Elution involved the following reagents (in order of how they affect separation of actinides), after which all cuts were acidified and sampled NNFD-7920-OP-181 as indicated:

1. 0.25 *M* AHIB with pH 3.9 into CB #1 (CXER-405) and CB #2 (CXER-406)
2. 0.25 *M* AHIB with pH 4.2 into CB #3 (CXER-407), CB #4 (CXES-305), CB #5 (CXES-306), CB #6 (CXCF-803) and CB #7 (CXCF-804)

3. 0.25 *M* AHIB with pH 4.6 into CB #8 (CXBK-312), CB #9 (CXBK-313) and CB #10 (CXBK-314)

The column was flushed with 500 mL of 0.5 *M* AHIB solution with pH = 4.8, which was collected in a 1 L poly bottle labeled "R-3" (CXCM-243).

Cuts #3, #4, #5, and #7 were recycled to join the balance of feed for 76CX-2, but cut #6 was retained for a 76CF run. Cuts #8 and #9 were saved to be consolidated later with comparable 76CX-2 cuts for Bk purification. Raffinate solutions from bottles R-1, R-2, and R-3 and cuts #1, #2, and #10 were transferred to T-66 on 4/29/15 for rework.

IX resin was removed from column C-402 on 4/29/15 per NNFD-7920-OP-173 and collected in a cut-off poly bottle. A neutron scan of the bottle indicated 120 cps above background, which is about ⅓ higher than a scan of Cf packages (90 cps) performed as an operability test of the neutron probe before 76CX-1 (Figure 14).

Neutron scans of C-401 and C-402 were performed. These scans indicated maximum readings of 90 cps for C-401 and 150 cps for C-402 (empty of resin).

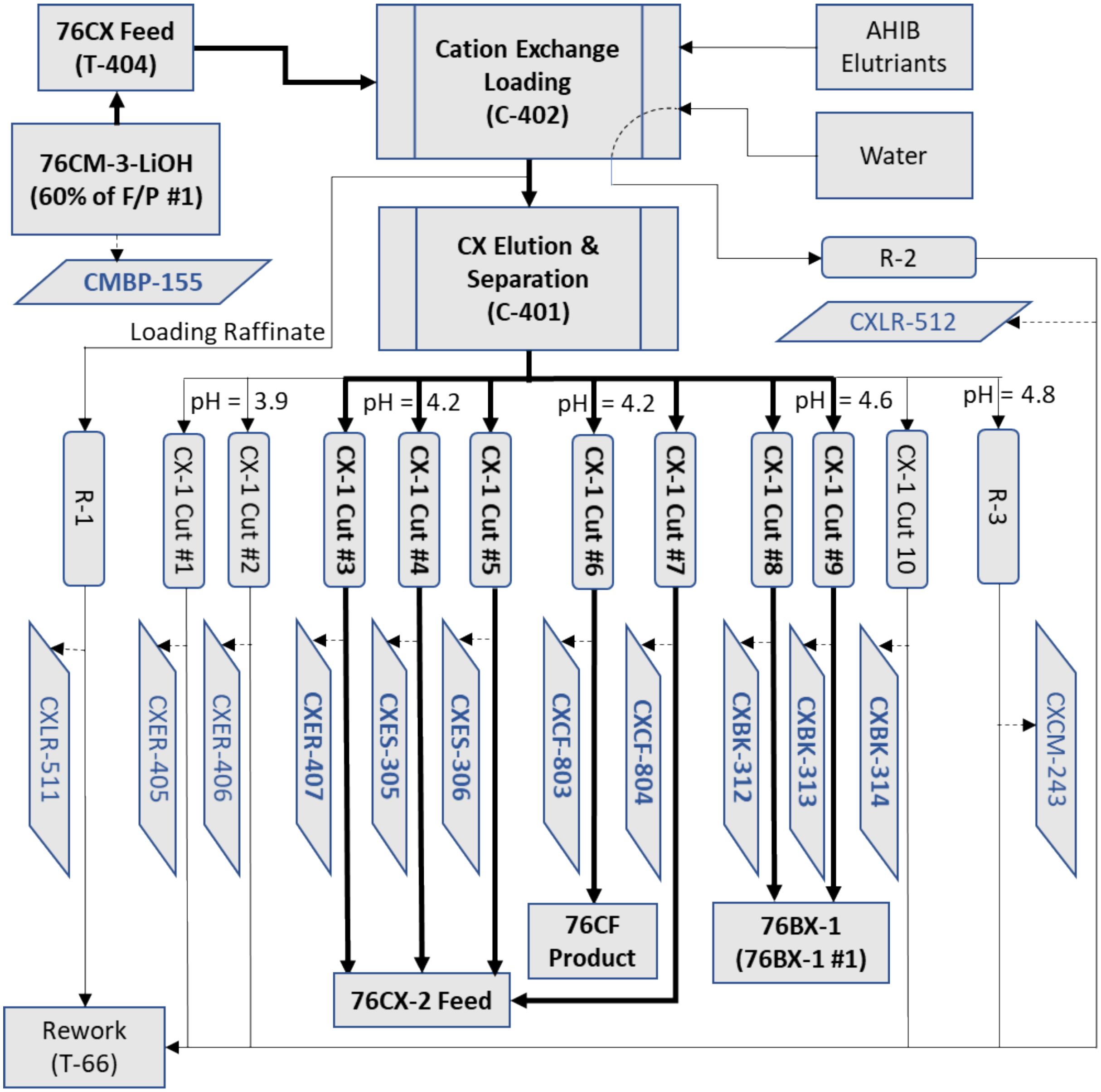


Figure 14. Flow sheet for AHIB cation exchange separation of transcurium elements—76CX-1.

Table 8. Sample results* for 76CX-1 transcurium elements separation

Sample Code	Date	Tank No.†	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk (mg)	^{253}Es (µg)
CMBP-155‡	4/23/15	F/P#1	0.042	0.002	0.001	0.033	0.008	84.74	17.37	16.79
CXLR-511	4/29/15	R-1						0.000		
CXLR-512	4/29/15	R-2						0.000		
CXER-405	4/29/15	CB#1						0.002		
CXER-406	4/29/15	CB#2	0.000					0.002		0.000
CXER-407	4/29/15	CB#3	0.000					0.002		0.001
CXES-305	4/29/15	CB#4				0.000		0.018		40.80
CXES-306	4/29/15	CB#5	0.954	0.001	0.000	0.011	0.003	6.160		0.538

Sample Code	Date	Tank No.†	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk (mg)	^{253}Es (µg)
CXCF-803	4/29/15	CB#6		0.001	0.000	0.011	0.002	45.32		
CXCF-804	4/29/15	CB#7		0.000		0.002	0.001	27.69	0.039	
CXBK-312	4/29/15	CB#8	0.001			0.000	0.000	0.267	9.313	0.013
CXBK-313	4/29/15	CB#9	0.014			0.000		0.076	1.017	0.008
CXBK-314	4/29/15	CB#10	0.016			0.005	0.001	0.032	0.008	
CXCM-243	4/29/15	R-3	0.004	0.005	0.001	0.015	0.004	0.092		0.019

* Values are *not* included for outlier results or results below limits of detection. Zero values indicate rounding down, *nominally from the next digit* (i.e., 0.0001 < value < 0.0005 would be rounded down to 0.000).

† F/P# Tank No. represents 76CM-3 Filtrate/Product bottle # and appears with a single-digit number or a “Clean” Bottle indication.

‡ No ^{249}Bk value is available for CMBP-155; value indicated is included from CMBP-154 (sampled 4/15/15).

5.2 76CX-2—AHIB CATION EXHANGE FOR TRANSCURIUM SEPARATION

4/29/15–5/27/15

76CX-2 began on 4/29/15 recharging with resin into C-402. Cuts from 76CX-1 CBs #3 (225 mL), #4 (80 mL), #5 (75 mL), and #7 (40 mL) were recycled to the 76CX-2 feed bottle—joining the balance of feed (40%, left after 76CX-1) was sampled only for an acid check. Each of the cut bottles (CBs) was flushed with water and followed by concentrated HNO_3 composited into the feed bottle. Feed was sampled only to confirm the free acid content. Actinides in the feed were loaded onto column C-402 per NNFD-7920-OP-179 and eluted from C-402 and through C-401 (Figure 15) per NNFD-7920-OP-180 the same day. Raffinate from loading was collected for recycle in an empty poly bottle labeled “R-1” and sampled as CXLR-513; however, this sample was misplaced, requiring R-1 to be resampled as CXLR-513. Elution involved the following reagents (in order of how they affect separation of actinides) residing on the elution column:

1. 0.25 *M* AHIB with pH 3.9 into CB #1
2. 0.25 *M* AHIB with pH 4.2 into CB #2, CB #3, CB #4, CB #5, CB #6, CB #7, CB #8, and CB #9
3. 0.25 *M* AHIB with pH 4.6 into and CB #10, CB #11, and CB #12

All cuts (volumes in parentheses) were acidified and sampled as indicated on 4/30/15 per NNFD-7920-OP-181:

1. Bottle R-1—CXLR-513 (1,000 mL)
2. Bottle R-2—CXLR-514 (600 mL)
3. Bottle R-3—CXCM-244 (910 mL)
4. CB #1—CXER-408 (230 mL)
5. CB #2—CXER-409 (225 mL)
6. CB #3—CXFM-120 (135 mL)
7. CB #4—CXES-307 (60 mL)
8. CB #5—CXES-308 (40 mL)
9. CB #6—CXES-309 (45 mL)
10. CB #7—CXCF-805 (155 mL)
11. CB #8—CXCF-806 (55 mL)
12. CB #9—CXCF-807 (40 mL)

13. CB #10—CXBK-315 (230 mL)
14. CB #11—CXBK-316 (235 mL)
15. CB #12—CXBK-317 (230 mL)

Used resin was removed from column C-402 on 5/1/15 and collected in a cut-off poly bottle. Resin was colored orange/red and indicated 20 neutron counts per second above background. Raffinate solutions from bottles R-1, R-2, and R-3 and cuts #1, #2, and #3 were transferred to T-66 on 5/1/15 for rework. Cut #12 was transferred to T-66 after some confusion was cleared up regarding its labeling: cut bottle #12 was labeled and introduced to the cubicle as part of 76CX-1, but it was not used until 76CX-2. (No cut #12 was collected as part of 76CX-1.)

Californium cuts in 76CX-2 #7and #8 were consolidated with 76CX-1 #6 into the "76CF Product" Kimax bottle on 5/1/15 and were joined later by 76CX-2 #6 and #9 on 5/5/15; each CB was flushed with 2 × 10 mL of water and followed by 1 mL of concentrated HNO_3 and another 20 mL water flush to "76CF Product"—multiple times, as needed, based on neutron readings of the CBs. The "76CF Product" Kimax bottle was sparged and sampled on 5/5/15 as CXCF-808 for run 76CF-1. An isotopic analysis of the Cf in the product was performed using inductively coupled plasma mass spectrometry (Table 9). Gamma spectrometry identified only ^{237}Np (3 g) at microgram or greater levels for nuclides present above the limits of detection.

Table 9. Isotopic results (atom %, as of 5/20/15) for Cf in sample CXCF-808 from CF product bottle—76CX-2

Isotope	Result	2σ Error
^{249}Cf	13.34	0.13
^{250}Cf	18.33	0.18
^{251}Cf	5.55	0.11
^{252}Cf	62.78	0.63

Berkelium cuts in CX-2 #10 and CX-2 #11 were consolidated with CX-1 cuts #8 and #9 in a 2 L poly bottle labeled "76BX-1 #1"; the CBs were flushed as described for the Cf cuts.

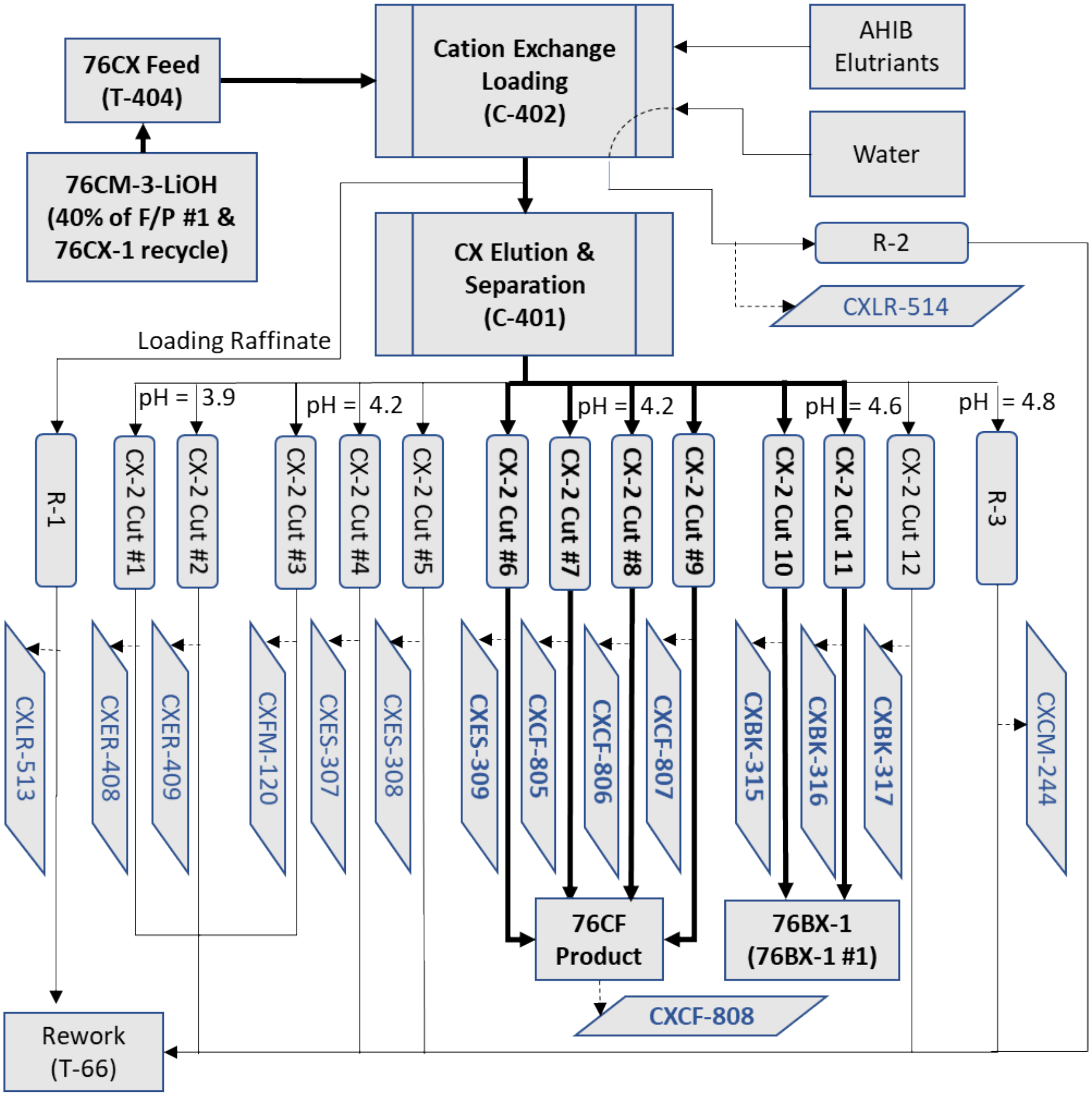


Figure 15. Flow sheet for AHIB cation exchange column separation of transcurium elements—76CX-2.

