ORNL/TM-2020/1839

Campaign 78—Production of ^{252}Cf and the Recovery of Curium Feed Material at the Radiochemical Engineering Development Center

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March 2021

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Californium-252 Program

CAMPAIGN 78—PRODUCTION OF ^{252}CF AND THE RECOVERY OF CURIUM FEED MATERIAL AT THE RADIOCHEMICAL ENGINEERING DEVELOPMENT CENTER

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March 2021

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1. BACKGROUND AND SUMMARY OF CAMPAIGN 78

1.1 BACKGROUND

The production of californium-252 (Cf-252) has been of interest to the US Department of Energy (DOE) since the 1970s because of its ability to emit neutrons (2.31×10^{12} neutrons/gram second) [1]. Today, Oak Ridge National Laboratory is responsible for the majority of Cf-252 sold around the world. This report aims to provide a complete summary of the chemical processing of the Cf and curium (Cm) from campaign 78 (C78) which started in January 2019.

1.2 FACILITIES

Today, most Cf-252 is produced by irradiating Cm targets in the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL). The Cm targets are placed in the flux trap of HFIR, and the Cm is transmutated into fission products and the transcurium actinides, berkelium (Bk), Cf, einsteinium (Es), and fermium (Fm). Because of the highly radioactive nature of the actinides and fission products that are produced in HFIR, all post-irradiation chemical processing manipulations are done in shielded hot cells at the Radiochemical Engineering and Development Center (REDC), which is conveniently located adjacent to HFIR. The Radiochemical Engineering and Development Center is a category II nuclear facility that hosts two buildings, 7920 and 7930, containing hot cells and associated radiochemical processing capabilities. Building 7920 is where the majority of the chemical processing for this process takes place and will be the focus of this report. Within 7920, there are four glovebox labs, three radioanalytical labs, three cold labs, two mini hot cells, and a bank of nine heavily shielded hot cells.

Connecting the cubicles (Cubs) is a conveyer system that allows for the transfer of materials and equipment from one Cub to another. Specially trained personnel handle the highly radioactive materials using a pair of mechanical manipulators in each Cub which is contained by an alpha seal window and a shielded window, consisting of several layers of leaded glass (for gamma shielding) and frames that are filled with mineral oil (for neutron shielding). This allows for the highly radioactive material to be handled with little to no dose to the worker. Figure 1 shows an illustration of the processing hot cells and accessory areas. Cubs 1 through 3 are used for fabrication of recycle Cm targets. Cubs 4-7 and Tank Pits 1-7 plus the Waste Tank Pit are used for chemical processing. Cub 8 is used for sample preparation and analysis, and Cub 9 is used for solid waste accumulation, monitoring and packaging. Chemical reagent tanks and process instrumentation are located above the hot cells in the chemical Makeup Area.

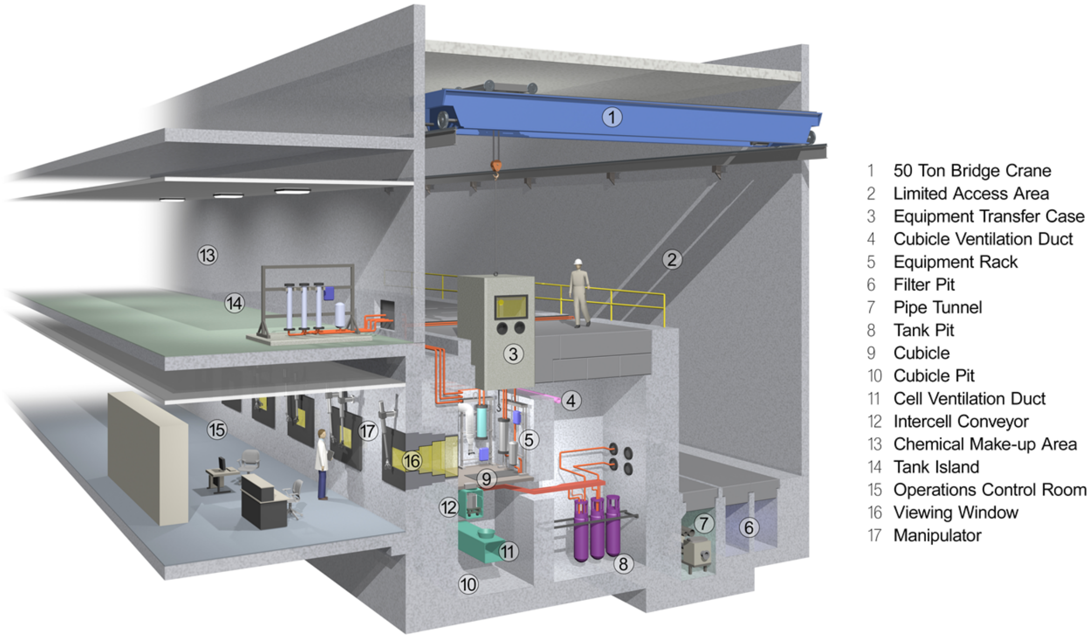


Figure 1. An illustration of the hot cell layout in the Radiochemical Engineering and Development Center Building 7920.

1.3 SUMMARY OF Campaign 78 (C78)

C78 started by irradiating four Cm targets (S-53, S-54, S-55, and S-57) in the flux trap of HFIR starting on May 1, 2018 and ending on September 28, 2018 for a total of four cycles (479–482). Table 1 shows the expected feed for C78 based on modeling calculations (Section 12 of this report) and measured rework content. After irradiation, the targets are left in the cooling pool to ensure short-lived ^{131}I (half-life = 8 d), has decayed to an acceptable limit. The targets were then transferred to Radiochemical Engineering and Development Center Building 7920 on December 27, 2018. Figure 2 shows a high-level flowchart of the processing operations that will be described throughout this report, as well as the naming conventions for each processing step. Chemical processing for C78 started on January 15, 2019, resulting in three ^{252}Cf nut packages containing 73.020 mg of ^{252}Cf. Table 2 summarizes the major feed and products of interest through this campaign. Other isotopes of interest that were produced in this campaign include 37.92 g of Cm microspheres, 13.8 mg of ^{249}Bk, microgram quantities of ^{253}Es and ^{254}Es, and nanogram quantities of ^{255}Es and ^{255}Fm.

Table 1. The estimated feed for C78 decay adjusted to February 1, 2017 and modified from the C78 plan found in Appendix A.

Item	^{131}I (Ci[a])	^{241}Am (g)	^{243}Am (g)	Cm (g)	^{244}Cm (g)	^{249}Bk (mg)	^{252}Cf (mg)	^{254}Es (µg)
C78 Targets[b]	0.2		0.3	24.0	6.5	17	97	2.2
Target Fabrication Rework[c]		0.1	0.3	5.0	1.6			
Campaign 77 Rework		0.2	0.9	4.9	1.5		7	
Total	0.2	0.3	1.5	33.9	9.6	17	114	2.2

[a] ~0.002 Ci is estimated to be generated per day from the fissions of ^{252}Cf until the ^{252}Cf is encapsulated for transfer to Building 7930.
[b] S-53, S-54, S-55, and S-57.
[c] 30 reject pellets

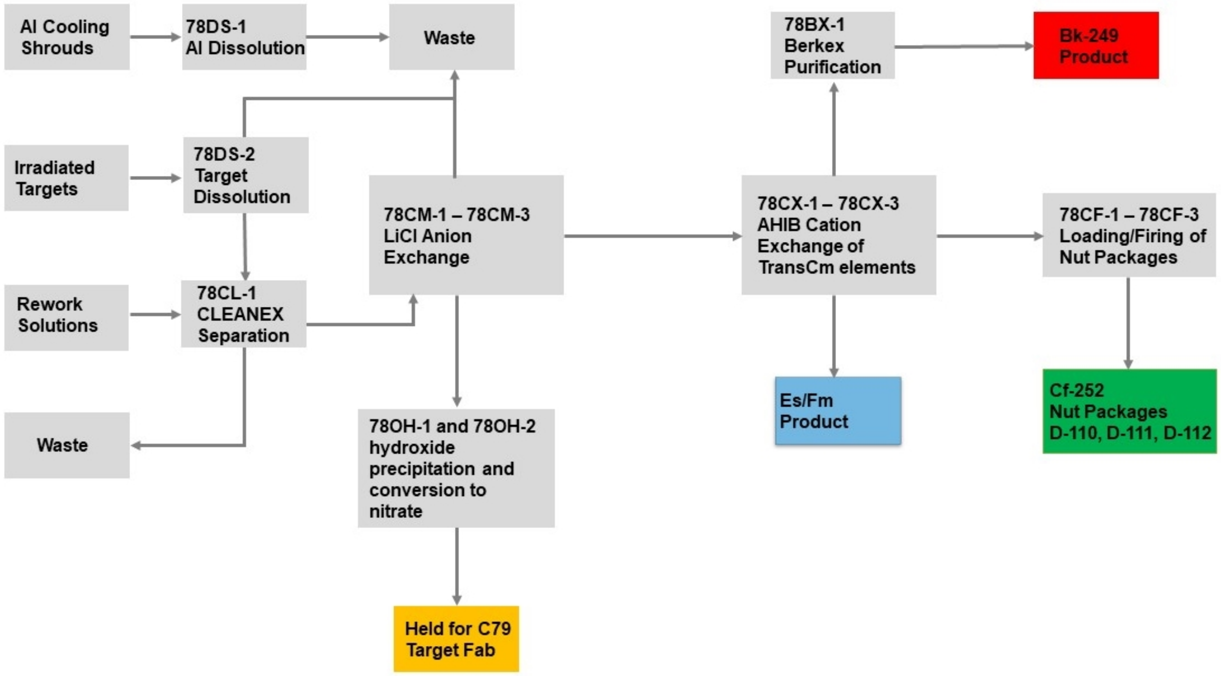


Figure 2. High-level flowchart of C78.

Table 2. Measured Feed, Products, Rework, and Waste of C78 Materials adjusted to February 1st, 2019.

Measured Process Material	Am (g)	^{243}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk (mg)	^{253}Es (µg)
Campaign Feed (Dissolver Products+Rework)	2.11	1.97	32.4	9.40	95.9		
AmCm Oxide Product	1.51	1.25	21.4	6.32			
Cf Nut Packages (D-110, D-111, & D-112)					76.2		
BX product						13.8	
Es Product							74.4
C78 Rework	1.46	1.21	7.23	2.13	9.41	13.8	

Blanks in the table refer to either values smaller than 1 × 10^{-5} or the sample was not analyzed for a specific isotope

2. ALUMINUM SHROUD AND TARGET DISSOLUTION (78DS)

Before chemical processing of the material can take place, the aluminum shroud and target body material need to be disposed of, which is done by dissolving the aluminum metal in a caustic solution containing sodium hydroxide (NaOH). In doing so, a soluble form of aluminum is formed ($Na_2Al_2(OH)_4$) and *f*-elements (and some fission products) form insoluble hydroxide compounds (i.e., $Cf(OH)_3$), which can be filtered out of the aluminum-containing solution [2].

2.1 ALUMINUM SHROUD DISSOLUTION [78DS-1]

January 15, 2019–January 19, 2019

Chemical makeup for aluminum shroud dissolution began on January 15 by preparing two batches of $NaNO_3$ (5.761 L of 2.25 M) in M-2 and one batch of NaOH (3.593 L of 10 M) in M-573. The shrouds were removed from the targets and stored until dissolution. Systems checks were performed starting January 17th. During calibration checks of the M-573 flow controller, it was noticed that the process valve would not stabilize. Operations were put on a temporary hold to address the issue. Once calibration tests were successfully complete, flushing the dissolution vessel (T-70) to F-111 was set up and tested with water.

On January 18, the dissolution of the aluminum shroud started by adding $NaNO_3$ (5.761 L, 2.25 M) to T-70 followed by the aluminum shroud. The mixture was then heated to roughly 92°C before NaOH (2.6 L, 10 M) was added slowly. Temperature stability was monitored and the addition of NaOH was to be stopped if temperature began to increase. After the NaOH was added, water was used to flush the process lines into T-70 vessel. The temperature was then increased to 103°C and was held for 30 min before it was cooled to 60°C. The solution in T-70 was then transferred to F-111 for disposal with 2 M NaOH and water flushes. Figure 3 illustrates the process used for DS-1.

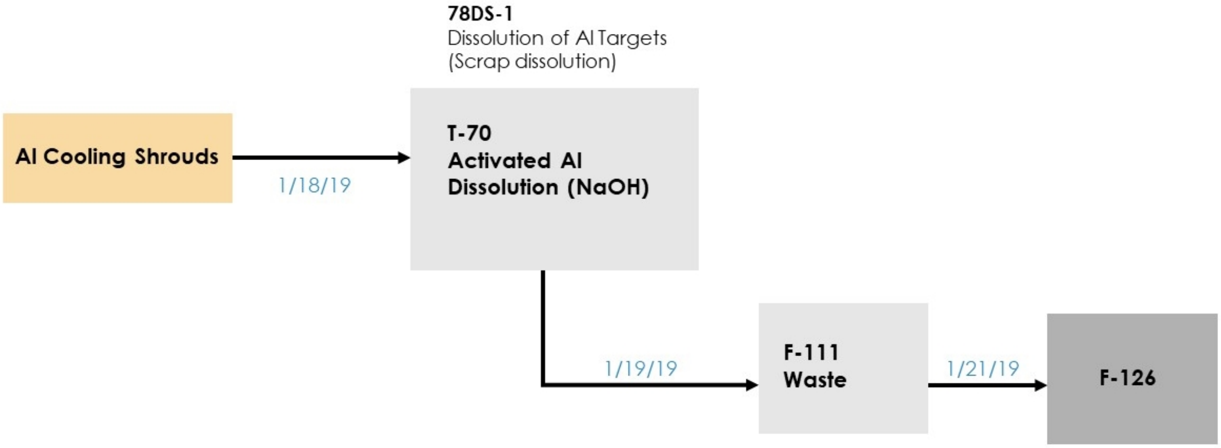


Figure 3. A flowchart showing a schematic of DS-1.

2.2 TARGET AND REJECT PELLET DISSOLUTION [78DS-2]

January 17, 2019–January 31, 2019

Chemical makeup and system checks for the target dissolution were performed from January 17 to 19. On January 20th, dissolution of the targets and reject pellets began by adding $NaNO_3$ (10.535 L, 2.25 M) to T-70 from M-2 followed by targets S-53, S-54, S-55, and S-57, and rework items such as pellets within pipe nipple 7 and container A, and powder within twist lock P and SS beaker. While attempting to heat T-70 to 92°C, the Eli-H79 (voltmeter) would not respond. The process was put on hold to address the issue. An electrician troubleshot the H-70B heating circuit and determined that the H-70B Variac failed. This was replaced and tested before successfully heating T-70 to 92°C. Sodium hydroxide (4.7 L, 10 M) was added slowly from M-573 while monitoring the temperature difference between the heating water in the dissolver jacket and the solution inside the dissolver tank until all NaOH was added.

This was done to control the rate of reaction by controlling the rate of NaOH addition and prevent a too-rapid reaction. After all the NaOH was added, water was used to flush the process lines into the T-70 vessel. The temperature was then increased to 103°C and held for 30 min before it was cooled to 60°C. The solution was then transferred to F-111 and rinsed three times with NaOH (2 M) followed by three rinses with water. Contents were sparged with air for 10 min and left to settle for 10 min before the Al-bearing solution was decanted through filter F-70 to F-111. The actinide and fission product oxides and Al-activation product, Si, were retained in T-70

On January 23, the actinide and fission product oxide solids were dissolved by three additions of concentrated HNO_3 (2.5 L, 15.7 M) followed by three water rinses (1.5 L each), bringing the total volume to 15.182 L. The solution was sparged with air while being heated to 103°C for 3 h before cooling to below 40°C. The solution was sparged for 30 min before an aliquot was taken for analytical analysis (DSDP-621A/B/C). The contents of T-70 were filtered to remove silica and any undissolved solids (UDS) and transferred to T-72 on January 25. The contents of T-72 were then evaporated down to roughly 30 L while back-flush and heel checks were performed on T-70 (analytical sample HCA-028, bottle #3120). T-72 continued to be evaporated down to roughly 30 L. Meanwhile, HNO_3 (2 × 5 L, 1 M) was used to flush the floor filter first to T-72 and then to T-66. T-72 was left with a volume of 30.9 L. Once T-72 reached 30 L, it was cooled before an additional floor filter rinse with HNO_3 (2 × 5 L, 1 M) from M-70 was added to T-72. The floor filter continued to get flushed with HNO_3 (9 × 2.5 L, 1 M) and water (2.5 L) to T-66. After sparging for 1 h, T-66 was then sampled as PVC-211. Assay results from samples taken during DS-2 are shown in Table 3 and a process flowchart is shown in Figure 4.

Table 3. Assay results taken from DS-2.

Sample code	Date	Tank	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk (mg)	^{137}Cs (Ci)	^{144}Ce (Ci)	^{154}Eu (Ci)
DSJW-427A	1/23/2019	F-111		0.35	0.047	0.069	0.005	0.047		7.71	0.952	0.0017
DSJW-427B	1/23/2019	F-111		0.29	0.039	0.068	0.005	0.047		7.71	0.862	0.0017
DSDP-621A	1/24/2019	T-70	0.517	0.35	0.047	26.2	7.40	91.7	16.2	3.52	146	1.13
DSDP-621B	1/24/2019	T-70	0.527	0.22	0.030	26.4	7.34	94.9	15.1	3.52	146	1.05
DSDP-621C	1/24/2019	T-70	0.525	0.52	0.071	26.6	7.42	93.0	16.1	3.60	150	1.17
DSDF-297	1/25/2019	T-70						0.001			0.003	0.0001
DSDF-298	1/25/2019	T-70						0.0003				
HCA-028	1/26/2019	T-70	0.014	0.04	0.006	0.023	0.006	0.14		0.017	0.215	0.0163
PVC-211	1/27/2019	T-66	0.179	0.06	0.008	0.001		0.004		0.002	0.002	0.0009

Blanks in the table refer to either values smaller than 1 × 10^{-5} or the sample was not analyzed for a specific isotope.

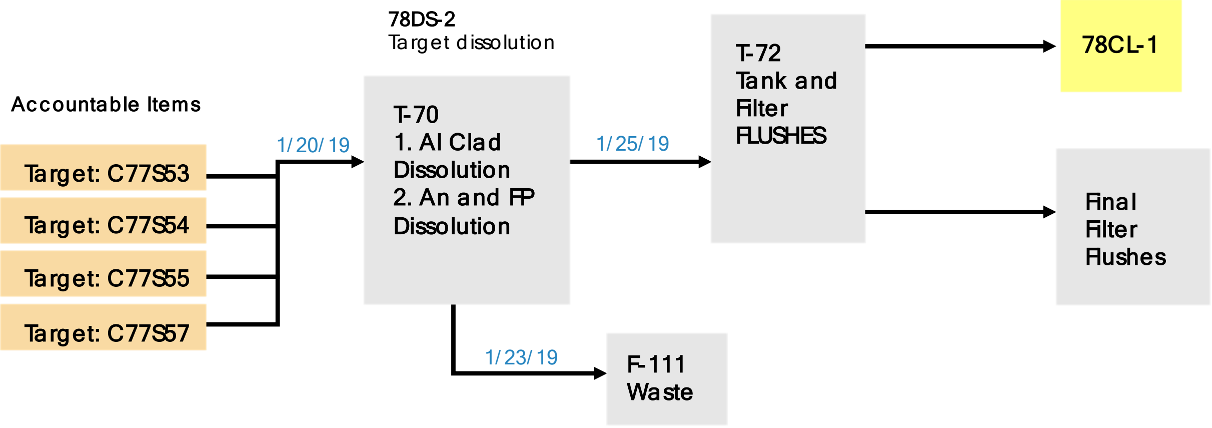


Figure 4. A flowchart showing a schematic of DS-2.

3. CLEANEX BATCH SOLVENT EXTRACTION FOR TRANSPLUTONIUM PURIFICATION [78CL]

The CLEANEX process is designed to separate trivalent actinide and lanthanide elements from other fission products and miscellaneous metals by extracting the An-Ln into HDEHP (di(2-ethylhexyl) orthophosphoric acid) in a bi-phasic extraction. HDEHP is dissolved in Exxsol D60 to make the Cleanex solvent 1M HDEHP [3]. This organic solution is less dense than and is immiscible in water. This allows for a physical separation of the more dense aqueous phase from less dense organic phase.

3.1 CLEANEX FEED ADJUSTMENT [78CL-1]

January 26, 2019–January 30, 2019

T-72 was sparged with air for 2 h before samples CLAF-898A, CLAF-898B, and CLAF-898C were taken for analysis (Table 4). An aliquot of the solution from T-72 (325 mL) was transferred to a clean bottle labeled “T-72 CLAF product C78.” for subsequent development of an improved separations process.

Makeup solution and system checks started on January 26th and feed adjustments started the next day. The feed solution was evaporated down for a total of three days. After evaporation, concentrated HCl was added to re-dissolve any precipitated solids such as zirconium molybdate, then diluted with water to a volume of ~10 L. The solution was sparged for 30 min while being heated to 90°C to reduce the HCl acidity and was subsequently cooled to room temperature. Once cool, additional water was added followed by sparging of the solution with air. A sample was taken to measure its acidity, which confirmed a normality of 1.7 (CLAF-899). To adjust the acidity within an acceptable range, NaOH was added to the reaction and an additional sample was taken (CLAF-900, Table 4). This sample was within an acceptable range (0.39 M). Concentrated NaOCl (2.5 L) and water (0.25 L) were added to the solution to oxidize all Mo species to Mo(VI). It is important to extract Mo(VI), as all unextracted Mo may hydrolyze in low acidity later in the Cleanex extraction, resulting in emulsions. The solution was sparged while heating and shut off once the temperature reached 70°C. Heating was continued without air sparging until the temperature reached 80°C, where it was held for 20 min before cooling.