Table 10. Sample results* for 76CX-2 transcurium elements separation

Sample Code	Date	Tank No.†	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk (mg)	^{253}Es (µg)
CXLR-513	4/30/15	R-1						0.001		0.000
CXLR-514	4/30/15	R-2						0.000		
CXER-408	4/30/15	CB#1	0.000					0.008		0.001
CXER-409	4/30/15	CB#2	0.000					0.008		0.002
CXFM-120	4/30/15	CB#3	0.000					0.002		0.014
CXES-307	4/30/15	CB#4	0.000					0.001		1.868
CXES-308	4/30/15	CB#5	0.000					0.011		5.325
CXES-309	4/30/15	CB#6						0.672		1.635
CXCF-805	4/30/15	CB#7		0.000	0.000	0.007	0.002	27.78		
CXCF-806	4/30/15	CB#8	0.001			0.000		2.793	0.006	
CXCF-807	4/30/15	CB#9	0.002					0.099	0.001	
CXBK-315	4/30/15	CB#10	0.008			0.000	0.000	0.065	4.304	0.004
CXBK-316	4/30/15	CB#11	0.004					0.029	2.119	0.007
CXBK-317	4/30/15	CB#12	0.000			0.000	0.000	0.021	0.009	0.005
CXCM-244	4/30/15	R-3	0.001	0.002	0.000	0.012	0.003	0.039		
CXCF-808‡	5/5/15	76Cf Pr.		0.001	0.000	0.020	0.005	84.12		2.054

* Values are *not* included for outlier results or results below limits of detection. Zero values indicate rounding down, *nominally from the next digit* (i.e., 0.0001 < value < 0.0005 would be rounded down to 0.000).

† Tank No. "Kimax" is a 1 L glass bottle in which the balance of feed and the transcurium cuts from 76CX-1 were consolidated for 76CX-2.
"76Cf Pr." (also a Kimax bottle) represents the Cf product cuts consolidated from 76CX-1 and 76CX-2.

‡ 76CF Product Bottle (sampled as CXCF-808) was a consolidation of 76CX-1 Cut #6 and 76CX-2 Cuts 6–9.

6. 76BX—BERKELIUM PURIFICATION USING BATCH SOLVENT EXTRACTION

Berkelium fractions from 76CX runs were further purified from Cf using Berkex batch solvent extraction followed by further refinement for dispensing as ^{249}Bk product. To prepare the feed solution for Berkex, the berkelium fractions from the CX runs (sampled as CXBK) were consolidated, and the berkelium was concentrated and purified from AHIB solution using cation exchange in Cub-4. The consolidated Bk solution was pressured through the small loading column (C-402) to load the Bk on the Dowex 50W-X8 resin and then eluted with 500 mL of 8 M HNO_3, the desired feed solution for Berkex.

After about a 1 week digestion period, Bk feed solution was treated with 0.30 M $NaBrO_3$ to oxidize Bk(III) to Bk(IV). The Berkex solvent extraction process was conducted in tank T-411 with each batch's contact facilitated by a 30 minute agitation followed by 10 minutes of settling and then slow and careful draining via a "PS dip line" to decant each heavy aqueous phase from the lighter organic phase. The extractant was 0.5 M HDEHP in dodecane, which was scrubbed with ~7.0 M HNO_3–0.3 M $NaBrO_3$ to drive entrained Cf into the aqueous phase; finally, the purified Bk was stripped from the extractant with 8.0 M HNO_3–1 M H_2O_2. Sufficient decontamination was achieved in one cycle to allow Bk product to be transferred to a laboratory glovebox for final purification and packaging for shipment.

6.1 76BX-1—BERKELIUM PURIFICATION USING BATCH SOLVENT EXTRACTION

4/30/15–5/12/15

The following Bk cuts from the 76CX runs were consolidated on 5/4/15 for further purification:

- Cut #8 from CX-1 (235 mL)
- Cut #9 from CX-1 (235 mL)
- Cut #10 from CX-2 (235 mL)
- Cut #11 from CX-2 (220 mL)

Each bottle was flushed with 10 mL of water and followed with 1 mL of concentrated HNO_3 routed to the feed bottle water, labeled "76BX-1 #1." The feed was sparged gently and sampled as BXBK-079. A new neutron probe was installed, and the alpha probe was tested (OK). The neutron probe read 2,150 cps above background for the D-85 nut package source. The Berkex feed was adjusted, and the berkelium was purified per NNFD-7920-OP-187, followed by 2 × 200 mL of water (via the 76BX-1 #1 bottle) to displace interstitial AHIB reagent. Raffinate was collected in poly bottles R-1 and R-2, except for the second water wash, which was collected in R-3.

Berkelium then was eluted with 2 × 200 mL of 8 M HNO_3 (via the 76BX-1 feed bottle [#1]) and 1 × 100 mL 8 M HNO_3, and eluates were collected in a bottle labeled "76BX-1 FEED/RAFFINATE." The feed bottle (#1) then was rinsed with water and passed through C-402 column, and the raffinate was collected in a bottle labeled "Circle 6." Next, 100 mL concentrated HNO_3 was added to FEED/RAFFINATE, sparged, and sampled as BXBK-080. R-1, R-2, R-3, and Circle 6 were sampled after shaking, respectively, as BXRF-207, BXRF-208, BXRF-209, and BXRF-210. These solutions were transferred to T-45 for rework at a later date (5/15/15). C-402 was scanned, which yielded a high neutron reading of 60 cps above background near the top of the resin chamber.

Berkex preparations were made 5/7/15–5/11/15 per NNFD-7920-OP-186 and NNFD-7920-OP-188. The Berkex batch solvent extraction was conducted in tank T-411 on 5/12/15 with oxidant and rinses routed through the FEED/RAFFINATE bottle. After extraction with 0.6 L of 0.5 M HDEHP in dodecane, the aqueous phase was drained to the emptied and rinsed 76BX-1 FEED/RAFFINATE bottle. Berkelium-laden organic was scrubbed twice with a combination of 90 mL of 2 M $NaBrO_3$ and 500 mL of 8 M

HNO_3, and each scrubbing was drained to SR-1 and SR-2 bottles, respectively. Finally, Bk was stripped from the extractant with two contacts of 30 mL of 30% H_2O_2 and 250 mL of 8 *M* HNO_3, which were collected in the Bk product bottle.

At the beginning of draining the second strip, the dip line on the phase separator broke at the bottom of the bulb. An estimated 5 mL of product solution was lost to the cubicle floor area. The inside of the dip line was flushed to the Bk product bottle with ~10 mL of water. The remaining strip was drained slowly from T-411 to the Bk product bottle via Tygon™ tubing, after which the product was sparged and sampled as BXBK-081. Extractant was drained from T-411 to the bottle marked WO/HDEHP, which was shaken and sampled as BXWO-036. T-411 was overfilled with water (to T-409), sparged, sampled as BXFL-038, and drained to T-45. The raffinate bottle was shaken and sampled as BXRF-211, SR-1 was sampled as BXRF-212, and SR-2 was sampled as BXRF-213.

The product was sent to lab 211 for further purification. Data from purification indicated the product recovered as shown in Table 11.

Table 11. Laboratory-purified ^{249}Bk material composition (decayed to June 24, 2015)

Isotope	Mass
^{249}Bk	13.5 mg
^{249}Cf	0.519 mg
^{252}Cf	0.738 ng

Figure 16 illustrates the Bk purification process (76BX-1). Table 12 lists the sample results for 76BX-1.

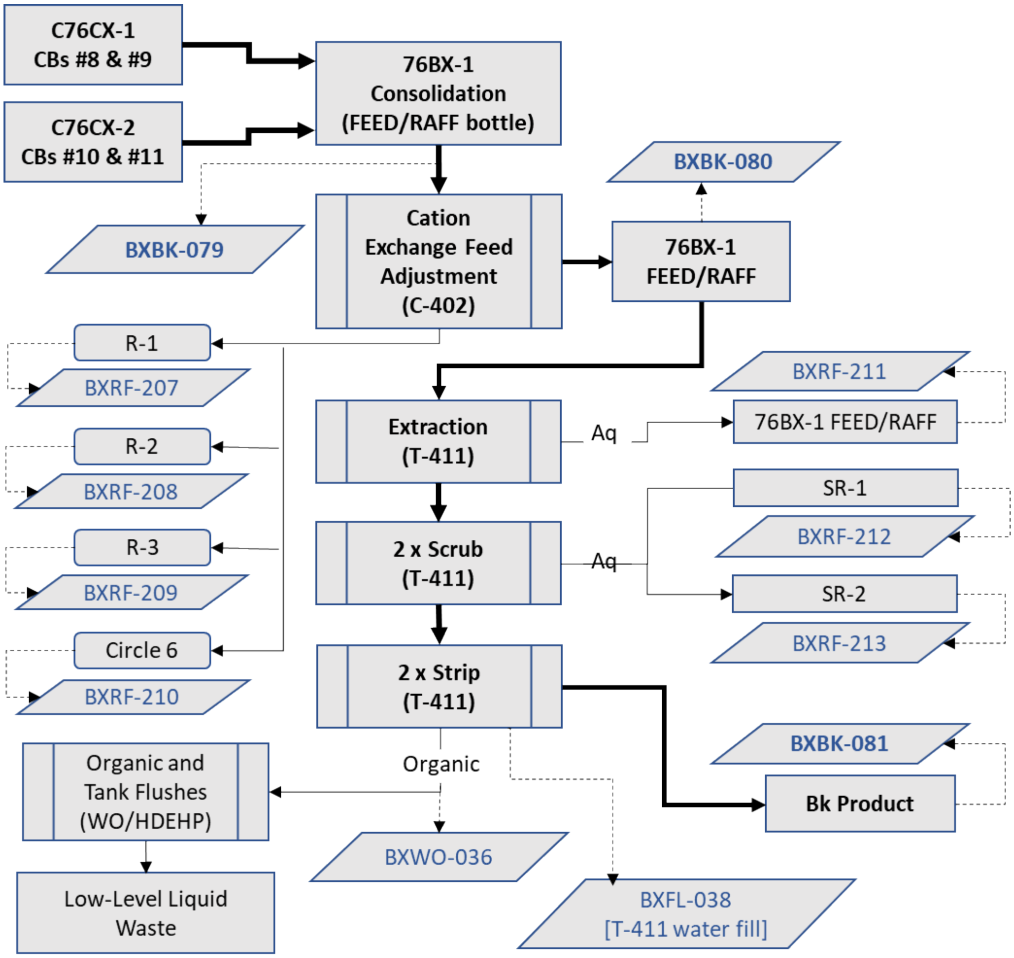


Figure 16. Berkelium feed adjustment and purification—76BX-1.

Table 12. Sample results* for 76BX-1

Sample Code	Date	Tank No.	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk (mg)	^{253}Es (µg)
BXBK-079	5/5/15	76BX-1 #1	0.026	0.000		0.002	0.000	0.378		0.025
BXBK-080	5/6/15	Feed/Raff	0.029			0.000		0.436		0.024
BXRF-207	5/7/15	76CX-1 R-1	0.000			0.000		0.001		0.000
BXRF-208	5/7/15	76CX-1 R-2	0.000							
BXRF-209	5/7/15	76CX-1 R-3								
BXRF-210	5/7/15	Circle 6						0.000		
BXFL-038	5/12/15	T-411							0.006	
BXWO-036	5/12/15	WO/HDEHP	0.024						0.041	
BXBK-081	5/12/15	Product	0.000			0.000	0.000	0.000	16.077	
BXRF-211	5/12/15	Raffinate	0.002			0.000	0.000	0.425	0.769	0.022
BXRF-212	5/12/15	SR-1	0.000			0.000		0.004	0.007	0.000
BXRF-213	5/12/15	SR-2							0.091	

* Values are *not* included for outlier results or results below limits of detection. Zero values indicate rounding *down*, nominally from the next digit (i.e., 0.0001 < value < 0.0005 would be rounded down to 0.000).

7. 76CF—CALIFORNIUM CAPSULE LOADINGS

Californium fractions from the CX runs were composited into the 76CF product bottle (see end of 76CX-2 run) and prepared for loading on cation exchange resin packages via vacuum transfer to tank T-408 in the Cub-4 high-pressure system. Each package consisted of a Pt cylinder with fritted ends that retained the cation resin. For loading, each Pt cylinder was placed in a Swagelok fitting assembly—basically, a Swagelok™ nut on both ends, leading to its reference as a “nut” package—and loaded up to 40 mg ^{252}Cf by pressurized transfer through the package (Figure 17). Interstitial liquid was displaced by N_2 after which the capsule was vented to relieve built-up pressure, disconnected, and loosely capped. Sequentially, three packages were loaded (during the primary loading runs), removed from their nut packages, inserted into a firing wand, cured for 24 hours and fired—at l50°C for 1 hour, 350°C for 1 hour, and 700°C for 4 hour—while connected to a vacuum-induced air flow during firing. Fired packages were placed back in their respective Swagelok™ nut packages for later pneumatic-line transfer to Building 7930 and further processing. Some of the Cf feed resisted loading and was collected for a later recycle CX run (76CX-3). Effluents from each primary loading run were collected in poly bottles that were monitored for neutron readings when sampled at the conclusion of loading and then later in a corner of Cub-4; the later readings are indicated in the 76CF run descriptions as they are believed to have been obtained in a more consistent manner and under more controlled conditions.

Nut package loadings were measured individually in Building 7930 as needed (from July 2016 to June 2018) to satisfy demand for ^{252}Cf sources. Loadings were determined in accordance with procedure NNFD-7930-OP-114, *Californium Assay System Cell G Workstation #4*. The procedure involves individually dissolving Cf out of a package with nitric acid and neutron counting the solution against one of two standards—CF-STD-4 (the smaller standard) or CF-STD-5 (the larger of the two standards)—depending on the estimated amount of Cf in solution. The results of these measurements were decayed to the date of nut-package loading and are shown on the process schematic and in the list of sample results for the 76CF runs.