Table 4. Assay results taken during feed adjustments of CL-1.

Sample code	Date	Tank	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-898A	1/27/2019	T-72	1.45	1.57	0.270	32.1	9.32	97.4	3.66	162	
CLAF-898B	1/27/2019	T-72	1.42	2.19	0.377	32.6	9.47	97.1	3.45	172	
CLAF-898C	1/27/2019	T-72	1.36	2.56	0.440	32.5	9.44	94.1	3.45	162	
CLAF-900	1/30/2019	T-72	0.959	2.02	0.346	31.2	9.06	84.9	3.19	44.8	0.756

3.2 CLEANEX EXTRACTION [78CL-1]

January 31, 2019–February 10, 2019

HDEHP (1 M, 25.45 L) in Exxsol D60 was added to T-72 for the biphasic extraction of trivalent actinides and lanthanides away from other fission products. The acidity of the solution was adjusted to roughly 0.1 N by adding additional NaOH. The solution was sparged with air for 60 min and then left to settle. Sample CLER-373 (#3152) was sent to analytical to measure acidity. Once the acidity was confirmed to be 0.19 N, additional NaOH was added to adjust the pH to roughly 0.05 N. The solution was sparged again for 60 min before it was left to settle. A sample for analytical analysis was pulled as CLER-374 (#3168). The acidity was measured to be at 0.064 M. From here, another adjustment was performed using NaOH to adjust the pH to roughly 0.02 M followed by sparging and sampling CLER-375 (#3153). Acidity and solution activity were within the desired values. Accurate acidity adjustments are necessary to ensure all actinides are extracted out of the aqueous phase (3 moles acid is released for each mole trivalent An-Ln extracted). Once below an acceptable activity, the aqueous phase was transferred to T-23 while the organic phase remaining in T-72 is scrubbed with HNO_3 (2 x 0.03 M) and HCl (0.04 M) and sampled for activity (CLSR-942 [#3166]). Upon attempting to transfer the two scrubbing solutions to T-23, insufficient vacuum disabled this from occurring. Multiple attempts to tighten connections, install a new special FDV (flow diversion valve), and install a new PVC (process vacuum condensate) ball valve were unsuccessful. Eventually, the phase separator was rebuilt, and thereafter, the transfer to T-23 was successful. Another aliquot of HCl (7.5 L, 0.04 M) was added to T-72 for an additional scrub. A sample was taken and sent to analytical for analysis as CLSR-943 (#3157). The additional scrubbing solution was transferred to T-23. Stripping of the organic solution began February 3. The first strip was performed with HCl (9 L, 6.7 M) and H_2O_2 (500 mL, 30%). The addition of H_2O_2 is critical to keep Bk in the trivalent state for later separation. The solution was sparged for 1 h and then left to settle before it was transferred to T-73 for additional sparging. Once complete, the solution was sent to T-43, where evaporate was begun. A second strip was performed by adding HCl (4.5 L, 6.7 M), H_2O_2 (250 mL, 30%), and water (250 mL). The solution was sparged for 30 min and was left to settle before it was moved to T-73 for diluent washing before being sent to T-43. Similar strip batches were performed five more times during February 3-9 while continuing to evaporate. Meanwhile, all the contents of T-72 were emptied on February 9. T-72 was rinsed four times with HCl (0.25 M, 2 L) and twice with water (2 L). An additional 20 L of water was added to T-72 before it was set to evaporate down to 10 L. Once cool, 10 L of T-72 was transferred to T-23, where it was sparged and sampled as CLSR-944. Strips 3 and 4 were sent to T-43. Strip 5 was transferred from T-73 to T-72, where it was sparged with air and then sent to T-43. Strip 6 was added to T-72, which consisted of a mixture of HCl (4.5 L, 6.7 M) and H_2O_2 (30 %, 250 mL), followed by a water (250 mL) rinse. T-72 was sparged and then left to settle for 30 min before an aqueous sample was taken as CLCP-450. The sixth strip was transferred from T-72 to T-73. Then, the organic phase from T-72 was sampled as CLWO-137. Meanwhile, T-43 was set to evaporate and additional HCl (4.5 L, 6.7 M) and H_2O_2 (30 %, 250 mL) were added to T-73 before it was sampled as CLCP-451. The aqueous strip was transferred from T-73 to T-43 while the organic phase in T-73 was sampled as CLWO-138. Three samples of the desired product were then taken from T-43 as CLCP-452A, CLCP-452B, and CLCP-452C. T-72's organic solution was previously sent to T-23. T-73 organic stayed in T-73. Sample results are shown in Table 5 for CL-1. A process flowchart is shown in Figure 5.

Table 5. Assay results taken on samples for CL-1.

Tank	Sample ID	Date	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{137}Cs (Ci)	^{144}Ce (Ci)	^{154}Eu (Ci)
T-72	CLER-375	2/1/2019	0.011	0.288	0.050	0.0302	0.0088	0.017	3.50	0.104	0.068
T-72	CLSR-942	2/2/2019	0.0009	0.0188	0.0032	0.0043	0.0013	0.0044	0.0608	0.0158	0.0015
T-72	CLSR-943	2/3/2019	0.0014	0.0014	0.0002	0.0065	0.0019	0.0056	0.00808	0.0254	0.0005
T-72	CLCP-450	2/6/2019				0.0128	0.0037	0.0354	0.00145	0.0541	0.0005
T-72	CLWO-137	2/6/2019	1.58	0.361	0.0620	0.0004	0.0001	0.0022	0.036	0.0124	0.0148
T-73	CLCP-451	2/8/2019	0.0204	0.0006	0.0001	0.0091	0.0027	0.0231	0.0005	0.0396	0.0003
T-73	CLWO-138	2/8/2019	0.424	0.153	0.0262				0.0032	0.0116	0.0028
T-43	CLCP-452A	2/8/2019	0.733	1.86	0.301	31.9	9.25	92.8	0.133	0.0149	0.626
T-43	CLCP-452B	2/8/2019	0.765	1.79	0.882	33.3	9.66	90.6	0.133	0.0145	0.665
T-43	CLCP-452C	2/8/2019	0.776	1.20	0.305	33.1	9.62	92.8	0.133	0.0153	0.587

Blanks in the table refer to either values smaller than 1×10^{-5} or the sample was not analyzed for a specific isotope.

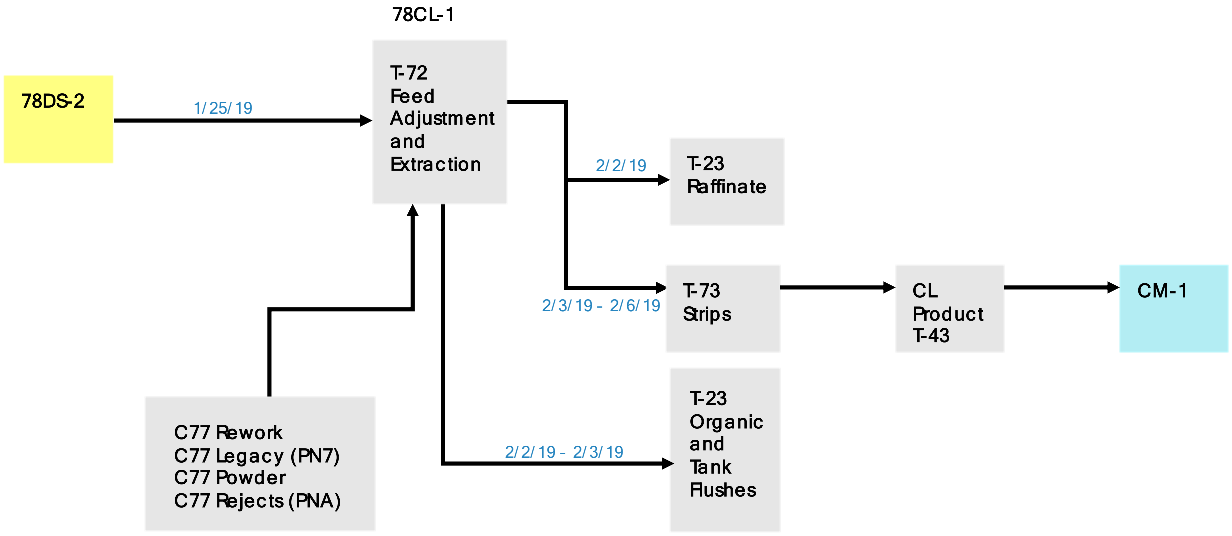


Figure 5. A flowchart showing a schematic of CL-1.

4. ACTINIDE AND LANTHANIDE SEPARATION BY LICL-BASED ANION EXCHANGE [78CM]

LiCl-based anion exchange is used for the separation of lanthanides from actinides as well as Am and Cm from transCm elements such as Bk, Cf, Es, and Fm. For this separation to occur, high chloride (Cl-) content must be present [4]. To achieve this, the feed solution is treated with LiCl salt that is dissolved in concentrated HCl. In the presence of high Cl- concentrations, lanthanide and actinide metals form anionic complexes such as $[AmCl_6]^{-3}$ [5], which can be separated on a chromatographic column based on their binding affinity, size to charge ratio, and the complex formed. Elution is achieved by decreasing the Cl- content. Elutriants are as follows: rare-earth 10 M LiCl, Am/Cm 9 M LiCl, and transCm 8 M HCl (no LiCl). The first two column runs (78CM-1 and 78CM-2; each loaded with half of the original feed solution) were eluted to separate lanthanides from actinides for waste disposal and to separate Am/Cm for target fabrication. The transCm element product was collected separately and then recycled in 78CM-3 to further separate Am/Cm, thus reducing the actinide (predominantly Cm) mass to an acceptably low level for the subsequent high pressure cation exchange separation of the actinide elements using elution chromatography with alpha-hydroxy isobutyric acid (AHIB) at various pH levels.

4.1 LiCl ANION EXCHANGE-1 [78CM-1]

February 4, 2019–February 13, 2019

Equipment checks and preparation started on February 4 by installing new HCV (hand control valve) units with new gaskets. Valves on (right rack 6) RR6 were installed and leak-checked the following day. The floor filer in Cub 6 was rebuilt with a 1-µm filter. Attempts were made to replace the needle block in T-634, but when loosening the nut, the manipulator started to bow from the applied force. This replacement was put on hold until the following day. M-706 and M-67 were flushed twice with water. The P-69 water loop was set to heat C-636 (anion column) to 70°C–80°C, hold for 1 h, and then cool to room temperature.