7.1 76CF-1—CALIFORNIUM LOADING INTO NUT PACKAGE D-101

5/13/15–5/21/15

Preparations began 5/13/15 per NNFD-7920-OP-182, and maintenance was performed as needed. Before loading the first capsule, 2.4 mL of concentrated HNO_3 was added to (and sparged in) the feed (76CF product bottle; from 76CX-2 CXCF-809) but was not resampled. The first capsule was loaded from T-408 on 5/19/15 with 220 mL of feed, D-101 (neutron reading >100,000 cps @ ¼ in. from the package), with the interstitial liquor was displaced by 50 mL of water followed by another 50 mL of water per NNFD-7920-OP-183. Effluent appeared orange/red color and was collected (including water flushes) in R-4. R-4 had a final neutron reading of ~17,000 cps above background and was sampled as CXLR-515, which was deemed to warrant recycle at the end of the primary Cf runs. Tank T-408 was flushed with 2 × 100 mL of water to a poly bottle labeled “4/42/15 Circle 4” (“4/42/15” is as-indicated in the log) with each water flush appearing orange with some solids. Water flushes were decanted to T-45. T-408 was overfilled (to T-405) with 0.3 *M* HNO_3. T-405 overflow was collected in poly bottle R-5, and T-408 acid wash was collected in R-6 with only background neutron readings indicated for both bottles; all flushes were transferred to T-45.

A “large amount of black solids” were found unexpectedly (i.e., according to the procedure) on the D-101 capsule’s top frit. Most of the solids (>90%) were scraped into a 250 mL bottle labeled “76CF 5-19-15,” and the frit top was flushed with ~130 mL of in-cubicle water to bottle 76CF 5-19-15—leaving behind only a few black spots, deemed to be OK to proceed. The bottle read ~50 cps neutrons above background. The liquid was decanted from the bottle with solids and transferred to T-45.

In follow-up operations on 5/21/15, R-4 was decanted to the original feed bottle (76CF product) and flushed with 50 mL of 0.3 M HNO_3 (contacting all surfaces and allowing solids to settle). The acid was decanted to 76CF product followed by two water flushes to R-4. Neutron readings on R-4 indicated a reduction to ~1,200 cps above background. Finally, 50 mL of 0.25 M AHIB was added to R-4; mixed, settled, and decanted; and followed by a water flush. Neutron readings on R-4 after the AHIB treatment indicated 40 cps above background. Solids remaining in R-4 were transferred to 76CF 5-19-15.

Beginning on 5/20/15, Capsule D-101 was dried at 150°C (after the 24 hour cure) and then fired through the temperature sequence and returned to its nut package after cooling per NNFD-7920-OP-184.

7.2 76CF-2—CALIFORNIUM LOADING INTO NUT PACKAGE D-102

5/20/15–5/21/15

During preparations for loading D-102, the neutron probe became unresponsive and had to be replaced; its operation was confirmed with a background reading of 210 cps and a reading of 3,300 cps on the D-85 source. Effluent from acid conditioning of resin was collected in R-5 and then sent to T-45 before loading but after confirming a background neutron count. The second capsule, D-102, was loaded with 195 mL of feed and followed with two 50 mL water flushes (to R-5) on 5/20/15 per NNFD-7920-OP-183 (final neutron reading >100,000 cps @ ¼ in. from the nut package). Effluent appeared clear with no solids and was collected (including water flushes) in R-5. R-5 had a final neutron reading of ~460 cps and was sampled as CXLR-516.

A “thin layer” of “reddish/black solids” were found unexpectedly on the capsule’s top frit when it was inspected after disconnecting it from the holder. The solids were removed with ~150 mL of water, which seemed to dissolve almost all of the solids, and placed in the bottle labeled “76CF 5-19-15” (joining the solids from D-101). The bottle read ~50 cps neutrons above background.

Per NNFD-7920-OP-184, capsule D-102 was fired through the temperature sequence beginning on 5/21/15 and returned to its nut package after cooling.

7.3 76CF-3—CALIFORNIUM LOADING INTO NUT PACKAGE D-103

5/21/15–5/22/15

The third capsule, D-103, was loaded with 200 mL of feed on 5/21/15 per NNFD-7920-OP-183 (final neutron reading >100,000 cps @ ¼ in.). Effluent appeared clear with no solids and was collected (including three 50 mL water flushes) in R-6. R-6 had a final neutron reading of ~1,600 cps above background and was sampled as CXLR-517.

Like D-102, a thin layer of reddish/black solids were found unexpectedly on the capsule’s top frit. The solids were removed with water—which seemed to dissolve almost all of the solids—and placed in the bottle labeled “76CF 5-19-15” (joining the solids from D-101 and D-102). A total of ~170 mL of solution was collected in the bottle labeled “76CF 5-19-15” from removing solids observed in the three nut packages (D-101, D-102, and D-103). The bottle read ~60 cps neutrons above background.

A small indentation was noticed on the edge of the frit upon inspection after loading. Per NNFD-7920-OP-184, capsule D-103 was fired through the temperature sequence beginning on 5/22/15 and returned to its nut package after cooling. (The distorted frit was not observed after calcination, but upon its later use in fabricating bulk wire, the contents seemed to spill out of the capsule unexpectedly.)

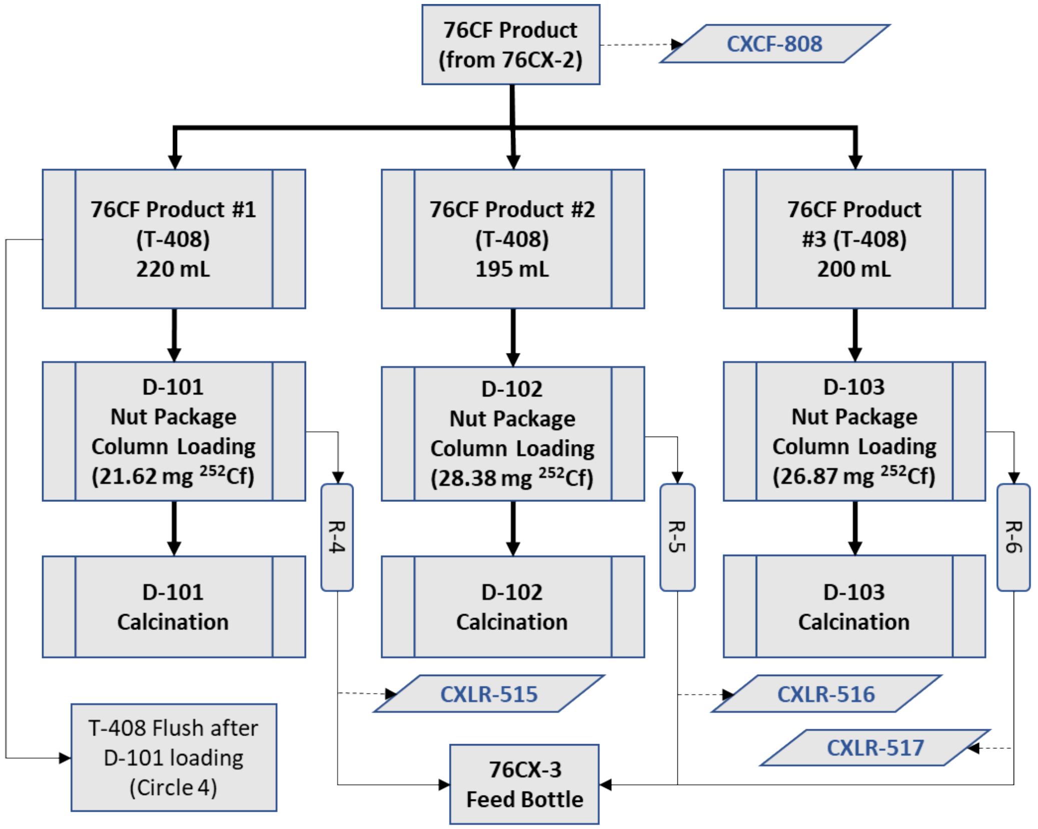


Figure 17. 76CF-1/-2/-3—Loading and calcining D-101, D-102, and D-103 nut packages.

Table 13. Sample results* for 76CF-1/-2/-3

Sample Code	Date	Tank No.*	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{253}Es (µg)
CXCF-808	5/5/15	76 Cf Product		0.001	0.000	0.020	0.005	84.12	2.054
N. Assay	5/19/15	D-101						**21.62**	
CXLR-515	5/19/15	R-4		0.000		0.001	0.000	9.318	0.209
N. Assay	5/20/15	D-102						**28.38**	
CXLR-516	5/20/15	R-5	0.003	0.000		0.001	0.000	0.070	
N. Assay	5/19/15	D-103						**26.87**	
CXLR-517	5/21/15	R-6	0.001			0.000	0.000	0.633	0.016

* Values are *not* included for outlier results or results below limits of detection. Zero values indicate rounding *down*, nominally from the next digit (i.e., 0.0001 < value < 0.0005 would be rounded down to 0.000).

8. 76CX-3—CATION EXCHANGE RECOVERY AND PURIFICATION OF LOADING-RESISTIVE CALIFORNIUM

Some of the Cf product from the 76CX-1 and 76CX-2 runs resisted loading onto the cation resins in the D-101, D-102, and D-103 nut packages. Loading-resistive Cf and other actinides were collected in poly bottles R-4, R-5, and R-6, which were consolidated to form feed for run 76CX-3. Californium and other actinides were recovered and separated by cation exchange involving AHIB as a complexing agent as described previously for the earlier CX runs. As before, actinide separation was determined by tracking the neutron peak from Cf and monitoring the alpha trace coming off the column during the 76CX-3 run.

8.1 76CX-3—CATION EXCHANGE RECOVERY AND PURIFICATION OF LOADING-RESISTIVE CALIFORNIUM

5/26/15–5/27/15

Preparations for 76CX-3 began on 5/26/15 in accordance with procedures NNFD-7920-OP-177 and NNFD-7920-OP-178. Effluents collected in R-5 (~300 mL) during D-102 loading and in R-6 (~290 mL) during D-103 loading were consolidated on 5/26/15 along with the liquid in "76CF Product" (decanted from R-4 solids, including acid, AHIB, and water flushes to recover actinides; ~500 mL total) into a 2 L poly bottle labeled "76CX-3 Feed" and sampled (after an acid adjustment as CXAF-110 (0.3 N H+). The C-401/C-402 system was tested with water, which was collected (but not sampled) in R-6.

Actinides in the feed were loaded from the 76CX-3 feed bottle (via T-404) onto column C-402 (Figure 18) on 5/27/15 per NNFD-7920-OP-179 with discharge collected in R-4. This was followed by 80 mL of water rinse, 300 mL of 0.3 M NH_4OH, and a second 80 mL of water in the "76CX-3 Feed" bottle and through the columns with discharge collected in R-5. Elution of actinides from C-402 through C-401 was performed with 0.25 M AHIB at pH 4.2 per NNFD-7920-OP-180 with effluent collected in CB #1–CB #6. Column C-401 then was treated with 0.5 L of 0.5 M AHIB solution at pH = 4.8 followed by 0.4 L of water, with effluent collected in a poly bottle labeled "Circle 16." Maximum neutron readings of the columns indicated 180 cps for C-401 and 210 cps for C-402 above background. Each of the CBs was acidified and sampled as follows:

1. CB #1—CXER-410 (235 mL)
2. CB #2—CXER-411 (230 mL)
3. CB #3—CXER-412 (75 mL)
4. CB #4—CXCF-810 (230 mL)
5. CB #5—CXCF-811 (160 mL)
6. CB #6—CXCF-812 (160 mL)

The contents of CB #4 provided feed for 76CF-4. Effluents collected in CB #1, CB #2, and CB #3, and in poly bottles R-4, R-5, and R-6 were recycled and rinsed to T-45. Resin in columns C-401 and C-402 were removed and collected in the C76 used CX resin bottle per NNFD-7920-OP-174 and NNFD-7920-OP-173, respectively.

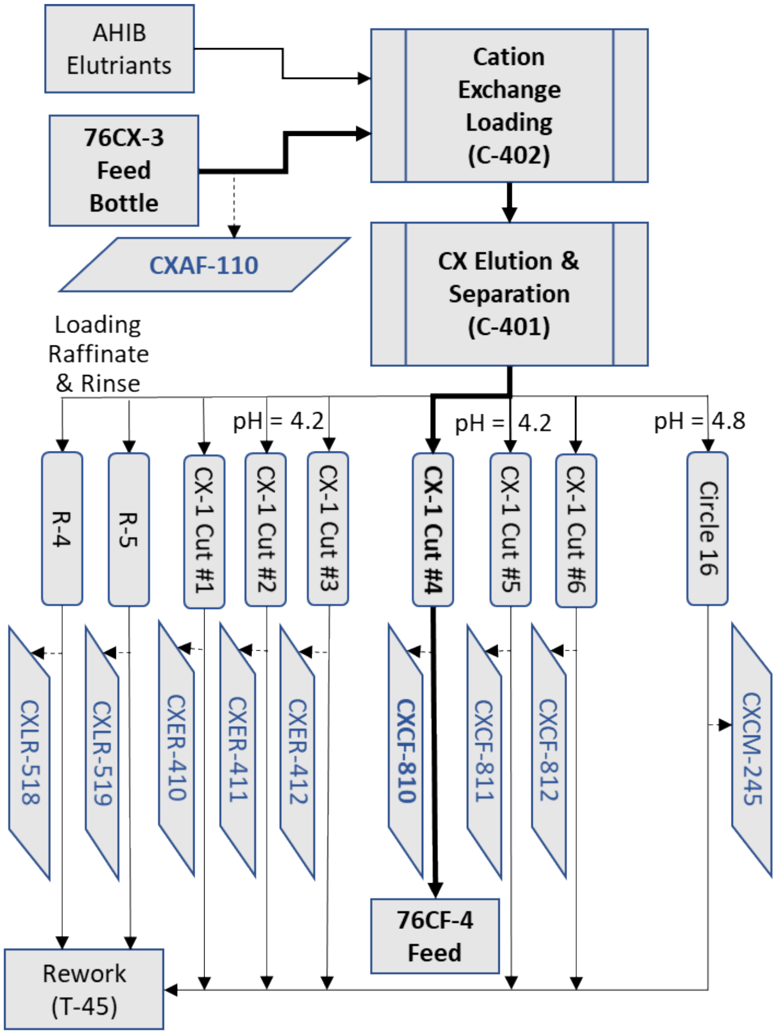


Figure 18. AHIB cation exchange column purification of 76CF-1/2/3 raffinates—76CX-3.