Feed solution preparation began on February 8 by evaporating 2 L from T-43. LiCl (1.5 L, 13.2 M) was added to T-43 followed by two additions of water (0.5 L). The tank was mixed by sparging air through the solution. The tank was then heated to 142.5°C and then cooled to below 50°C. HCl (126 mL, 12 M) was added to the tank to adjust the acidity to about 1 M and flushed the route with stock LiCl (200 mL, 13.2 M). The feed solution was filtered to remove insoluble Al, then transferred to T-65, where it was observed to be opaque in color with foam and solids present. To flush the line from T-43 to T-65, HCl (0.75 L, 1 M) was added five times. The solution was sparged with air for 5 min before the same route above was flushed with water (0.5 L) four times followed by additional HCl (4 L, 0.5 M) and water (4 L). The mixture was then sparged for 30 min before a sample was collected for analysis CMAF-175 (#3220). The mixture was set to evaporate.

Meanwhile, the filter used between T-43 and T-65 was back-flushed with HCl (0.5 M, 4 L) and water (4 L). HCl (2.5 L, 12 M) and two flushes of water (0.5 L) were added to T-43. This boiled down to roughly 10 L and then was sparged for 30 min before the reaction stopped.

T-65 continued to heat/evaporate until the temperature reached 142.5°C. It was then cooled and HCl (405 mL, 12 M) and LiCl (200 mL, 13.2 M) were added. The mixture was sparged with air for 15 min before it was heated to 120°C for 10 min and then cooled to below 50°C. In conjunction with this, C-636 was pretreated and adjusted to the correct flow rate with synthetic feed and LiCl stock solution. In preparation for loading half of the feed, ~2.5 L of T-65 was transferred to T-635. Column loading/elution started February 12 by transferring 2.4 L of hot feed solution through the anion column, C-636, to T-61 using pump P-636. The flow rate was set to 1.5 L/h and the feed was loaded until less than 0.2 L remained in T-635. The column was washed with 10 M LiCl, rare-earth elutriant, (3 × 500 mL, 1 × 1 L, and 2 × 500 mL). The wash solution was then switched to Am/Cm elutriant, 9 M LiCl, was precut, and was then sent to T-630 and the main Cm cut was collected in T-633. The Cf was stripped into T-632 using the Cf elutriant, 8 M HCl (2 × 500 mL, 1 × 1 L, 1 × 500 mL). The column was then flushed with 0.5 M HCl into T-72 and the pump was shut off. All fractions collected were adjusted with HCl and water before they were sampled and sent for analytical analysis (Table 6). A process flowchart is shown in Figure 6.

Table 6. Assay results for samples taken during CM-1.

Tank	Sample ID	Date	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{137}Cs (Ci)	^{144}Ce (Ci)	^{154}Eu (Ci)
T-65	CMAF-175	2/11/2019	0.56	2.17	0.372	31.1	9.04	86.4	0.186	133	0.578
T-61	CMCR-624	2/12/2019		0.007	0.001	0.01	0.003		0.006	0.006	0.29
T-630	CMCR-625	2/12/2019		0.005	0.0009	0.11	0.031	0.02	0.0009	0.46	0.003
T-633	CMAP-313	2/12/2019		0.603	0.104	11.2	3.24	0.105	0.0034	0.102	0.006
T-632	CMBP-164	2/12/2019	0.026	0.392	0.067	4.52	1.31	46.6	0.0387	0.062	0.047
T-72	CMCR-626	2/12/2019	0.288	0.002	0.0003	0.03	0.008	0.328	0.0009	0.073	

Blanks in the table refer to either values smaller than 1×10^{-5} or the sample was not analyzed for a specific isotope.

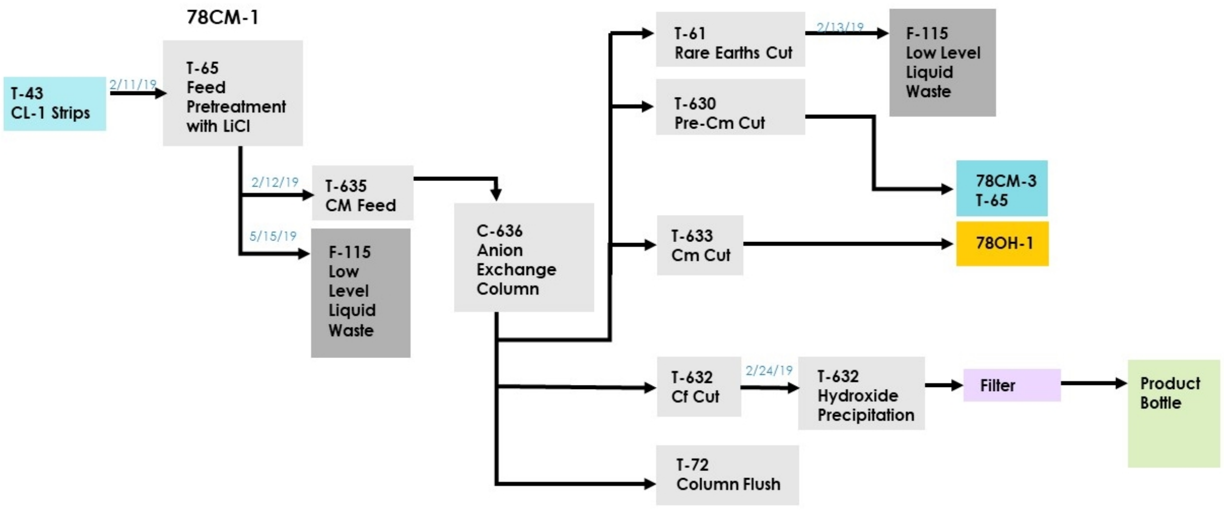


Figure 6. A flowchart showing a schematic of CM-1.

4.2 LiCl ANION EXCHANGE-2 [78CM-2]

February 12, 2019–February 18, 2019

Feed adjustments were made by adding HCl (500 mL, 12 M) and water (2 × 500 mL) to T-65. The solution was sparged for about 30 min with air. T-65 was heated to 142.5°C before it was shut off. Additional HCl (223 mL, 12 M) and LiCl (200 mL, 13.2 M) were added and sparged for 15 min. T-65 was set to heat to 120°C for 10 min before it cooled to 50°C. Roughly 2.5 L of the hot solution was pulled to T-635 while 3 × 500 mL of synthetic feed was added to T-65 and was sparged with air. In conjunction with this, the column (C-636) was pretreated and vented. The synthetic feed from T-65 was pulled into T-635 before T-635 was loaded onto the column (C-636). The column was washed with rare-earth elutriant and collected in T-61. The Am/Cm precut was collected in T-631 and the main cut of Cm was collected in T-634. The Cf main cut was then collected in T-632. The column was then flushed to T-72 before the pump shut off. T-61, T-631, and T-634 were adjusted with HCl (12 M) and water before they were sent for chemical analysis (Table 7).

Table 7. Sample results taken from fractions of CM-2.

Tank	Sample ID	Date	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{137}Cs (Ci)	^{144}Ce (Ci)	^{154}Eu (Ci)
T-631	CMCR-628	2/15/2019						0.09	0.001	0.42	0.002
T-632	CMBP-165	2/15/2019	0.01	0.590	0.102	5.03	1.46	93.2	0.062	0.26	0.071
T-634	CMAP-314	2/15/2019	0.134	1.09	0.187	17.8	5.16	0.008	0.002	0.32	0.004
T-61	CMCR-627	2/15/2019	0.031	0.091	0.003	0.10	0.03		0.005	0.66	0.321
T-72	CMCR-629	2/15/2019	0.073	0.133	0.023	0.05	0.014	0.803	0.002	0.11	0.002

Blanks in the table refer to either values smaller than 1×10^{-5} or the sample was not analyzed for a specific isotope.

The Cf main cut in T-632 was then transferred back to T-65 where it was evaporated down to below 4 L. The Am/Cm precut in T-630 was also transferred to T-65 and rinsed once with HCl (2 L, 1 M) and twice with water (1.5 L each). When trying to transfer the Am/Cm precut from CM-2 in T-631 to T-65 on February 16th, issues were encountered with the P-653 pump. Instead, T-65 continued to heat to 120°C and then was cooled. In addition to transfer issues, sample analysis showed that the solution in T-631 was uneconomical to recover via CM-3, so it was transferred to F-115 the next day and flushed with water.

Next, NaOH (10 L, 2 M) was added to T-61 with water. This was sparged for 15 min before it was sent to F-126 with water. A process flowchart for CM-2 is shown in Figure 7.

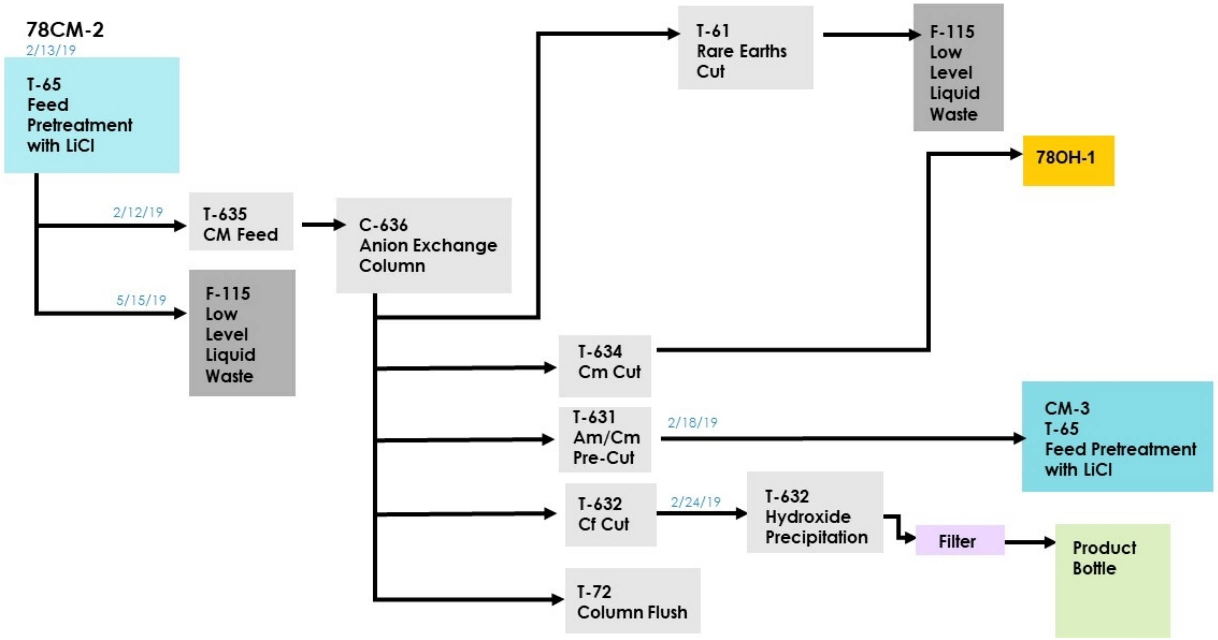


Figure 7. A flowchart showing a schematic of CM-1.