Table 14. Sample results* for 76CX-3

Sample Code	Date	Tank No.	^{252}Cf (mg)	^{253}Es (µg)
CXAF-110	5/26/15	76CX-3 Feed	7.777	0.192
CXLR-518	5/27/15	R-4	0.000	
CXLR-519	5/27/15	R-5	0.000	
CXER-410	5/27/15	CB #1	0.000	0.000
CXER-411	5/27/15	CB #2	0.001	0.001
CXER-412	5/27/15	CB #3	0.003	1.749
CXCF-810	5/27/15	CB #4	7.650	
CXCF-811	5/27/15	CB #5	0.169	
CXCF-812	5/27/15	CB #6	0.016	
CXCM-245	5/27/15	Circle 16	0.026	0.001

* Values are *not* included for outlier results or results below limits of detection. Zero values indicate rounding *down*, nominally from the next digit (i.e., 0.0001 < value < 0.0005 would be rounded down to 0.000).

9. 76CF-4—CAPSULE LOADING WITH LOADING-RESISTIVE CALIFORNIUM

Some of the Cf feed resisted loading into nut packages D-101, D-102, and D-103 and was collected and recovered during a recycle cation exchange run (CX-3). The product of CX-3 (230 mL) was collected in CB #4, designated 76CF-4.

Nut package D-104 loading was measured in Building 7930 in accordance with NNFD-7930-OP-114, when it was needed (in August 2016) to satisfy demand for ^{252}Cf sources. The result of this measurement was decayed to 5/27/15, the date of nut-package loading as shown on the process schematic and in the list of sample results for the 76CF-4 run.

With recovery and packaging of loading-resistive Cf, a total of 85.78 mg ^{252}Cf was confirmed by neutron assay to have been loaded in the four product nut packages. Nut Packages D-102, D-103, and D-104 were transferred pneumatically to Building 7930 on 12/27/16.

9.1 76CF-4—CALIFORIUM LOADING INTO NUT PACKAGE D-104

5/29/15–6/1/15

Loading-resistive Cf collected in CB #4, labeled "76CF-4" (230 mL), was loaded onto nut package D-104 resin beginning 5/29/15 per NNFD-7920-OP-183. Raffinate was collected in R-4 (which read 80 cps above background) and was poured to T-45. An inspection of D-104's top frit revealed no solids and an intact frit. On 6/27/15, T-404, T-405, and T-408 were flushed to T-45 (MSA-810; representing all recycle from nut package loadings) per SOWP042488/0, which then was jetted and flushed to F-115.

Per NNFD-7920-OP-184, capsule D-104 was fired through the temperature sequence beginning on 5/30/15 and reinstalled in its nut package after cooling (Figure 19). Californium nut packages D-101, D-102, D-103, and D-104 were all placed in a holder on top of the Cub-4 box furnace.

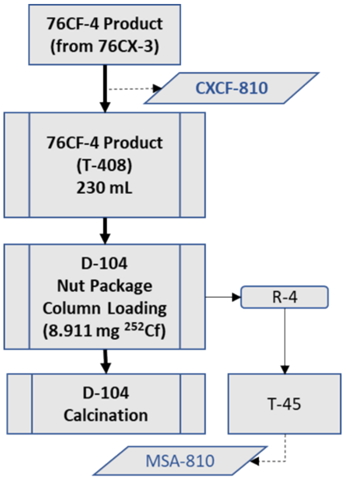


Figure 19. 76CF-4—Loading and calcining D-104 nut package.

Table 15. Sample results* for 76CF-4

Sample Code	Date	Tank No.	^{252}Cf (mg)
CXCF-810	5/27/15	76CF Cut #4	7.650
N. Assay	5/27/15	D-104	**8.911**
MSA-810	6/27/15	T-45	0.029

* Values are *not* included for outlier results or results below limits of detection. Zero values indicate rounding *down*, nominally from the next digit (i.e., 0.0001 < value < 0.0005 would be rounded down to 0.000).

10. 76OH—HYDOROXIDE PRECIPITATION FOR LICL REMOVAL FROM TRANSCURIUM ACTINIDES

Like 76CM transcurium cuts, Am/Cm products from a LiCl anion exchange column runs contain LiCl salts that must be removed before it can be concentrated and stored properly. These salts and other contaminants that do not precipitate are removed by precipitating actinides as hydroxides, filtering the precipitated actinides in a deep-bed filter (DBF), and re-dissolving them in nitric acid (similar to the treatment performed for transcurium cuts during the 76CM-3(LiOH) run). The precipitation process requires techniques to add reagents (sequentially, hydroxides, acids and rinses), agitate by sparging, allowing precipitates to settle, and then decanting the liquor while leaving precipitates undisturbed in the tank. In addition to removing LiCl entrained into the Am/Cm cuts during CM runs, this process converts the purified products from a chloride form in an HCl solution to a nitrate form in a HNO_3 solution suitable storage and further processing.

Curium products (and a pre-Cm cut) consolidated in T-60 at the end of the 76CM runs (05/17/15) served as feed for this run. Recall that this material was contaminated with extra Pu (likely from another process being conducted in the cubicles). The composite of Cm products was sampled in T-60 as CMAP-307A, CMAP-307B, and CMAP-307C. After this feed was adjusted, half of the feed solution was used in 76OH-1, and the remainder was used in 76OH-2. The flow sheets and results tables of the feeds and the products for these two 76OH runs were consolidated below. Curium products from 76OH runs were retained for later addition to Cm recovered during C77; no targets were fabricated directly from Cm recovered during this campaign, C76.

10.1 76OH-1 HYDROXIDE PRECIPICATION FOR LICL REMOVAL FROM TRANSCURIUM ACTINIDES

6/3/15–6/19/15

Reagent make-ups, system preparations, and DBF recharging began on 6/3/15 per procedure NNFD-7920-OP-154, NNFD-7920-OP-155, and NNFD-7920-OP-206, respectively. MDV-637 required replacement when a retaining nut on the stem sheared off. On 6/16/15 and according to NNFD-7920-OP-156, consolidated Cm products from the 76CM runs were evaporated to ~16 L in T-60, then diluted to 18 L, and sampled as OHPF-049. Nine liters was transferred to T-61 as feed 76OH-1, which was chased by 2 × 4.5 L of water and sampled in T-61 as OHPF-050.

On 6/17/15, 8 L of 10 *M* NaOH was made up and added to T-61 (and chased with 2 × 2 L of water). This yielded a final solution with 2 to 3 *N* OH^- which was sparged for over 2 hours to neutralize the acid and precipitate the actinides. After settling for more than 2 hours, T-61 liquid then was transferred through the DBF (via T-444), collected in T-21 (via T-430) in 3 × 4.5 L batches and flushed with two 4 L water—leaving the precipitate in T-61. To dissolve the precipitate, T-61 was treated once with 2.5 L of 8 *M* HNO_3 + 1 L of water flush plus 3 × (2.5 L of 1 *M* HNO_3 + 0.5 L of water), each of which was passed sequentially through the DBF (to dissolve precipitates collected thereon) with effluent collected in T-43. T-61 also was flushed (and sparged) with 3 × 2 L of water through the DBF to T-43, which was evaporated to 5 L but not sampled. T-21 was acidified with 7.25 L of 6 *M* HCl and sampled on 6/19/15 as OHFL-068.

10.2 76OH-2 HYDROXIDE PRECIPICATION FOR LICL REMOVAL FROM TRANSCURIUM ACTINIDES

6/18/15–6/28/15

Feed in T-60 for 76OH-2 was evaporated from 9 L to 6 L on 6/18/15 in anticipation of extra dilution from water-flushing a to-be-emptied T-60. The DBF was disassembled and emptied of filter aid, which was collected in a bottle labeled "76OH-1 Used Filter Aid," which was moved to Cub-6 for decanting to T-66. DBF parts were flushed with water, reassembled, pressure tested, and recharged with fresh filter aid. The balance of the T-60 feed and flushes were transferred to T-61 on 6/22/15. A pH check resulted in 9 L of 10 M NaOH being metered into T-61 to neutralize the acid, precipitate the actinides, and yield 2–3 N OH^- in the final solution; T-61 was not resampled. On 6/23/15, the T-61 solution was fed through the DBF (Figure 20), collected in T-21, and flushed with 3 × 4.5 L of 2 M NaOH and 2 × 4 L of water—with care taken to leave the precipitate intact in T-61.

On 6/23/15, T-61 was treated with 2.5 L of 8 M HNO_3 (plus an additional line rinse with 2 × 0.5 L of water). The acidic solution was transferred through the DBF to T-43. This system was flushed to T-43 with 3 × 2.5 L of 1 M HNO_3 and 3 × 2 L water flushes, after which T-61 was sampled as MSA-806. T-60 was sampled as MSA-807 after it was concentrated to ~15 L. T-21 was sampled as OHFL-070 after some HCl and water flushes were collected in it. T-43 was evaporated to a volume of ~4.5 L to gain freeboard. T-43 then was sampled on 6/25/15 as OHDP-031, transferred to T-300 for storage, and flushed with 4 × 2 L of 1 M HNO_3 and 2 × 2 L water. Finally, it was sampled as CMAP-308A, CMAP-308B, and CMAP-308C. Added 1.5 L of concentrated HNO_3 and ~15.5 L of rinse water to T-43, evaporated down to ~5 L and sampled as MSA-809.

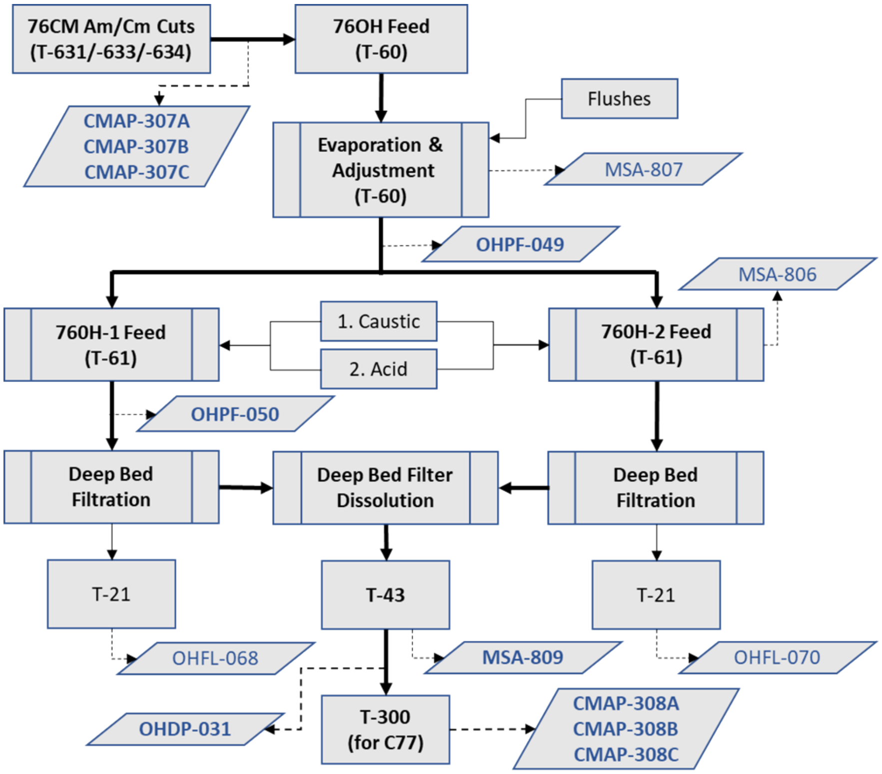


Figure 20. 76OH-1 and 76OH-2—Actinide purification by hydroxide precipitation.

Table 16. Sample results* for 76OH-1 and 76OH-2

Sample Code †	Date	Tank No.	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)
CMAP-307A	5/18/15	T-60	0.178	3.959	0.858	44.952	10.609	0.020
CMAP-307B	5/18/15	T-60	0.344	3.779	0.819	43.869	10.353	0.064
CMAP-307C	5/18/15	T-60	1.421	3.188	0.691	45.627	10.768	0.088
OHPF-049	6/16/15	T-60		3.602	0.781	36.879	8.704	0.746
OHPF-050	6/16/15	T-61	0.447	1.457	0.316	17.231	4.067	
OHFL-068	6/19/15	T-21	0.049	0.017	0.004	0.166	0.039	
OHDP-031	6/25/15	T-43	0.357	3.523	0.764	39.312	9.278	0.032
MSA-806	6/25/15	T-61		0.005	0.001	0.007	0.002	
MSA-807	6/26/15	T-60	0.007	0.001	0.000	0.012	0.003	
CMAP-308A	6/27/15	T-300	0.363	3.584	0.777	46.151	10.892	
CMAP-308B	6/27/15	T-300	0.379	3.392	0.735	44.142	10.418	0.022
CMAP-308C	6/27/15	T-300	0.379	3.712	0.804	44.084	10.404	0.010
OHFL-070	6/26/15	T-21		0.025	0.005	0.280	0.066	
MSA-809	6/28/15	T-43	0.001	0.003	0.001	0.038	0.009	

* Values are *not* included for outlier results or results below limits of detection. Zero values indicate rounding *down*, nominally from the next digit (i.e., 0.0001 < value < 0.0005 would be rounded down to 0.000).

† OHPF-049 represents total feed for both 76OH runs (half for each run)**.** OHPF-050 and OHFL-068 represents feed and waste, respectively, for Run 76OH-1. All other results apply to Run 76OH-2.

11. CAMPAIGN 76 REWORK

During the campaign, various rework solutions were collected in F-115 and T-23 from several C76 processes as CY15F115CL-1 and CY15T23CL-1A, respectively, and actinides were recovered during Cleanex extraction stages described previously. Actinides removed from the aqueous by extraction in F-115 were stripped from the organic phase in operations conducted in Tank T-77. The strip aqueous from the F-115 Cleanex extraction was added to T-23 for recovery as part of CY15T23CL-1B. Actinides recovered from T-23 C76 rework solutions and from the CY15F115CL-1 extraction were consolidated and characterized as CY15T23CL-1 (after CY15T23CL-1B extraction) and were stored in T-302 until purification during belated C76 oxalate precipitation runs (in late-2016/early-2017) and subsequent use in C77.

11.1 CY15F115CL-1 CLEANEX REWORK

1/17/15–9/08/15

Aqueous solutions transferred to F-115 were essentially waste materials bearing small amounts of actinides and contaminants collected from different processes. (Note that these processes included some not related to C76; but essentially all transplutonium actinides related to the Cf campaign.) Actinides in this stream were recovered by a series of batch liquid–liquid extractions in F-115 (CY15F115CL-1A through -1E) according to NNFD-7920-OP-123. Each batch extraction involved several sequences of sparging the organic and aqueous phases followed by settling, sampling, and adjustments as needed (typically, adding NaOH and HDEHP/Exxsol D60) to maintain extraction conditions. When aqueous-phase samples indicated that contents met disposal goals or limits, the raffinate was transferred to F-126 for disposal to waste.

The extractant was transferred to tank T-77 on 8/19/15, where it was stripped of recovered actinides using eight strip cycles of ~6 *N* HNO_3 over the period 8/21/15–8/27/15 per NNFD-7920-OP-146. Strips were decanted to T-604 where they were washed batchwise with diluent (Exxsol D60) to remove entrained organic (otherwise, the HDEHP extractant could decompose during evaporation, forming a tar which could sorb a significant amount of actinide elements). Diluent-washed strips then were consolidated in T-60, where they were evaporated, adjusted for acidity batchwise through 8/27/15 to serve as CY15T23CL-1B feed, and sampled as CLCP-428A and CLCP-428B. The stripped organic was sampled in T-77 as CLWO-122, and the diluent-washed organic was sampled in T-604 as CLWO-123.

Actinide products recovered during the CY15F115CL-1 extraction are listed in Table 17, and wastes are shown in Table 18 with actinide disposal goals and limits. Aqueous strips from CY15F115CL-1 augmented rework solutions in T-23 for further recovery during the CY15T23CL-1B extraction.

Table 17. Sample results for actinide products recovered during CY15F115CL-1

Sample Code	Date	Tank No.	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)
CLCP-428A	8/28/15	T-60	0.569	0.101	2.418	0.669	1.925
CLCP-428B	8/28/15	T-60	0.569	0.101	2.413	0.668	1.925

Table 18. Sample results for CY15F115CL-1 wastes *

Nuclide	Disposal Goal	Disposal Limit	-1A Raff. (SFW-839)	-1B Raff. (SFW-843)	-1C Raff. (SFW-847)	-1D Raff. (SFW-853)	-1E Raff. (SFW-859)	Organic † (post-strip)
^{241}Am (mg)	≤25	50	0.002	0.011	0.010	0.013	0.028	0.003
^{244}Cm (mg)	≤15	15	0.27	1.24	1.15	1.45	3.17	0.021
^{252}Cf (μg) ‡	≤50	200				6.00 (n)		1.33 (n)

* Values are *not* included for outlier results or results below limits of detection.
† Organic includes post-strip organic results in T-77 and organic from diluent wash in T-604.
‡ Californium-252 results determined either by Gross α counts (α) or by Gross neutron counts (n) from fission

11.2 CY15T23CL-1A CLEANEX REWORK

8/24/15–9/1/15

Recovery of actinides from aqueous rework solutions collected in T-23 began with the addition of HDEHP/Exxsol D60 extractant on 8/24/15 per NNFD-7920-OP-146. After four acid adjustments and sparges, CLER-307 sample results indicated that all limits were met for disposing T-23 raffinate from the CY15T23CL-1A extraction, so the aqueous was decanted to T-21 on 8/28/15, sampled as CLER-308 to confirm meeting disposal limits, and transferred to F-115 on 9/1/15 for disposal. The product of CY15T23CL-1A was characterized after the CY15T23CL-1B extraction.

11.3 CY15T23CL-1B CLEANEX REWORK

8/27/15–12/29/15

The final adjustment of aqueous strip solutions collected in T-60 was completed on 9/4/15 and sampled as CLCP-430, leading directly into actinide recovery by Cleanex extraction in T-23 per NNFD-7920-OP-146. After five acid adjustments, sparges/settlings, and samplings, CLER-313 sample results indicated that limits were met for disposing T-23 raffinate from the CY15T23CL-1B extraction. The aqueous raffinate was decanted to T-21 between 9/10/15 and 9/13/15, and the organic phase was transferred from T-23 to T-60 for further scrubbing. The organic in T-60 then was scrubbed twice (between 9/14/15 and 9/15/15) with both scrubs decanted to T-21. A composite CY15T23CL-1 aqueous waste solution was sampled as CLSR-929 on 9/15/15 and transferred to F-115 for disposal.

The scrubbed organic was stripped eight times in T-60 (9/15/15–9/30/15) with each strip product decanted to T-40, where they were concentrated. Stripping spanned 2 weeks because of problems pumping strip solutions to T-40 and other troubleshooting/repair issues. The composite product (after water flush and sparging) for CY15T23CL-1 was sampled on 10/6/15 as CLCP-433A, CLCP-433B and CLCP-433C with wide-ranging analytical results. The composite strip was transferred to T-302 on 12/21/15 and resampled on 12/29/15 as CLCP-434A, CLCP-434B, and CLCP-434C, yielding more consistent sample results. Stripped organic was sampled from T-60 on 9/30/15 as CLWO-124, and diluent-washed organic was sampled from T-604 on 10/3/15 as CLWO-125. Actinide products recovered during the CY15T23CL-1 extraction are listed in Table 19, and wastes are shown in Table 20 with actinide disposal goals and limits.

Table 19. Sample results for actinide products recovered during CY15T23CL-1

Sample Code	Date	Tank No.	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)
CLCP-433A	10/6/15	T-40	1.769	0.313	9.198	2.546	11.42
CLCP-433B	10/6/15	T-40	0.788	0.139	4.745	1.313	11.45
CLCP-433C	10/6/15	T-40	0.829	0.147	0.040	0.011	11.48
CLCP-434A	12/29/15	T-302	0.839	0.149	4.513	1.249	10.42
CLCP-434B	12/29/15	T-302	0.839	0.149	4.698	1.300	10.53
CLCP-434C	12/29/15	T-302	0.839	0.149	4.681	1.296	10.77

Table 20. Sample results for CY15T23CL-1 wastes*

Nuclide	Disposal Goal	Disposal Limit	Aqueous Wastes (CLSR-929)	Post-strip Organic † (CLWO-124 & -125)
^{241}Am (mg)	≤25	50	0.873	0.021
^{244}Cm (mg)	≤15	15	7.80	0.170
^{252}Cf (µg) ‡	≤50	200	25.4	0.687 (n)

* Values are *not* included for outlier results or results below limits of detection.

† Organic includes post-strip organic results in T-77 and organic from diluent wash in T-604.

‡ Californium-252 results determined either by Gross α counts (α) or by Gross neutron counts (n) from fission

12. 76OX AMERICIUM/CURIUM OXALATE PRECIPITATION AND PURIFICATION (OX)

Americium and curium recovered from C76 rework runs (CY15F115CL-1 and CY15T23CL-1) were purified from ionic contaminants in four batches by two-cycle OX runs. Feed for these runs was rework product stored in tank T-302) since December 2015; it was resampled on 9/26/16 as CLCP-435A and CLCP-435B to confirm the contents. The T-302 solution was transferred to T-54 in three increments (plus waster flush) and adjusted to 4.0 L at ~1 *M* free acid per NNFD-7920-OP-199. Subsequently, it was transferred batchwise for each OX run to a stirred-agitator tank (T-451) via tank T-430 (where the solution could be observed by an in-cubicle camera) per NNFD-7920-OP-200. Some filtrates were recycled to either T-54 or T-451 in OX runs subsequent to the first cycle of the first run.

Oxalic acid (0.8 *M*) was added to each batch of feed to form actinide oxalate precipitates. For both cycles of each OX run, the slurry was filtered in F-454, and the filtrate was collected for tertiary recovery of actinides. Acid was added to T-451 to re-dissolve any residual precipitates and passed through F-454 to re-dissolve the filtered solids. For the first cycle, re-dissolved solids were collected in an intermediate receiving tank (T-458) and recycled back to T-451 for the second precipitation cycle per NNFD-7920-OP-201. For the second cycle, re-dissolved solids were collected in T-43, where they were collected and sampled as OXDP-755A and OXDP-755B on 1/7/17 at the end of 76OX-4. The composite product of four 76OX runs was boiled to a temperature slightly above 119°C (almost to dryness) to destroy the oxalate form per NNFD-7920-OP-202. The composite became part of the feed for C77's CL-1 run when it was added to T-72 on 2/9/17.

Actinide residues in the OX filtrates were recovered by multi-batch, tertiary precipitations. OX filtrates were recycled to T-451 and adjusted to conditions amenable to actinide precipitation using concentrated ammonium hydroxide per NNFD-7920-OP-203. The tertiary precipitates were filtered as an intermediate product. Re-filtered precipitates accumulated on F-454 were re-dissolved with nitric acid. Since re-filtration products are considered to be of good quality (like first cycle precipitation products), and since the small amount of actinides in re-filtered product reduce the excess free-acid content only slightly (as intended), the product solutions bearing actinides recovered from tertiary precipitations are recycled for re-dissolving the first-cycle precipitate of the subsequent run in the series. Filtrates from tertiary precipitations were collected after each filtration run in T-40, where they were treated by evaporation to destroy the oxalate before joining scrub solutions from the C77 CL-1 run.

12.1 76OX-1 AMERICIUM/CURIUM OXALATE PRECIPITATION AND PURIFICATION

12/19/16–12/28/16

Feed for 76OX-1 was transferred from T-54 to the precipitator (T-451) on 12/19/16, during which it was observed to be "light brown in color/no solids" in transfer tank T-430. The first cycle was completed the next day, and the second cycle was completed on 12/21/16. The dissolved product was sampled in T-43 on 12/24/15 as OXDP-752.

The filtrates from both cycles were collected in T-456 and T-458 and sampled as OXFL-623 and XFL-624, respectively. Soluble actinides in the filtrates were not recovered at the end of 76OX-1; however, to free up T-456 (for first-cycle use during 76OX-2), 76OX-1's first-cycle filtrate was transferred to a Kimax bottle labeled "76OX-1 T456 Filtrate" for later recovery with filtrates generated during 76OX-2. The flow sheet and sample results for 76OX-1 were incorporated into the flow sheet and sample results for 76OX-2 at the end of next section.

12.2 76OX-2 AMERICIUM/CURIUM OXALATE PRECIPITATION AND PURIFICATION

12/27/16–12/30/16

Feed for 76OX-2 was transferred from T-54 (OXPF-419) to the precipitator (T-451) on 12/28/16, during which it was observed to be "slightly brown with no solids" in transfer tank T-430. The first and second cycles were completed on 12/29/16. Product (a composite of 76OX-1 and 76OX-2) was sampled in T-43 as OXDP-753.

Filtrates collected from 76OX-1 and 76OX-2 were treated after 76OX-2 (12/29/16–12/30/16) to recover small amounts of soluble actinides by re-precipitating and re-filtering in 5 batches. Upon re-dissolution, recovered intermediate product was collected in T-458 and sampled as OXDP-754 for later use to dissolve the first cycle precipitate in 76OX-3. The flow sheets and sample results for 76OX-1 and 76OX-2 are shown in Figure 21 and Table 19.

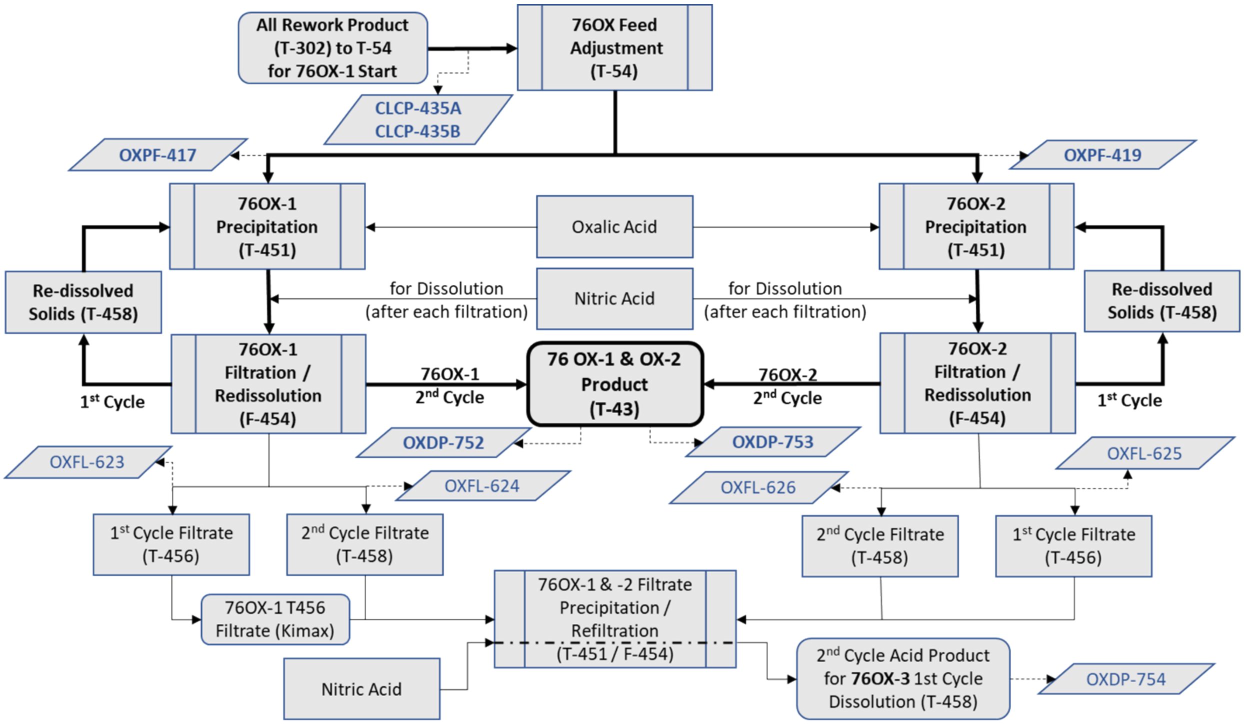


Figure 21. 76OX-1 and 76OX-2—Purification of actinides in recovered rework by oxalate precipitation.

Table 19. Sample results* for 76OX-1 and 76OX-2

Sample Code †	Date	Tank No.	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)
CLCP-435A	12/13/16	T-302	0.891	0.164	4.813	1.332	9.287
CLCP-435B	12/13/16	T-302	0.960	0.163	4.786	1.325	9.517
OXPF-417	12/14/16	T-54	0.296	0.064	3.993	0.942	2.963
OXFL-623	12/21/16	T-456	0.107	0.023	0.799	0.188	0.893
OXFL-624	12/21/16	T-458	0.002	0.000	0.017	0.004	0.023
OXDP-752	12/24/16	T-43	0.284	0.062	2.144	0.506	3.388
OXPF-419	12/27/16	T-54	0.014	0.003	0.195	0.046	0.194
OXFL-625	12/29/16	T-456	0.008	0.002	0.055	0.013	0.058
OXFL-626	12/29/16	T-458	0.001	0.000	0.011	0.003	0.006
OXDP-753	12/29/16	T-43	0.270	0.059	2.448	0.578	3.917
OXDP-754	12/30/16	T-458	0.018	0.004	0.247	0.058	0.157

* Values are *not* included for outlier results or results below limits of detection. Zero values indicate rounding *down*, nominally from the next digit (i.e., 0.0001 < value < 0.0005 would be rounded down to 0.000).