4.3 LiCl ANION EXCHANGE-3 [78CM-3]

February 17, 2019–March 1, 2019

On February 17th, new resin was loaded into C-636. During the column loading process, L-11 goose neck became clogged. To fix this problem, the HCV unit needed to be placed at the HCV-C636-1 position and new flat gaskets needed to be installed. This was not possible because of a broken tape on the manipulator.

The recycled Transcurium products from 78-CM-1 and 78-CM-2 were combined in T-65, and then heated to 142.5°C, with sparging up to 80°C. After cooling the feed solution in T-65 to room temperature, HCl (239 mL, 12 M) and LiCl (200 mL, stock solution) were added. The mixture was sparged with air for 15 min and heated to 120°C where it was held for 10 min to homogenize the feed solution, then cooled to ambient temperature for loading. Roughly 3 L of hot feed in T-65 was transferred to T-635 and the actinides were loaded on the resin. (C-636 had been previously preconditioned with synthetic feed pulled from T-635 through the column to T-61). Hot feed was then pumped through the column and T-635 was rinsed with synthetic feed and transferred to the column. Once all of the feed was loaded onto the column, five fractions of 10 M LiCl RE elutriant (500 mL) were fed through the column and collected in T-61. Then, three fractions of 9 M LiCl Am/Cm elutriant (500 mL) plus another 1 L were passed through the column and collected in T-630. Seven additional fractions of the Am/Cm elutriant (4 × 500 mL, 3 × 250 mL) followed by 500 mL of the Cf elutriant were collected in T-631. Two more fractions of the Cf elutriant (500 mL) were collected in T-632. The column flushes to follow were collected in T-72. Tanks were acidified by adding calculated amounts of HCl (12 M) and water. The contents of T-72 will become rework, and T-632 will go on as the main product. The following samples were taken for analysis and are shown in Table 8. A process flowchart for CM-3 is shown in Figure 8.

Table 8. Sample assay results taken during CM-3.

Tank	Sample ID	Date	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{137}Cs (Ci)	^{144}Ce (Ci)	^{154}Eu (Ci)
T-630	CMCR-631	2/19/2019				0.002		0.037	0.002	0.015	0.002
T-631	CMAP-315	2/19/2019	0.031	0.487	0.084	4.93	0.611	0.037	0.005	0.102	0.005
T-632	CMBP-166	2/19/2019		0.257	0.044	0.107	0.031	85.9	0.044	0.199	0.058
T-61	CMCR-630	2/19/2019			0.001			0.001	0.001	1.49	0.007
T-72	CMCR-632	2/19/2019	0.231	0.006	0.001	0.080	0.023	3.21	0.003	0.101	0.003

Blanks in the table refer to either values smaller than 1 × 10^{-5} or the sample was not analyzed for a specific isotope.

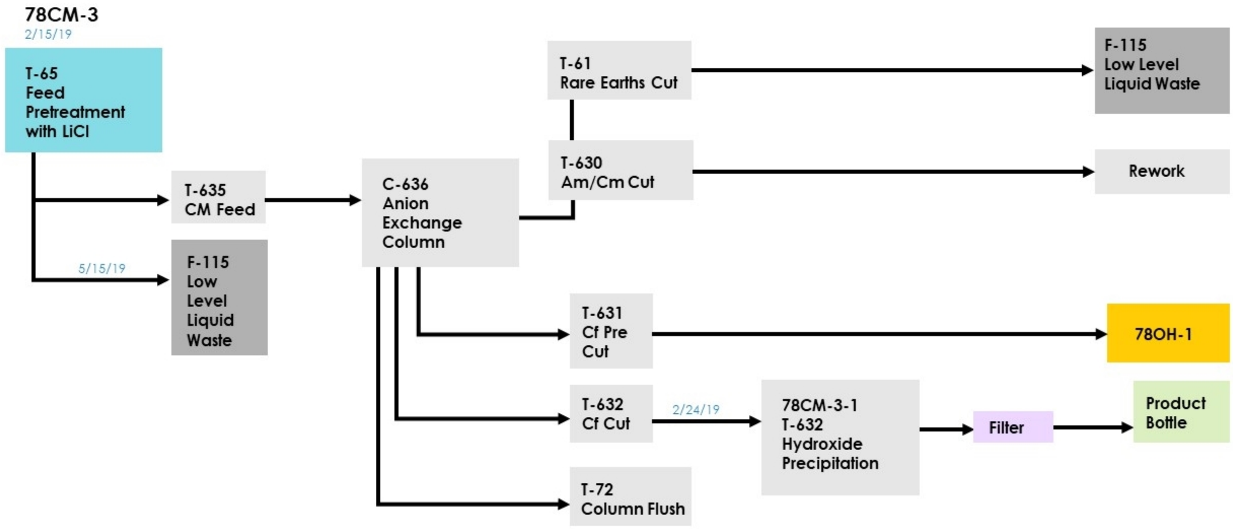


Figure 8. A flowchart showing a schematic of CM-3.

4.4 PURIFICATION OF TRANSCURIUM ACTINIDES BY LiOH [78CM-3-1]

February 24th, 2019–February 28th, 2019

After the LiCl anion exchange was complete, the transcurium products were converted from chloride to nitrate form by hydroxide precipitation, filtration, and re-dissolution of the precipitate in nitric acid. This conversion was essential to prevent corrosion during the transcurium actinide elemental separation which is performed in stainless steel equipment. This process also removed any LiCl left over from the anion exchange operations. To do this, 5 M LiOH was added to the main Cf product in T-632 and then filtered through a glass frit to collect all the precipitates (transcurium hydroxides $An(OH)_3$). The precipitate was then redissolved in HNO_3 and saved for the next separation (AHIB cation exchange).

Purification of the transcurium actinides by hydroxide precipitation began on February 24 by slowly adding $FeCl_3{\cdot}6H_2O$ (0.98 g) in water (50 mL) to T-632 as a carrier for the hydroxide precipitation. Sparging with air was increased and LiOH (5 M, 1 L) was added followed by two water washes (250 mL). T-632 continued to be sparged with air for an additional 2 h. The solution was then filtered through a glass frit collecting the first 700 mL as the first batch, which had a neutron reading of 3,300 counts/s. The solution was then pumped to the intermediate bottle, and the solution was filtered again. This process was repeated three times until neutron counts had decreased to1,000 c/s, and the remaining solution was transferred back to T-65. The second batch was then pumped from T-632 through the filter to the product bottle. This solution was filtered three additional times before it was sent to T-65. Batch three was then pulled from T-632, filtered three times, and sent to T-65. Batches four and five were filtered once before sending the solution to T-65. The solid filtrate was then washed with water that was pumped from T-632 through the filter. After several water washes, the solution was counted and sent to T-65. To redissolve the solids left on the filter, a funnel was installed on the top of the glass frit filter and HNO_3 (14 mL, 15.8 M) was slowly added and allowed to soak for about 15 min. Water (10 mL) was then added and allowed to sit on the filter for about 15 min. Additional rinses of T-632 with HNO_3 (0.2 M) were transferred through the filter and collected in the product bottle. Again, HNO_3 (14 mL, 8 M) was added to the filter and left to sit for roughly 15 min followed by water (10 mL). T-632 was rinsed with HNO_3 (0.2 M), which was sent through the filter to collect in the intermediate bottle. Counts were about 300 counts/s on all sides. This rinse and counting method were performed twice more until the filter counts decreased to 320 counts/s and the product bottle counts reached 22,000 counts/s. Additional water (50 mL) was added to the filtrate/product bottle and it was stirred by sparging with a dip wand. Two samples of the filtrate/product bottle CMBP-167 A (#3185) and CMBP-167 B (#3217) were submitted for chemical analysis (Table 9).

Table 9. Sample results from CM3-1.

Sample ID	Date	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{137}Cs (Ci)	^{144}Ce (Ci)	^{154}Eu (Ci)
CMBP-167A	2/26/2019	0.055	0.0095	0.076	0.022	63.2	0.061	0.243	0.065
CMBP-167B	2/26/2019			0.081	0.024	63.0	0.061	0.243	0.065

Blanks in the table refer to either values smaller than 1×10^{-5} or the sample was not analyzed for a specific isotope.

4.5 ADDITIONAL HYDROXIDE PRECIPITATION [78CM-3-2]

February 26, 2019–March 8, 2019

An additional hydroxide precipitation was performed due to losses from the first precipitation. To remove more ^{252}Cf material from the filter, the filter was flushed with HCl (1 L, 8 M) followed by 1 L of water to T-632. The tank was sparged with air for 30 min before it was pulled back through the filter to T-65. The filter was pumped dry. Additional HCl (6.5 L, 1 M) was added to T-632 and pulled through to T-65 followed by an addition of water to T-632, which ended again in T-65. T-65 was sampled as CMCF-544 (#3245) for chemical analysis. T-65 was then evaporated to roughly 2 L. Water (4 L) was added to T-65 and it was sparged with air for 1 h before it was sampled as CMBP-168 (bottle # 3237). T-65 was evaporated down to roughly 2 L. All of the solution in T-65 was then transferred to T-632. The route was then flushed with water (250 mL) four times.

On March 5th, a filter was installed between T-632 and T-65. To perform an additional hydroxide precipitation, $FeCl_3$ (0.98 g in 50 mL of water) was slowly added to T-632. The solution was sparged with air and LiOH (1 L, 5 M) was added followed by two additions of water (250 mL). While adding LiOH, the flow rate became increasingly slower than normal. This was caused by either a clog at the V-M706-6 line or because the HV-639 toggle valve failed. The clog progressively worsened as water was added. Gentle tapping on the check valve seemed to allow the flow rate to speed up again. During the second rinse with water, the check valve was tapped again, but this time, it broke. A work request to have this fixed was placed. It is estimated that about 200 mL of water did not make it to T-632. T-632 was subsequently sparged for about 2 h. Later that night, the M-706-6 check valve was changed and attempts were made to transfer water to T-632, but the solution would not transfer. To address this, the valve at the HV-639-1 position was removed. During this process, water shot out of the line. A new valve was installed, and the water was successfully transferred to T-632. The solution was transferred from T-632 to the product bottle through a glass frit. Solids collected on the filter. Liquid in the product bottle was sent to T-65 while T-632 continued to be filtered. This was accomplished in six batches. The filter was washed multiple times with water, sending all of the solution to T-65 as described above. To dissolve the precipitate, HNO_3 (14 mL, 15.8 M) was added to the filter dropwise. The solution was allowed to sit for 15 min before it was pulled through to the sample bottle. The filter was washed with water (10 mL) and then with HNO_3 (14 mL, 8 M) and finally with water (10 mL). In between these rinses, T-632 was also rinsed with HNO_3 (100 mL, 0.2 M) through the filter into the sample bottle. In total, the product bottle contained roughly 700 mL and was sampled as CMBP-169A (bottle #3271) and CMBP-169B (bottle #3268) for chemical analysis (Table 10).

Table 10. Sample results from CM-3-2.

Sample ID	Date	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{137}Cs (Ci)	^{144}Ce (Ci)	^{154}Eu (Ci)
CMCF-544	2/28/2019	0.015		0.003	0.032	0.009	21.3	0.009	0.037	0.0099
CMBP-169A	3/7/2019	0.012	0.019	0.003	0.027	0.008	18.8	0.011	0.045	0.013
CMBP-169B	3/7/2019	0.004	0.019	0.003	0.027	0.008	20.4	0.011	0.049	0.013

5. SEPARATION OF ACTINIDES USING AHIB CATION EXCHANGE [78CX-1 AND -2]

AHIB (α-hydroxyisobuteric acid) in combination with a Dowex 50W-8X cation exchange column allows for the separation of individual transcurium elements from one another. This is an important step in this chemical processing campaign not only to separate a pure Cf stream, but also to separate other elements of interest, such as Bk, Es, and Fm. This separation is controlled by eluting with AHIB in different pH ranges [5]. A key to good separations is the use of very small size resin particles and high-pressure operation. The use of separate loading and elution columns

5.1 AHIB CATION EXCHANGE FOR TRANSCURIUM SEPARATION [78CX-1]

February 21, 2019–March 9, 2019

Reagent and equipment preparations began on February 21. All AHIB solutions were sampled to ensure correct pH ranges before use. Two columns were prepped for this separation. C-401 was loaded with Dowex 50W-X8 resin (30–45 µm) and C-402 was loaded with Dowex 50W-X8 resin (45–56 µm). Loading of the actinides onto the columns began on March 5. The feed collected from 78CM3-1 was loaded in two batches. The first batch was pulled from the feed bottle to T-404 through columns C-402 and C-401 and collected in the raffinate bottle labeled “R-1.” Followed by the second transfer of feed (150 mL) from the feed bottle to T-404 through columns C-402 and C-401 where the raffinate was collected in the bottle labeled “R-2”. The columns were then washed with water (80 mL), NH_4NO_3 (300 mL), and again with water (80 mL), which was all collected into R-2. The first cut was collected by eluting with AHIB (0.25 M, pH 3.9) to the bottle “78CX-1 #1 cut.” Cut #2 was collected next using the same AHIB pH. Cuts #3 through #8 were collected using AHIB (0.25 M, pH 4.2) to elute. 78CX-1 cuts #5 and #4 contained the Es fractions, cut #6 contained Es and Cf, and cuts #7 and #8 contained only Cf. Eluting with AHIB (0.25 M, pH 4.6) resulted in cuts #9, #10, and #11, which contained Bk. The columns were washed with AHIB (0.5 L, 0.25 M, pH 4.8) and then water (0.4 L) and the washes were collected in R-3. After the washes, the columns were counted using an in-cell neutron probe. C-401 had a max reading of 100 counts/s and C-402 had a max reading of 2 counts/s. All fractions were acidified, mixed, and sampled (2–3 mL) as shown in Table 11. Cut #7 was transferred to the 78CM3-2 product bottle. The resin was then removed from C-402, counted, and stored. Cuts #10 and #11 were transferred to T-45. A new feed bottle labeled “78CX-2 feed bottle” was added to the cell where the filtrate product bottle 78CX-3 cut #2 and 78CX-1 cut #9 were added to it. The bottle was sparged for 30 min and then sampled as CXAF-112 (#3222) to check the molarity of the solution. Sample results are shown in Table 11 and a process flowchart for CX-1 is shown in Figure 9.

Table 11. Sample results taken from CX-1.

Sample	Sample ID	Date	Element of Interest	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk (mg)	^{253}Es (µg)
Cut #1	CXER-421	3/5/2019									
Cut #2	CXER-422	3/5/2019		0.0001					0.0003		0.0004
Cut #3	CXER-423	3/5/2019									0.0004
Cut #4	CXES-320	3/5/2019	Es								0.0009
Cut #5	CXES-321	3/5/2019	Es						0.0001		0.0169
Cut #6	CXES-322	3/6/2019	Es	0.0024					0.0005		9.66
Cut #7	CXCF-822	3/6/2019	Cf	0.174					30.4		8.54
Cut #8	CXCF-823	3/6/2019	Cf	0.0057			0.0006	0.0002	25.7		
Cut #9	CXBK-325	3/6/2019	Bk	0.0014			0.0002		1.61	7.79	
Cut #10	CXBK-326	3/6/2019	Bk	0.0047	0.0005				0.0293	0.044	0.0026
Cut #11	CXBK-327	3/6/2019	Bk	0.0049	0.0002		0.0232	0.0067	0.0080		
R-1	CXLR-531	3/5/2019									
R-2	CXLR-532	3/5/2019									
R-3	CXSF-291	3/5/2019		0.0043	0.0013	0.0002	0.0443	0.0013	0.0306		
78CX-2 Feed	CXAF-112	3/11/2019		0.277			0.0295	0.0086	42.4	0.011	8.38

Blanks in the table refer to either values smaller than 1×10^{-5} or the sample was not analyzed for a specific isotope.

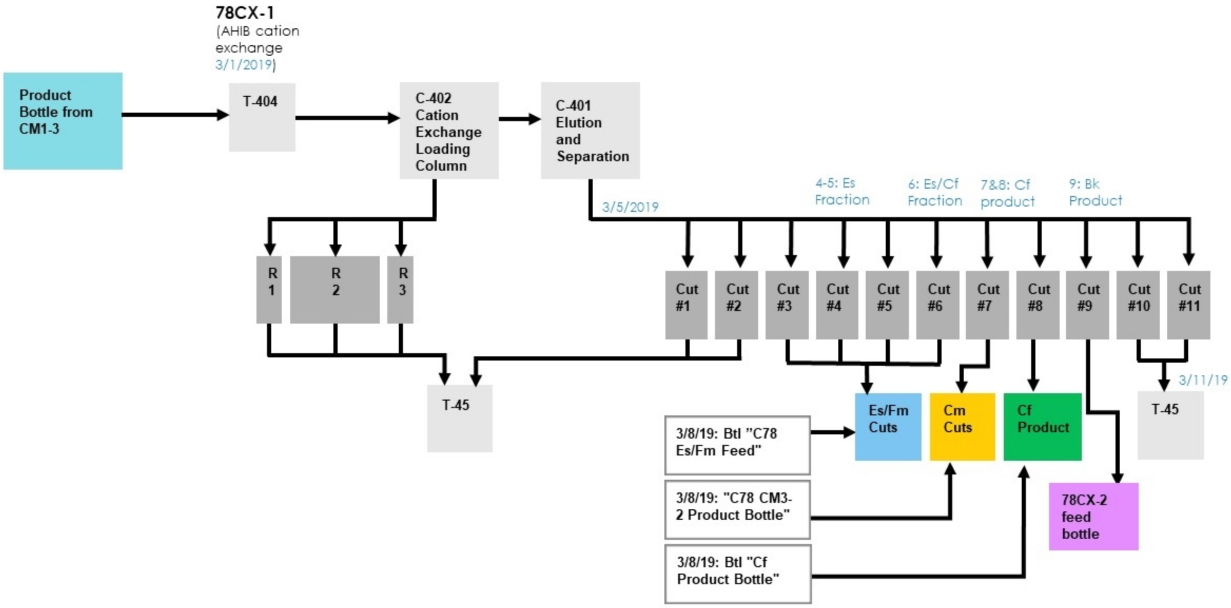


Figure 9. A flowchart showing a schematic of CX-1.

5.2 AHIB CATION EXCHANGE [78CX-2]

March 11, 2019–March 14, 2019

New resin was loaded onto C-402 in preparation for the next AHIB column run which commenced on March 11th. Column loading began on March 12, by first diluting the feed to 1.5 L and sparging for 30 min. The first batch of feed (800 mL) was loaded onto C-402 and the raffinate was collected in R-1. Once R-1 was full (approximately 900 mL), the collection was switched to R-2 for the final loading of feed (700 mL). The feed bottle was rinsed out with water (80 mL), which was collected in R-2, followed by NH_4NO_3 (300 mL, 0.3 M), which was collected in R-3, and lastly with water (80 mL), which was collected in R-2. To start eluting from C-402 through C-401, AHIB (pH 3.9) was added to the fraction bottle for cuts #1 (220 mL) and #2 (220 mL). Next, AHIB (pH 4.2) was added to collect cut #3 through #10. Then, for cuts #11 through #13, AHIB (pH 4.6) was used. After all cuts were taken, the columns were washed with AHIB (pH 4.8, 470 mL) and then with water (900 mL), which was collected in R-4. All cuts were diluted, acidified, and mixed as needed. Each cut was sampled, and results are shown in Table 12. A process flowchart for CX-2 is shown in Figure 10.

Table 12. Sample results taken during CX-2.

Sample	Sample ID	Date	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk (mg)	^{253}Es (µg)
Feed	CXAF-112	3/11/2019	0.277			0.0295	0.0086	42.4	11.0	8.38
Cut #1	CXER-424	3/12/2019						0.0004		
Cut #2	CXER-425	3/12/2019						0.0006		0.0032
Cut #3	CXER-426	3/12/2019						0.0003		0.0039
Cut #4	CXES-323	3/12/2019						0.0003		0.0172
Cut #5	CXES-324	3/12/2019	0.0007					0.0004		2.71
Cut #6	CXES-325	3/12/2019	0.0020					0.040		7.16
Cut #7	CXES-326	3/12/2019	0.0013					0.178		0.883
Cut #8	CXCF-824	3/12/2019	0.0562					9.71		
Cut #9	CXCF-825	3/12/2019	0.0155	0.001	0.0002	0.0154	0.005	34.4		
Cut #10	CXCF-826	3/12/2019	0.0327			0.0002		5.20	0.002	
Cut #11	CXBK-328	3/12/2019	0.0004					0.952	11.7	
Cut #12	CXBK-329	3/12/2019	0.0066	0.001	0.0002	0.0001		0.0241	0.039	
Cut #13	CXBK-330	3/12/2019	0.0011			0.0116	0.003	0.0034	0.001	
R-1	CXLR-533	3/12/2019	0.0001					0.0018		0.0005
R-2	CXLR-534	3/12/2019						0.0006		0.0002
R-3	CXLR-535	3/12/2019						0.0002		
R-4	CXSF-292	3/12/2019	0.0015			0.02	0.006	0.0221		
Es/Fm	CXES-327	3/18/2019	0.0037					0.317		16.1

Blanks in the table refer to either values smaller than 1 × 10^{-5} or the sample was not analyzed for a specific isotope.