† OHDP-753 represents total product for both 76OX-1 and 76OX-2

12.3 76OX-3 AMERICIUM/CURIUM OXALATE PRECIPITATION AND PURIFICATION

1/4/17–1/6/17

Feed for 76OX-3 was transferred from T-54 to the precipitator (T-451) on 1/4/17, during which it was observed to be “slightly brown with no solids.” The feed was not sampled except for an acid check. First cycle filtrate was collected in T-456. Acid used for dissolving the re-filtered precipitate (after 76OX-2 and retained in T-458) was used to dissolve the first cycle precipitate for 76OX-3. Second cycle feed also was not sampled except for an acid check. Filtrate from the second cycle precipitation was collected in T-458, and the precipitate was dissolved and pumped to T-43, completing the second cycle on 1/6/17. No product sample was obtained for 76OX-3, *per se*.

As was done after 76OX-2, the filtrates from 76OX-3’s standard filtrations were treated by precipitation and re-filtration in five batches to recover soluble actinides on 1/5/17–1/6/17. The filtrates were sent directly to T-40 for collection and evaporation. The accumulation of filtered product (on F-454) was re-dissolved with acid routed through T-456 (to dissolve any residual actinides), collected in T-458, and used to dissolve the first cycle precipitate in 76OX-4. The flow sheet and sample results for 76OX-3 are included in next section with the flow sheet and sample results for 76OX-4.

12.4 76OX-4 AMERICIUM/CURIUM OXALATEPRECIPITATION AND PURIFICATION

1/6/17–1/16/17

Feed for 76OX-4 was transferred from T-54 to the precipitator (T-451) on 1/6/17, during which it was observed to be “cloudy with no solids.” As the last OX run of the series, the feed volume was lower than that used for the other OX runs (~1 L compared to ~4 L or more in the others). The feed was not sampled except for an acid check. The first cycle precipitation required the use of an alternative sparging method for T-451 because the mixer seized up 15 minutes into operation. This alternative method was achieved by applying the in-cell vacuum to a vacuum/pressure port on T-451 with the sample bottle removed from the sampler needle block. This allowed air to be pulled into T-451 via the sampler line that is near the bottom of the tank and thus bubbling (sparging) the solution in T-451. Acid product from 76OX-4 was used to dissolve the filtered precipitate. The dissolved solids were sampled in T-451 as OXPF-422.

During the second cycle (1/7/15), the T-451 sparge was not working (later deemed to be a plugged vacuum jet), and solution was running out of the needle block. The vacuum was quickly removed from sampler to stop siphoning liquid, but not before ~1.8 L leaked to Cub-4 floor. This malfunction left 1.4 L in T-451. Solution in sample lines was pushed out of needle block with an air purge of T-451. Solution in the sump was evaporated to ~1.0 L and sampled as MSA-169. Solution from Cub-4 sump was flushed to two 1 L Kimax bottles labeled "76OX-4B" (MSA-171B; 0.65 L) and "76OX-4C" (MSA-171C; 0.9 L). Although not logged, it was presumed (based on similarities in the sample code, sample date and analytical results) that a bottle labeled "76OX-4A" also received solution from the leak that collected in the Cub-4 floor sump, and this bottle was sampled as MSA-171A (0.9 L).

Second cycle filtrate was collected in T-458, and acid was added to dissolve the precipitate in F-454 and send it to T-43, which was sampled as OXDP-755A and OXDP-755B on 1/7/17. Only product evaporation and system flushing remained to be done beyond that. T-43 was evaporated from 1/13/17 to 1/16/17 until temperature leveled off at 119.4°C to destroy oxalate residues. Flushes and filtrates (collected in T-40) were evaporated to 119.3°C to destroy oxalate residues as well and were sampled as OXFL-628A and OXFL-628B. The composite of flushes and tertiary filtrates was transferred to T-23 on 1/13/17, where it was joined by scrub solutions from the 77CL-1 run. The flow sheets and sample results for 76OX-3 and 76OX-4 are shown in Figure 22 and Table 20.

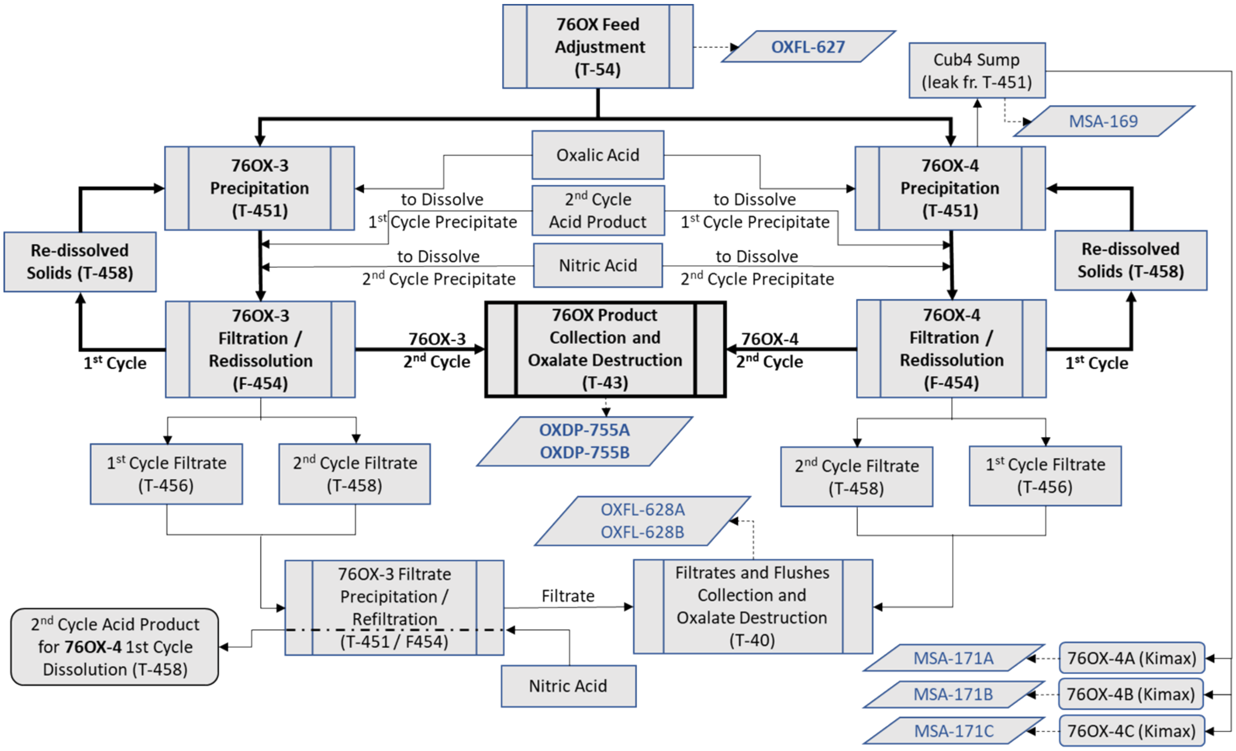


Figure 22. 76OX-3 and 76OX-4—Purification of actinides in recovered rework by oxalate precipitation.

Table 20. Sample results* for 76OX-3 and 76OX-4

Sample Code †	Date	Tank No.	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)
OXDP-755A	1/7/17	T-43	0.572	0.124	4.251	1.003	6.771
OXDP-755B	1/7/17	T-43	0.572	0.124	4.370	1.031	6.770
OXFL-627	1/8/17	T-54	0.007	0.001	0.006	0.001	0.088
MSA-169	1/10/17	Cub-4 Sump	0.021	0.005	0.282	0.067	0.401
MSA-171A	1/12/17	76OX4A	0.017	0.004	0.231	0.055	0.334
MSA-171B	1/12/17	76OX4B	0.009	0.002	0.125	0.030	0.179
MSA-171C	1/12/17	76OX4C	0.013	0.003	0.172	0.041	0.247
OXFL-628A	1/11/17	T-40	0.017	0.004	0.138	0.033	0.158
OXFL-628B	1/11/17	T-40	0.018	0.004	0.138	0.033	0.158

* Values are *not* included for outlier results or results below limits of detection. Zero values indicate rounding *down*, nominally from the next digit (i.e., 0.0001 < value < 0.0005 would be rounded down to 0.000).

† OHDP-755A&B represents total product for all four 76OX runs

13. CAMPAIGN 76 SUMMARY

Table 21 summarizes runs in the context of the involved processes and, overall, follows the order of processing described in the preceding sections. Key product recoveries are shown in bold font, and intermediate products (typically feeds for another process) are shown in red font. For simplicity, rework streams are not delineated.

Table 21. Summary of principal product recoveries* for C76

Sample Code	Process Component	Sample Date	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk (mg)	^{253}Es (µg)
C76 Plan †	C76 Input		**3.9**	**0.8**	**46.5**	**10.0**	**110**	**24**	
C76 Plan †	Recovery		2		35		80	15	
DSDP-560	DS Product	2/28/15	0.70	0.08	40.8	9.50	90	22	
CLAF-890	**CL Feed**	**3/5/15**	**3.25**	**0.71**	**44.9**	**10.6**	**102**		**106**
CLCP-424	CL Product	3/17/15	3.26	0.71	48.7	11.5	109		58.4
CMAF-172	CM Feed	3/20/15	3.05	0.66	42.8	10.1	91		50
CMAP-303, 304,305	CM Am/Cm Products	3/31/15–4/15/15	3.36	0.73	41.2	9.7	1.2	0.2	
CMBP-154	CM Trans-Cm Product	4/15/15	0.07	0.02	0.2	0.04	77	17.4	21
CMBP-155	CX Feed	4/23/15			0.03	0.01	85		17
CXBK-312, 313,315,316	CM Bk Products BX Feed	4/29/15–4/30/15					0.4	16.8	0.03
BXBK-081	BX Bk Product	5/12/15						**16.1**	
CXCF-808	CX Cf Product / Nut Pkg feed	5/5/15			0.02		84		2
N. Assays	**Nut Pkgs D-101 to D-104**	**5/19/15–5/27/15**					**86**		
CMAP-307	Am/Cm Composite OH Feed	5/18/15	3.78	0.82	45.0	10.6	0.1		
CMAP-308	OH Product	6/27/15	**3.58**	**0.78**	**44.1**	**10.4**	0.2		
CLCP-434	Rework Product	12/29/15	0.9	0.15	4.7	1.3	11		
OXDP-755	OX Product	1/7/17	**0.6**	**0.12**	**4.3**	**1.0**	**7**		

* Average or median values are shown for the indicated sample groups—at lower precisions than indicated in sections describing individual runs—mainly to track **principal products of Campaign 76 (in bold font)**, and do not include waste materials (i.e., plutonium, dissolved structural metals, lanthanides, etc.). Values shown in red font are intermediate products, feeds or recycles (e.g., outputs of one process that feed another). Short-lived isotopes (e.g., ^{249}Bk and ^{252}Cf) have been decayed to dates when the samples were acquired. Values are *not* included for outlier results, for results below limits of detection, or for constituents with no related analyses.

† C76 Plan values (from Appendix A) are a combination of calculated target contents (based on target loadings and irradiation conditions) and measured rework material from Campaign 75 (chemical processing recycle [CY11T23CL-1], and target scrap [35 pellets]). The value for Am is the sum of ^{241}Am and ^{243}Am in the plan.

Table 22 contains a series of radiochemical analysis results and predictions made from ORIGEN or TCOMP codes of key nuclide production during HFIR irradiation. "Reworks" added to these predictions are from analyzed values. Values containing model predictions are shaded blue in the table, and values representing analytical results from the indicated set of 2 or 3 samples are left unshaded for contrast purposes. Predicted values for ^{249}Cf include contributions from decay of ^{249}Bk. Blanks (unshaded) for ^{249}Bk sample results indicate that no analysis was obtained for that set of samples.

The first Origen/TC predictions (also designated as "O/TC" in later table rows) include only calculated nuclide production from HFIR irradiation of targets being recovered in C76. The next entry "O/TC + reworks in 76DS-2" includes the aforementioned irradiation predictions plus 67 unirradiated reject pellets and reject Am/Cm powder left from Campaign 75 operations which had been added to the dissolver in 75DS-2. Two additional shaded entries appear for "TC+76DS-2 + Reworks into 76CL-1" and includes reworks from the conclusion of C75 (CY11T23CL-1). Each of these entries have been decayed to dates of associated analytical results representing that material—3/5/2015 for the 76CL-1 feed sample and 3/17/2015 for the product.

A brief discussion of the predictions contrasted with comparable analyzed values – with some context from the C76 Plan – follow the table. This discussion focuses on the main products of the campaign: ^{252}Cf and Cm which replenishes Cm consumed in producing ^{252}Cf and serves as feed for fabricating new targets for irradiation and production of ^{252}Cf.

Table 22. Model predictions (blue background) and final sample (white background) results

Analysis Method*	Decay Date	^{244}Cm (g)	^{245}Cm (g)	^{246}Cm (g)	^{247}Cm (g)	^{248}Cm (g)	^{Total}Cm (g)	^{249}Cf (mg)	^{250}Cf (mg)	^{251}Cf (mg)	^{252}Cf (mg)	^{249}Bk (mg)
Origen/TC Targets	3/2/15	6.691	0.063	20.459	0.549	4.607	32.493	7.814	23.128	6.337	99.060	26.890
O/TC+ reworks in 76DS-2	3/2/15	9.803	0.151	28.388	0.771	6.344	45.584	7.814	23.128	6.337	99.060	26.890
76DS-2 Product (DSDP-560)	3/1/15	9.497	0.171	24.823	0.682	5.558	40.836	12.449	23.252	6.745	90.635	15.513
O/TC+76DS-2 + Reworks from CY11T23CL-1	3/5/15	11.262	0.219	31.617	0.863	7.098	51.196	11.306	28.273	8.076	111.399	26.730
76CL-1 Feed (CLAF-890)	3/5/15	10.591	0.234	27.087	0.748	6.112	44.877	19.414	27.652	8.310	101.790	3.410
O/TC+76DS-2 + Reworks from CY11T23CL-1	3/17/17	11.246	0.219	31.589	0.862	7.101	51.017	11.944	28.264	8.076	111.293	26.092
76CL-1 Product (CLCP-424)	3/17/15	11.502	0.254	29.416	0.812	6.638	48.736	20.994	29.852	8.987	109.137	
76CM-1 Feed (CMAF-172)	3/20/15	10.091	0.222	25.807	0.713	5.823	42.757	17.565	24.965	7.519	91.115	
C76 Reworks CY15T23CL-1 CLCP-434	12/29/15	1.296	0.048	2.662	0.074	0.598	4.681	2.486	3.396	1.065	10.533	
C76 Totals†		11.387	0.270	28.469	0.787	6.421	47.438	20.051	28.361	8.584	101.648	

* Origen is the Oak Ridge Isotopic Generation and Depletion Code [5], and TCOMP (or TC) is the Transmutation Computation Code[6], [7]. Modeled results take advantage of Origen's strength in tracking nuclide branching ratios and decay while also incorporating TCOMP's parameterized neutron absorption cross sections for irradiation of production targets.

† C76 Totals are the sum of 76CM-1 Feed and C76 Reworks recovered in CY15T23CL-1 from analytical results.