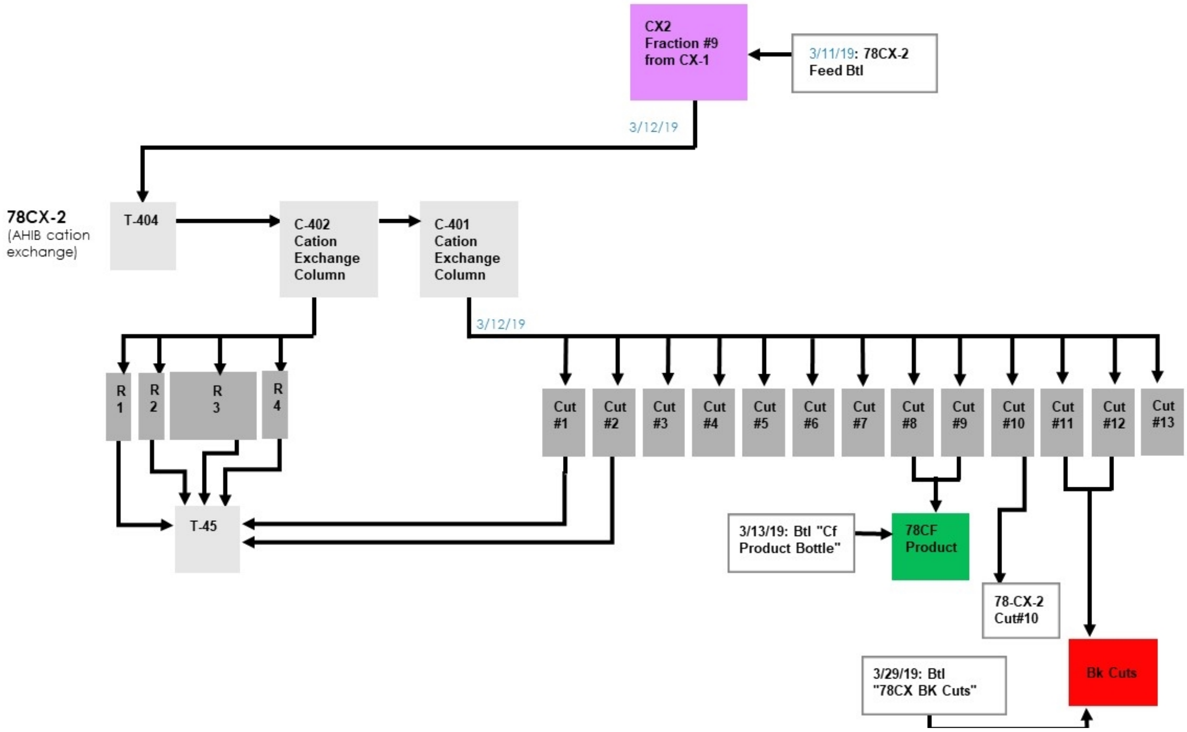


Figure 10. A flowchart showing a schematic of CX-2.

Cut #9 was combined with the C78 Cf product bottle followed by flushing the cut #9 container with water (35 mL), HNO_3 (1 mL, 15.8 M), and water (20 mL). Cut #8 was also combined into the C78 Cf product bottle along with two water (10 mL) rinses. Cut #10 was transferred into a bottle labeled "78CX-2 cut #10." Resin from C-402 was counted and emptied into a bottle, labelled C78 resin. Resin from C-401 was also counted and removed in a similar manner. Cuts #3 through #7 were combined into "C78 Es/Fm feed" and rinsed with water and nitric acid for a final volume of roughly 850 mL. The bottle was sparged with air and a sample was taken as CXES-327 (bottle #3340) for chemical analysis. Cuts R-1, R-2, R-3, R-4, #1, and #2 were flushed to T-45 with water (50 mL).

5.3 AHIB CATION EXCHANGE [78CX-3]

March 15, 2019–April 1, 2019

New resin was loaded onto C-401 and C-402 as preparations for the next AHIB column run began on March 15. Water (200 mL) was pumped to evacuate T-404 and was discharged into R-1. Hot feed (600 mL) from the C78 Es/Fm feed bottle was pumped onto the columns and collected in R-1 until it reached 800 mL; then, the column collection was switched to R-2. The feed bottle was rinsed with NH_4NO_3 (300 mL, 0.3 M) and the solution was transferred where. AHIB (0.25 M, pH 3.9) was pumped through the columns to start elution and cuts #1 through #8 were collected. The solution was changed to 0.5 M AHIB (pH 4.8) to rinse the column and was collected in R-3. All cuts and fractions were acidified, mixed, and sampled (Table 13).

Table 13. Sample results taken during CX-3.

Sample	Sample ID	Date	^{252}Cf (mg)	^{253}Es (µg)
Cut #1	CXER-427	3/19/2019	0.0010	0.0002

Cut #2	CXER-428	3/19/2019	0.0022	0.0002
Cut #3	CXES-328	3/19/2019	0.0005	0.0134
Cut #4	CXES-329	3/19/2019	0.0004	1.24
Cut #5	CXES-330	3/19/2019	0.0002	5.44
Cut #6	CXES-331	3/19/2019	0.0002	3.85
Cut #7	CXES-332	3/19/2019	0.0017	2.18
Cut #8	CXCF-827	3/19/2019	0.185	0.881
R-1	CXLR-536	3/19/2019	0.0005	
R-2	CXLR-537	3/19/2019	0.0006	0.0005
R-3	CXCM-249	3/19/2019	0.0500	0.088

Blanks in the table refer to either values smaller than 1 × 10^{-5} or the sample was not analyzed for a specific isotope.

The resin was removed from the columns and collected in a C78 resin storage bottle. On March 29, the Bk cuts, 78CX-2 cut #11 and cut #12, were combined into a “78CX Bk cuts” bottle and sampled for pH analysis as CXBK-331 (#3313). 78CX-2 cut #10 was combined with the “C78 Cf product” bottle and was sampled as CXCF-828 (#3341, Table 14). Cuts #2 through #7 were transferred to the “C78 Es/Fm product” bottle on April 1. A process flowchart for CX-3 is shown in Figure 11.

Table 14. Cf-252 assay results from CX-3.

Sample	Sample ID	Date	^{252}Cf (mg)
C78 Cf product	CXCF-828	3/29/2019	73.3

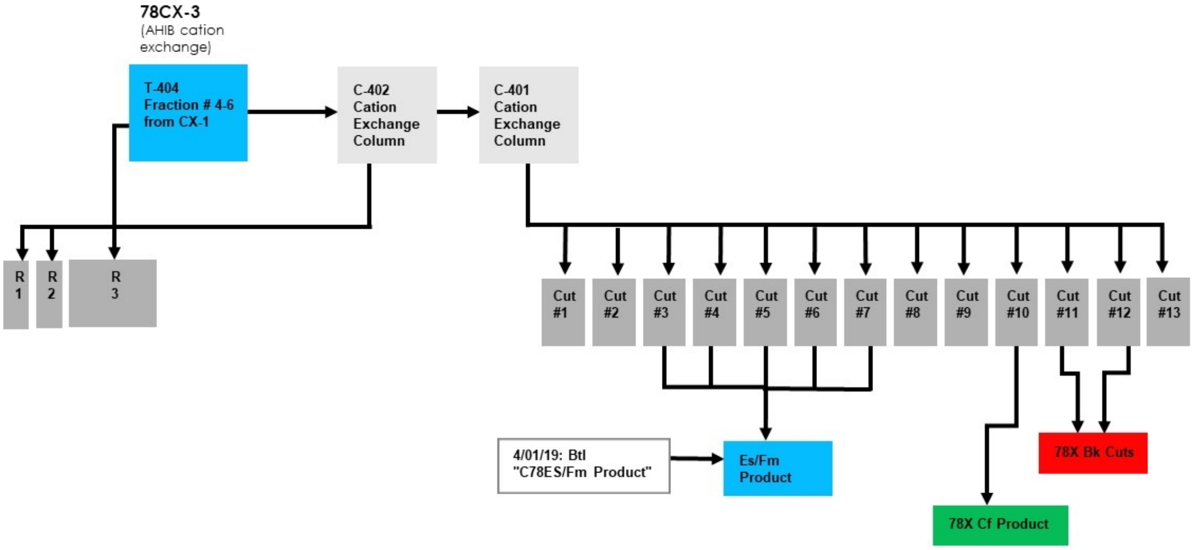


Figure 11. A flowchart showing a schematic of CX-3.

6. PURIFICATION OF BERKLIUM [78BK]

6.1 BERKEX [78BK-1]

March 25, 2019–May 1, 2019

Reagent preparations and column loadings started on March 25th by loading about 25 mL of Dowex 50W-X8 (42–56 µm) slurried into C-402. Leak testing and feed adjustments followed. Water was tested through T-404 to C-402 to the effluent line and collected into R-1 with no issues. Feed adjustments began on March 31 by leak checking the path from T-404 through the column (C-402), and then through the effluent line to R-1. Once no leaks were detected, 500 mL of the feed (78CX Bk cuts) was transferred to T-404. The feed was then fed through C-402 and collected in R-1. The feed bottle was rinsed twice with water (200 mL), which was loaded on C-402 and collected in R-2. Then, HNO_3 (300 mL, 8 M) was transferred into the original feed bottle. In four batches, the acid was transferred onto the column and collected in the "feed/raff" bottle. The Bk cuts bottle was rinsed with water (100 mL) twice, put through the column, and collected in R-3. Once the column run was complete, additional HNO_3 (100 mL, 15.8 M) was added to the feed/raff bottle. All cuts were sampled, and the results are shown in Table 15.

Table 15. Sample results taken prior to BK-1.

Sample name	Sample ID	Date	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{249}Bk (mg)	^{252}Cf (mg)
78BX-1 feed/raff	BXBK-084	3/31/2019	0.0067	0.0014	0.0003	0.0003	13.4	0.913
R-1	BXRF-221	3/31/2019						0.0004
R-2	BXRF-222	3/31/2019						
R-3	BXRF-223	3/31/2019						0.0005

Blanks in the table refer to either values smaller than 1 × 10^{-5} or the sample was not analyzed for a specific isotope.

Additional purification of the Bk continued with preparations for an AHIB cation exchange column run which started on April 15 by unloading resin for column C-402, and reagent makeup began on April 22. HDEHP (0.5 M) was washed twice with a 250-mL mixture of $NaBrO_3$ (0.32 M) and HNO_3 (6.8 M). Then, HNO_3 (1 L, 15.8 M), $NaBrO_3$ (200 mL, 2 M), and water (25 mL) were added to T-411. The tank was then sparged with air for 15 min before it was transferred to T-45. The feed from the 78BX-1 feed/raff was transferred to T-411. The bottle holding the feed was rinsed with $NaBrO_3$ (90 mL, 2 M) and water (10 mL) before it was also transferred to T-411. T-411 was sparged for 5 min before the old feed/raff bottle was rinsed with HDEHP (500 mL) in dodecane. The organic phase was then transferred to T-411, where it was sparged for 30 min and left to settle for 10 min. The aqueous phase was drained into the 78BX-1 feed/raff bottle. Two scrubs were preformed using $NaBrO_3$ (90 mL, 2 M) and HNO_3 (500 mL, 8 M), where the aqueous was once again drained into SR-1 and SR-2, respectively. Two back-extractions were performed using H_2O_2 (30 mL, 30%) and HNO_3 (250 mL, 8 M), and the HNO_3 was collected in the bottle labeled "78BX-1 product bottle." The organic phase was drained into the WO/HDEHP bottle and T-411 was rinsed with water (3.1 L) and HNO_3 (1.5 L, 8 M). The final sample results are shown in Table 16. A flowchart of the process flow of BK-1 is shown in Figure 12.