It was predicted that 99 mg of ^{252}Cf would be produced in irradiating the targets. Another 12 mg was added from 75CL-1 reworks; no ^{252}Cf contribution was indicated or expected from unirradiated C75 reject pellets and powder. Of the 111 mg ^{252}Cf predicted in the calculations, essentially all of it was measured as product in 76CL-1 (sample CLCP-424 in Table 22). Process losses (mostly to rework) soon become evident as the Cf moves through the campaign. Eventually, 86 mg ^{252}Cf was packaged as recoverable product which was slightly better than the 80 mg ^{252}Cf expected as product in the C76 Plan (Table 21).

The contrast between predictions with analyzed values for curium is discussed generally in terms of total curium since the isotopic values tend follow the total; deviations from this tendency are noted. The models predicted production of about 32.5 g of Cm with another 13 g added to the 76DS-2 run in the form of unirradiated reject pellets and powder from C75. Sample analyses indicated that the predictions overestimated production regularly by 5% to 14% in Table 22 which leveled out at ~44 g (see Table 21) by the end of the campaign which was consistent with the C76 Plan. It is notable that ^{245}Cm was *under*estimated in the predictions shown in Table 22 by comparable margins (5% to 16%).The next to last row in Table 22 shows radioanalytical results from processing all the rework solutions collected during C76—through the 76OH runs—that underwent a full Cleanex processing in CY15T23CL-1. The last row in Table 22, "C76 Totals" are the sum of analytical results for 76CM-1 Feed and C76 Reworks recovered in CY15T23CL-1, essentially capturing all materials recovered during C76.

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APPENDIX A. CAMPAIGN 76 PLAN

Date: January 13, 2015

To: Distribution

From: D. E. Benker D. Benker

Subject: **Plans for Campaign 76: Processing of Five AmCm Targets plus Rework Materials**

A group of five targets were discharged from the HFIR after cycle 456 on November 2, 2014. The targets were transferred to Building 7920 on January 7, 2015 and Campaign 76 is scheduled to start on February 16, 2015. Table 1 lists the materials estimated in the targets plus the rework materials that will be processed in Campaign 76. A tentative schedule for the various processing steps is shown in Attachment 1. The time scheduled for the various steps includes some contingency to account for some minor processing problems, but in general it assumes that there are no major equipment failures, interferences from other program work, or administrative impediments to operations. Detailed instructions for the campaign operations are given in Attachment 2. The goal is to complete the processing steps in a timely manner to replenish the californium stock for the Californium Production Program and allow the quick resumption of other scheduled work. The Bk-249 will be separated from the californium to minimize the dilution of the Cf-252 isotope from the in-growth of the Cf-249 decay daughter. If there is a suitable customer the Bk-249 will purified and transferred to the alpha laboratories for final purification and shipment, otherwise the Bk-249/Cf-249 mixture will be discarded. As final products, we hope to recover about, 2 g of americium, 35 g of curium, 15 mg of Bk-249, and 80 mg of ^{252}Cf. Plans for preparing a new batch of americium-curium targets for irradiation will be formalized at a later date.

Table 1. Feed Material for Campaign 76, adjusted to February 2, 2015

Item	^{131}I, Ci	^{241}Am, g	^{243}Am, g	Cm, g	^{244}Cm, g	Ln g	^{249}Bk, mg	^{252}Cf, mg	^{254}Es, µg
Irradiated Targets[a]	0.4[b]		1.0	34.2	7.1	1	24	97	2
Rework[c]		0.8	2.1	12.3	2.9	25		13	
Total		0.8	3.1	46.5	10.0	26	24	110	2

[a] S-42, S-43, S-44 S-45, and S-46.

[b] Includes 0.03 Ci accumulated from ^{252}Cf fissions plus 0.34 Ci left from the fissions during the irradiation. It is estimated that ~0.002 Ci will be released per day from ^{252}Cf during the chemical processing until the ^{252}Cf is encapsulated for transfer to Building 7930.

[c] Rework includes, Campaign 75 chemical processing recycle (CY11T23CL-1), and Campaign 75 target scrap (35 pellets).

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Attachment 1: Schedule for Campaign 76

Date	Process	Purpose
02/16/15 to 02/21/15	Target Dissolution (76DS-2)	Separate the actinides from the Al target material and place in nitric acid solution
02/21/15 to 03/02/15	Cleanex Batch Solvent Extractions (76CL-1)	Remove chemical impurities and in-grown plutonium
03/02/15 to 03/13/15	LiCl Anion Exchange (76CM-1, 2, and 3)	Separate AmCm, transcurium elements, and residual lanthanides
03/13/15 to 03/16/15	LiOH Precipitation (76CM-3)	Convert transcurium from LiCl solution to small volume of dilute HNO_3 solution
03/16/15 to 03/23/15	High Pressure Ion Exchange (76CX-1 and 2)	Separate Cf from residual actinides
03/23/15 to 03/30/15	Berkex (76BX-1)[a]	Remove residual Cf so that the Bk product can go to the alpha laboratories unshielded.
03/30/15 to 04/06/15	Cf Package Loading (76CF-1, 2, and 3)	Load Cf onto storage packages suitable for transfer to Building 7930
03/23/15 to 04/09/15	Cf Rework Consolidation	Decontaminate equipment and consolidate various rework solutions for recovery in Cleanex stage (T-23).
04/09/15 to 04/16/15	Hydroxide Precipitation (76OH-1, and 2)	Transfer AmCm product from LiCl solution to HNO_3 solution for storage.
04/16/15 to 05/06/15	Cleanex Batch Solvent Extraction (F115 & T23)	Recovery of trace actinides recovered in secondary and tertiary processing of low-level effluent streams.

[a] deleted if no customer is identified.

Attachment 2: Description of Processing Steps for Campaign 76

Dissolution of Scrap aluminum (76DS-1)

The coolant flow-shrouds from the five targets will be dissolved in NaOH–$NaNO_3$ solution and the dissolvent disposed of to F-126. This step is done primarily to test the dissolution system is working properly, but it also provides a convenient means for the disposal of the highly-activated aluminum shrouds. The scrap aluminum will be charged to T-70 along with 7.7 L of 2.1 *M* $NaNO_3$. The dissolver will be heated to 92°C and 3.2 L of 10 *M* NaOH will be metered to the dissolver (<2.5 L/h) to effect the dissolution of the aluminum. After the required NaOH is metered into the dissolver, the temperature will be increased to ~104°C for a 30 min digest period (the digest period may be extended if 104°C cannot be attained) to ensure completed dissolution of the aluminum. The solution will be sampled and disposed of to F-126, and T-70 will be flushed with 2 × 10 L of 2 *M* NaOH and 3 × 10 L water via J126A06. (Note: T-737 may be substituted for J126A06 for these transfers. However, do not use T-734 as it is made of tantalum and strong caustic is corrosive to tantalum).

Dissolution of the target aluminum and actinide oxides in the irradiated targets and target fabrication recycle (76DS-2)

After the scrap aluminum dissolution is completed, 13.2 L of 2.1 *M* $NaNO_3$ will be added to T-70 and the five irradiated targets will be charged for the caustic dissolution. In addition to the irradiated targets reject Al-Cm pellets from Campaign 75 will also be added. The dissolver solution will be heated to 92°C and 5.6 L of 10 *M* NaOH will be metered to T-70 to control the aluminum dissolution process. When all the caustic has been added to T-70, the temperature will be increased to ~104°C and held for a 30 min digest period to ensure completed dissolution of the aluminum (the digest period may be extended if 104°C is not attainable). After cooling to <60°C, the aluminum-bearing solution will be vacuum transferred via T-737 through F-70 to F-111 and the undissolved actinide will be washed with 3 × 5 L of 2 *M* NaOH and 3 × 5 L of water leaving a 3-4 L heel in T-70 to cover the actinides and facilitate decay heat removal. The F-111 solution will be sampled for activities. If the activities are within the disposal limits, then it may be discarded to F-126 when convenient.

After completing the forward flushes to F-111, the acid dissolution of the oxides will begin by back flushing F-70 to T-70 with 3 × 2.5 L of conc. HNO_3 followed with 3 × 1.5 L of water to give a solution of ~15 L of 8.0 *M* HNO_3 in T-70. This solution will be simmered at 103°C for three hours (or longer if 103°C is not attainable) while being air sparged to dissolve the actinide oxides. After cooling, the dissolver solution will be transferred through F-720 to T-72 and combined with Campaign 75 rework materials. T-70 and F-70 will be flushed to T-72 with 2 × 3 L 4.0 *M* HNO_3, 2 × 3 L 1.0 *M* HNO_3, and 2 × 3 L water. Each of the water flushes will be sampled in T-70. After completing the flushes of T-70, F-720 will be flushed directly to T-72 with 2 × 5 L 1.0 *M* HNO_3 and to T-66 (or T-79) with 9 × 5 L of 1.0 *M* HNO_3 and 1 × 5 L water.

Purification of the transplutonium elements using batch solvent extraction (76CL-1)

The composite HNO_3 solution from the oxide dissolution of the irradiated targets and rework solutions will be adjusted in T-72 to feed conditions for the mainline Cleanex extraction. After sampling the composite solution at high volume to confirm the material balance, the solution will be boiled to 130°C or 3-4% above the chart zero. When this point is reached, a metered water addition (1 L/h) will be added to T-72 and the boiling continued until 10 L of water have been added. T-72 will be cooled and 1.25 L of conc. HCl will be added and the solution heated to boiling or 120°C. After cooling, the T-72 solution will be diluted to ~14 L with water (or sufficient volume to cover the sp gr probes) and sampled for acid. If the total H^+ is >30 moles, the evaporation sequence will be repeated. If the total H^+ is <30 moles, the solution will be adjusted to 0.25 *M* H^+ and sodium hypochlorite (2.5 L) will added and the solution digested at 80°C for 30 minutes.

The adjusted feed solution in T-72 will be contacted with 30 L of 1.0 *M* HDEHP in NPH diluent. The aqueous phase acidity will be adjusted stepwise with 5.0 *M* NaOH to ~0.03 *N* to extract the actinide elements. When the total gross alpha counting results show that <0.15-g ^{244}Cm (1-2% of the ^{244}Cm in the feed) remains in the aqueous raffinate, the raffinate solution will be transferred to T-23 for a subsequent second-stage contact. The loaded organic phase will be scrubbed with 2 × 10 L of 0.03 *N* HCl and the scrubs will be sampled and combined with the aqueous raffinate in T-23.

The first stage organic phase in T-72 will be adjusted to 0.2 *M* Adogen to hold the iron during the stripping operations. The actinides will be stripped with 6.0 *M* HCl—0.5 *M* H_2O_2 to T-604 for a diluent wash stage and then on to T-43. Hydrogen peroxide will be added to the strip solution to reduce the berkelium to the trivalent state so that it will strip with the other actinides.

Separation of lanthanides, americium-curium, and transcurium elements using LiCl chromatographic anion exchange (76CM-1, 2, and 3)

The separation of the lanthanides, americium-curium, and transcurium elements will be done using a chromatographic anion exchange process and the specialized equipment on the Cubicle 6 right rack. The feed will be adjusted to a highly concentrated salt solution of LiCl which will promote the formation of anion complexes of the lanthanide and actinide elements. Since the complexing strength varies for each element, they will move through the column at different rates and a chromatographic separation can be achieved by using appropriate eluants. Two primary runs will be made to prepare a lanthanide stream free of actinides that can be disposed of to waste, and an americium-curium product that is free of both lanthanides and transcurium elements. The transcurium products from the primary runs will be recycled to a third run to further purify the transcurium elements from both lanthanides and americium-curium.

When preparing the highly concentrated salt solution for the feed, small amounts of such impurities as aluminum, zirconium, or sodium will form insoluble compounds. In order to prevent these solids from plugging the equipment the feed will be adjusted to the run conditions and pre-filtered before making the final feed adjustment for the actual run. The initial feed adjustment will be done in T-43 and includes (1) evaporation to a low volume, (2) addition of ~20 moles of stock LiCl solution, (3) evaporation to 142.5°C, and (4) addition of enough conc. HCl to make the solution ~12.0 *M* LiCl–1 *M* HCl. The solution will be filtered from T-43 to T-65 via the floor filter, and T-43 will be flushed to T-65 with 4 × 750 mL synthetic feed solution (12 *M* LiCl–1 *M* HCl). The feed adjustment in T-65 for the ion exchange run is similar. The solution will be boiled to 142.5°C and cooled, conc. HCl will be added to make the solution 1.0 *M* HCl, and the solution will be heated to 120°C to dissolve any hydrolyzed zirconium and lower the acid concentration to ~0.1 *M* HCl. The final volume will be about 5 L.

The ion exchange column (C-636) will be filled with 1.4 L of HCl treated Dowex 1X10 (or X8) resin and approximately one half of the feed will be used for each of the two primary runs. The adjusted feed solution is pumped through the column to load the actinide elements and four eluant solutions will be used, (1) 10 *M* LiCl to remove residual lanthanides, (2) 9 *M* LiCl (3-4 L) to elute americium and curium, (3) 8 *M* HCL–0.1 *M* H_2O_2 (2-3 L) to elute the transcurium elements, and (4) 0.8 *M* HCl (4 L) to elute impurities. All the eluants will typically be added in 0.5 L batches and the eluants with LiCl will also contain 0.1 *M* HCl–0.1 *M* hydroxylamine hydrochloride and 2.5 vol. % methyl alcohol. The progress of the elution will be monitored using the in-cell neutron and alpha probes to determine when to switch the column effluent to the various collection tanks. In the primary runs, the cuts will be made to ensure there are no actinides in the raffinate and no transcurium elements in the americium-curium fraction. The loading raffinate and the portion of the 10 *M* LiCl eluant estimated to be extremely low of actinides will be collected in T-61. Just prior to the elution of the americium and curium, one or two fractions will be collected in T-630 and T-631. The americium and curium will be collected in T-633 (or T-634), the transcurium (plus a little americium-curium) will be collected in T-632, and the column flush will be collected in T-72. The americium-curium products will be composited in T-40, and the pre-curium cuts will be evaluated for disposal, rework, or product as necessary to maintain freeboard for the subsequent runs.

The combined transcurium fractions from T-632 along with any other recycle fractions will be transferred to T-65 and combined with the remaining tank heel to prepare feed for the recycle run. The volume for this final run should be ~3 L. The resin in C-636 will also be changed. The elution for the recycle run and the elution goals are essentially the same as for the primary runs with the exception that the HCl concentration of the transcurium eluant is lowered from 8 to 1 *M*. All of the feed solution will be pumped through the column for loading and the raffinate and lanthanide eluants will be collected in T-61. The pre-curium cut will be collected in T-630 and the americium-curium will be eluted to T-631. The transcurium product will be eluted to T-632 and the column will be flushed back to T-65 with 8.0 L of 0.8 *M* HCl for a heel check.

The 76CM-3 transcurium product will be converted to the nitrate form and adjusted to the required volume and acidity using a LiOH precipitation technique. This involves (1) adding 200-mg of iron (~1-g ferric chloride) as a precipitate carrier, (2) precipitating the actinide hydroxides with 1.0 L of 5 *M* LiOH, (3) aging the precipitate for 2-h, and (4) filtering through a glass frit to collect the precipitate. The precipitate will then be washed with 2 × 200 mL water to the filtrate collection tank, and finally dissolved with strong nitric acid (14 mL of conc. HNO_3) and a dilute nitric acid wash (100 mL of 0.2 *M* HNO_3) to a glass collection bottle. The dissolution sequence will then be repeated using a second nitric acid addition (14 mL of 8.0 *M* HNO_3) and a final 3 × 150 mL water flush to the product bottle. The final volume should be approximately 0.8 L at 0.3 *M* HNO_3. If a significant amount of the transcurium elements (>5%) remain on the filter, a second dissolution of the filter solids will be programmed and another transcurium product batch will be collected. The product collection bottle will be put in a bottle carrier and be transferred to Cub 4 for the start of the cation ion exchange (CX) runs.

Separation of actinides using AHIB (α-hydroxyisobutyrate) cation exchange (76CX-1 and 2)
The combined transcurium element product from the 76CM runs which has been precipitated and redissolved in HNO_3 will be further purified and the transcurium elements separated in the AHIB cation exchange runs. These purification-separation runs will use the chromatographic cation exchange equipment on the right rack of Cub 4. The actinides will be first loaded on the short column (C-402) containing Dowex 50W-X8 resin. The separation of the elements will be accomplished by pumping a complexing agent, ammonium alpha-hydroxyisobutyrate (AHIB), of various concentration and pH through the columns. The complexing strength for the elements are different, so they will move through the system at different rates which allows the elements to eventually form distinct bands on the larger elution column (C-401), which is also filled with Dowex 50W-X8 resin. The eluant solutions will be collected into small bottles and analyzed to determination which fractions contain Cf, Bk, rework, or

waste. The resin in the loading column (C-402) will be replaced after each run; however, the resin in the elution column (C-401) will be used for all two runs before it is discarded.

For the first run, both the loading and elution columns will be filled with fresh resin and both columns will be washed with 1.0 L of 0.3 *M* NH_4NO_3 and 600 mL water to convert the resin from the acid form to the ammonium form. The loading column is unheated, but the elution column will be heated to 70°C for the run. About 60% of the initial feed solution will be transferred in batches to the vacuum-pressure transfer pot and the pot will be pressurized with nitrogen to 45 to 50 psig to yield a flow rate of 1.5 to 2.0 L/h through the loading column. After the feed is loaded, the column is washed with 80 mL water, 300 ml 0.3 *M* NH_4NO_3, and 80 mL water. The loading column effluent is then routed to the elution column and the high pressure pump is used to transfer the eluants through both columns at ~1 L/h. The first eluants, 0.25 *M* AHIB-pH 4.2, will slowly move the actinides from the loading column and onto the longer elution column where the actinides are slowly separated into distinct bands. In a typical run, about 1.5 L to 2.0 of 0.25 *M* AHIB-pH 4.2 is used to elute the californium, ~700 mL of 0.25 *M* AHIB-pH 4.6 is used to elute the berkelium, and 450 mL of 0.50 *M* AHIB-pH 4.8 is used to elute any remaining americium-curium. After the elutions, 450 mL of water is used to flush the system free of any remaining reagent solutions. Approximately 10-12 cuts of varying volume will be collected during the run. Both the n_f probe and an inline alpha detector will be used to determine when to route the effluent to a separate bottle. All the bottles will be acidified and sampled for analysis to determine disposition of the solution, i.e., Cf and Bk product fractions or recycle.

The remaining 40% of the feed plus recycle fractions will be used in the second and final run. The run conditions and product handling will be similar to the first run. Depending on the quality of the Cf/Bk separation, a third recycle run may be added.

Berkelium purification using batch solvent extraction (76BX-1)

If a customer is found for the berkelium, the product fractions from 76CX runs will be further purified from californium using Berkex batch solvent extraction and the equipment on the Cub 4 right rack. To prepare the feed solution for Berkex, the berkelium fractions from the CX runs are composited and the berkelium is concentrated and purified from AHIB solution using ion exchange. The composited berkelium solution is pressured through the small loading column (C402) to load the berkelium on the resin, the resin is washed with 2 × 100 mL of water to remove residual AHIB reagent, and the berkelium is removed with 500 mL of 8 *M* HNO_3, which is the desired feed solution for Berkex.

After about a one week digestion period, berkelium feed solution will be made 0.30 *M* $NaBrO_3$ to oxidize Bk(III) to Bk(IV), which will then be extracted into 500 mL of 0.5 *M* HDEHP in dodecane in T-411. After a 30 min sparge period, the organic will be scrubbed with 2 × 590 mL 8.0 *M* HNO_3–0.3 *M* $NaBrO_3$. Finally the organic will be stripped with 2 × 280 mL 8.0 *M* HNO_3–1 *M* H_2O_2. If a sufficient decontamination is acquired in one cycle, the product will be transferred to a laboratory glove box for final purification and packaging for shipment. If not, additional runs may be made to achieve the necessary purification.

Loading Cf onto storage/transfer packages (76Cf-1 through 3)

The californium fractions from the CX runs will be composited and prepared for loading on cation exchange resin packages. The packages consist of a Pt cylinder with fritted ends containing cation exchange resin. The Pt cylinders are placed in a Swagelok fitting assembly and loaded with 40-45 mg ^{252}Cf. The loaded packages are held for 24 h and then are fired at 150°C for 1 h, 350°C for 1 h, and

700°C for 4 h. The fired packages will be placed in the Swagelok nut package for later transfer via the pneumatic transfer line to Building 7930 for further processing.

Removal of LiCl from the americium-curium product (76OH-1 and 2)

The americium-curium product from the LiCl anion exchange runs will contain from 80 to 100 moles of LiCl. The large amount of salt in this solution must be removed before the americium-curium product can be properly concentrated and stored. The technique that has been used in the past and described below involves precipitating and filtering the actinides and redissolving in clean nitric acid. An alternate step would involve diluting the salt content and using several batch Cleanex extractions in a large tank to extract the actinides from the salt and then back extract the actinides in strong nitric acid.

The americium-curium collection in T-40 will be evaporated to a low volume (~20 L) and one-half of the volume will be transferred to T-47 and flushed with 2 × 2.5 L of water. Enough 10 M NaOH (CP Grade) will be slowly added (~100 mL/min) to T-47 (while sparging) to neutralize the acid, precipitate the actinides, and make a final solution that 2 to 3 N OH^- in T-47. After 30 min of additional sparging and a >4 h settling period, the filtrate will be transferred through the deep-bed filter to T-20. It may be necessary to use both T-444 and T-430 as pressure and vacuum transfer tanks to move the solution through the filter. The bulk of the precipitated actinides should remain in T-47. This operation requires some technique on the part of the technicians to filter the solution through the deep bed filter and not disturb the filter bed. The precipitate in T-47 will be washed with 2 × 4.5 L of 2.0 M NaOH and 3 × 4.0 L of water. Each wash will be added to T-47, sparged 30 min, settled 30 min and transferred via the deep bed filter to T-20.

The discharge of the deep bed filter will be changed to route the flow to T-54 for collection of the acid dissolution product. The actinides will be dissolved by the slow addition of 4 L of 8 M HNO_3 plus 2×0.5 L water flushes to T-47. After 1 h of sparging, the acid dissolution product will be transferred through the deep bed filter to T-54. T-47 will be flushed with 3×2.5L of 1 M HNO_3 and 3×2.0 L water to T-54. T-54 will have to be boiled periodically to keep the volume <17. T-20 will be acidified, sampled, and (if confirmed low in actinides) disposed of in preparation for the second precipitation run. The remaining feed in T-40 will be transferred and flushed to T-47 and the precipitation, filtering, and washing sequence will be repeated collecting the dissolved americium-curium product in T-54. The acid product in T-54 will then be concentrated for transfer to a storage tank.

If the deep bed filter is filled with precipitate before the flushing is completed, the inlet line will be back flushed to T-47, and the filter will be flushed to T-20 with caustic and water. The filter discharge will be routed to T-54 and the precipitate will be dissolved with acid to T-54. The routes will then be reset to the original configuration and the filtration from T-47 to T-20 continued until all the filtrate and programmed flushes are completed.

Rework Collections (CY15F115CL-1 & CY15T23CL-1)

During the campaign, various rework solutions will be collected and processed in T-23 and F-115 Cleanex extraction stages. The recovered actinides will be processed, cleaned up, and stored.

APPENDIX B. LIST OF SAMPLE CODES

[Note that some listed codes may not be appear in this report.]

REDC Processing Sample Code List

Sample Code	Description	Aq/Org
AC	Analytical Consolidation Aqueous	Aqueous
ACO	Analytical Consolidation Organic	Organic
AF	A-Bank Adjusted Feed	Aqueous
AP	A-Bank Product	Organic
AS	A-Bank Scrub	Aqueous
AW	A-Bank Waste	Aqueous
AXAF	Anion Exchange Adjusted Feed	Aqueous
AXNP	Anion Exchange Neptunium Product	Aqueous
AXPU	Anion Exchange Plutonium Product	Aqueous
AXRF	Anion Exchange Raffinate (and col. Pretreat and wash)	Aqueous
BP	B-bank Product	Aqueous
BU	Originally B-Bank U product, now B-bank organic stream)	Organic
BX	B-bank back extractant, i.e, strip	Aqueous
BXBK	Berkex Berkelium Product	Aqueous
BXEX	Berkex Extractant	Aqueous
BXFL	Berkex Flush	Aqueous
BXRF	Berkex Raffinate	Aqueous
BXST	Berkex Strip	Aqueous
BXWO	Berkex Waste Organic	Organic
CLAF	Cleanex Adjusted Feed	Aqueous
CLCP	Cleanex Clean Product	Aqueous
CLER	Cleanex Extraction Raffinate	Aqueous
CLEX	Cleanex Extractant	Organic
CLSR	Cleanex Scrub Raffinate	Aqueous
CLST	Cleanex Strip	Aqueous
CLWO	Cleanex Waste Organic	Organic
CMAE	LiCl AIX Am/Cm Elutriant	Aqueous
CMAF	LiCl AIX Adjusted Feed	Aqueous
CMAP	LiCl AIX Am/Cm Product	Aqueous
CMBP	LiCl AIX Transcurium product	Aqueous
CMCE	LiCl AIX Californium Elutriant	Aqueous
CMCF	LiCl AIX Column Flush	Aqueous
CMCR	LiCl AIX Column Raffinate	Aqueous
CMPF	LiCl AIX Precipitation feed	Aqueous
CMP	Curium Product?	Solid
CMRE	LiCl AIX Rare Earth Elutriant	Aqueous
CMSF	LiCl AIX Synthetic Feed	Aqueous
COAF	Curium Oxide Adjusted Feed	Aqueous
CORF	Curium Oxide Raffinate	Aqueous
CP	C-Bank Product	Aqueous
CSA	Cold Sample Aqueous	Aqueous
CSO	Cold Sample Organic	Organic
CW	C-Bank Waste?	Organic
CX	C-Bank Back extractant, i.e., strip	Aqueous
CXAF	But Column Adjusted Feed	Aqueous

REDC Processing Sample Code List

CXBK	But Column Berkelium Product	Aqueous
CXCF	But Column Californium Product	Aqueous
CXCM	But Column Am/Cm Product	Aqueous
CXER	But Column Elution Raffinate	Aqueous
CXES	But Column Einsteinium Product	Aqueous
CXFM	But Column Fermium Product	Aqueous
CXLR	But Column Loading Raffinate	Aqueous
CXSF	But Column System Flush	Aqueous
DP	Dissolution Product? Primarily for T-555.	Aqueous?
DSDF	Dissolution Dissolver Flush primarily for T-70	Aqueous
DSDP	Dissolution Dissolver Product primarily for T-70	Aqueous
DSJD	Dissolution Jacket Dissolvent primarily for T-70	Aqueous
DSJW	Dissolution Jacket Waste primarily for T-70	Aqueous
HCA	Heel Check Aqueous	Aqueous
HCO	Heel Check Organic	Organic
HDP	HDEHP Concentration	Organic
MSA	Miscellaneous Sample Aqueous	Aqueous
MSO	Miscellaneous Sample Organic	Organic
NPPA	Neptunium Pa Removal Column Product	Aqueous
OHDP	Hydroxide Precipitation Dissolved Product	Aqueous
OHFL	Hydroxide Precipitation Flush	Aqueous
OHPF	Hydroxide Precipitation Precipitator Feed	Aqueous
OXDP	Oxalate Precipitation Dissolved Product	Aqueous
OXFL	Oxalate Precipitation Flush	Aqueous
OXPF	Oxalate Precipitation Precipitator Feed	Aqueous
PBMS	Plutonium Beryllium Miscellaneous Sample	Aqueous
PC	Process Condensate	Aqueous
POAF	Plutonium Oxide Column Adjusted Feed	Aqueous
POFL	Plutonium Oxide Column Flush	Aqueous
PORF	Plutonium Oxide Column Raffinate	Aqueous
PUAF	Pubex Adjusted Feed	Aqueous
PUCP	Pubex Curium Product	Aqueous
PUP	Plutonium Oxide Product	Solid
PUPP	Pubex Plutonium Product	Aqueous
PUWO	Pubex Waste Organic	Organic
PVC	Process Vacuum Condensate	Aqueous
REAE	BR-6 LiCl AIX Am/Cm Elutriant Obsolete	Aqueous
REAF	BR-6 LiCl AIX Adjusted Feed Obsolete	Aqueous
REAP	BR-6 LiCl AIX Am/Cm Product Obsolete	Aqueous
REBP	BR-6 LiCl AIX Transcurium Product Obsolete	Aqueous
RECE	BR-6 LiCl AIX Californium Elutriant Obsolete	Aqueous
RECF	BR-6 LiCl AIX Column Flush Obsolete	Aqueous
RECR	BR-6LiCl AIX Column Raffinate Obsolete	Aqueous
RERE	BR-6LiCl AIX Rare Earth Elutriant Obsolete	Aqueous
RESF	BR-6LiCl AIX Synthetic Feed Obsolete	Aqueous
SF	Synthetic Feed	Aqueous

REDC Processing Sample Code List

SFL	Off-Gas Scrubber Feed Liquor	Aqueous
SFW	Acidified Plant Waste	Aqueous
SLW	Off-Gas Scrubber Liquor Waste	Aqueous
SPW	Neutralized Plant Waste	Aqueous
STCM	Am/Cm Storage	Aqueous
SP	Stored plutonium product	Aqueous