Table 16. Sample results taken after BK-1.

Sample name	Sample ID	Date	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{249}Bk (mg)	^{252}Cf (mg)
Feed bottle	BXRF-224	4/26/2019	0.0002	0.0003		0.0003		0.945
WO/HDEHP	BXWO-038	4/26/2019	0.0045	0.0010	0.0002			

SR1	BXRF-225	4/26/2019						0.0169
SR2	BXRF-226	4/26/2019						0.0003
Product	BXBK-085	4/26/2019					13.8	
T-411	BXFL-040	4/26/2019						0.0001

Blanks in the table refer to either values smaller than 1 × 10^{-5} or the sample was not analyzed for a specific isotope.

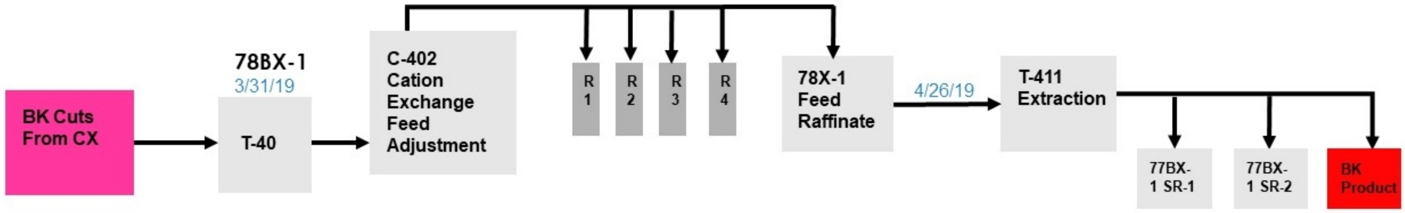


Figure 12. A flowchart showing the process flow of BX-1.

7. CALIFORNIUM CAPSULE LOADING [78CF]

The Cf separated using AHIB cation exchange process (CX) was collected into one bottle labeled “C78 Cf product.” Each nut package consisted of a platinum cylinder filled with Dowex 50W-8X resin fritted on both ends and capped with Swagelok nuts. Each of these packages can hold up to 40 mg of Cf; thus, the number of nut packages will vary depending on the campaign. Here, there were a total of three nut packages named “D-110,” “D-111,” and “D-112”. Californium was loaded onto each package using a pump. The Cf was then converted into a sulfate or oxysulfate form by removing the buts and inserting the cylinder into a firing wand. The assemblies were then cured for 24 h before they were heated to 150°C for 1 h, 350°C for 1 h, and 700°C for 4 h under a stream of air being pulled by a vacuum. After conversion, they were reassembled with their nuts and transferred to Building 7930 for storage and further processing. A flowchart of the process flow of 78CF is shown in Figure 13.

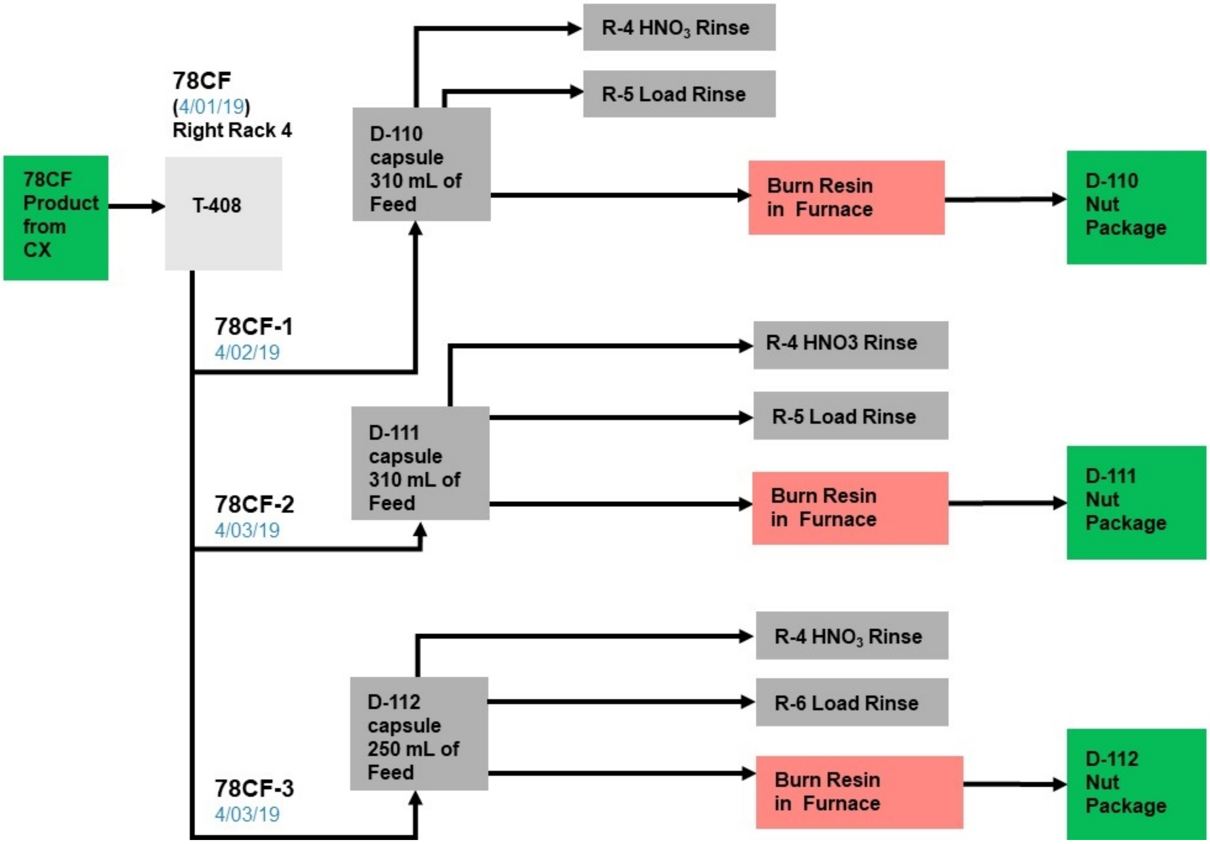


Figure 13. A flowchart showing a schematic of 78CF.

7.1 CALIFORNIUM LOADING INTO NUT PACKAGE D-110 [78CF-1]

April 1, 2019–April 4, 2019

Equipment and leak checks were started on April 1 and capsule loading began on April 2. The capsule was first rinsed with HNO_3 (150 mL, 0.3 M), which was collected in R-4. The feed bottle was sparged for 15 min with air and a glass sample “thief.” During this process, two thieves broke off into the feed bottle. Feed (310 mL) was then pumped through D-110 to R-5 and was rinsed twice with water (50 mL). The capsule was allowed to vent for 30 min. The capsule was then disconnected from the rack and the bottom cap was installed onto the nut package. The top reducer was removed and transferred to the leach bottle. The platinum capsule was removed from the nut package and was installed into the firing wand and placed into the box furnace. The furnace was heated to 150°C for 1 h, followed by 350°C for 1 h, and 700°C for 4 h before cooling to room temperature. After it was cooled, the capsule was reinstalled into the nut package and the top and bottom nuts were tightened.

7.2 CALIFORNIUM LOADING INTO NUT PACKAGE D-111 [78CF-2]

April 3, 2019–April 5, 2019

Capsule loading was performed on April 3 by first rinsing the resin with HNO_3 (150 mL, 0.3 M), which was collected into R-4. Roughly 310 mL of feed was loaded onto D-111 and the solution was collected into R-5 along with two water (50 mL) rinses. When R-5 was checked with the neutron probe, it indicated elevated readings (192 counts/s). As a result, a sample of R-5 was sent or analysis (CXLR-538, Table 17) before it was added to the Cf product bottle and then rinsed with HNO_3 (2 x 250 mL, 0.3 M) to T-23. Meanwhile, D-111 was removed from the rack and the frits were inspected. Small black specks were present on the top frits, but the bottom and top were in good condition. The capsule was removed from the packaging and placed into the second firing wand. The assembly sat for 24 h before heating occurred. The furnace was heated to 150°C for 1 h, then 350°C for 1 h, and 700°C for 4 h before cooling. After it was cooled, the capsule was reinstalled into the nut package and the top and bottom nuts were tightened.

Table 17. Sample results from R-5 taken during package loading.

Tank	Sample ID	Date	Pu (g)	^{252}Cf (mg)	^{253}Es (μg)
R-5	CXLR-538	4/3/2019	0.0044	0.704	0.0167

7.3 CALIFORNIUM LOADING INTO NUT PACKAGE D-112 [78CF-3]

April 3, 2019–April 5, 2019

The capsule was first rinsed with HNO_3 (150 mL, 0.3 M), which was collected in R-4. The feed bottle was sparged for 15 min with air. During this sparging process, two thieves broke off into the feed bottle. Feed (250 mL) was then pumped through D-112 to R-6 and D-112 was rinsed twice with water (50 mL) and the washes added to R-6. The capsule was allowed to vent for 30 min. The capsule was left on the rack until the firing of D-112 was complete. It was then disconnected from the rack and the bottom cap was installed onto the nut package. The top reducer was removed and transferred to the leach bottle. The platinum capsule was removed from the nut package and was installed into the firing wand and placed into the box furnace. The furnace was heated to 150°C for 1 h, then 350°C for 1 h, and 700°C for 4 h before cooling. After it was cooled, the capsule was reinstalled into the nut package and the top and bottom nuts were tightened.

8. AMERICIUM/CURIUM HYDROXIDE PRECIPITATION [78OH]

8.1 AMERICIUM/CURIUM HYDROXIDE PRECIPITATION [78OH-1]

April 5, 2019–April 11, 2019

Sample preparations and evaporation of the Am/Cm material in T-40 began on April 5. During the cleaning and recharging of the deep-bed filter (DBF), it was noticed that there was a crack in the water line in Cub 4 resulting in a leak. This was fixed and leak checks were successfully completed. On April 8, T-40 was sparged for 15 min and then 10 L of solution was transferred to T-47. T-40 was back-flushed with water (500 mL) and the system was flushed forward with water (4.5 L) twice and all washes were transferred to T-47. T-47 was sparged for roughly 30 min and a sample was taken for chemical analysis OHPF-055 (#3334). Sodium hydroxide (8 L, 9.95 M) was slowly added to T-47 followed by two water (2 L) rinses. T-47 was then sparged for 30 min and was left to settle for 2 h. Three batches of T-47 were pulled into T-444 and transferred through the DBF to T-21. Once all of the solution was transferred, NaOH (4.5 L, 2 M) was added to T-47. The tank was sparged for 30 min and then allowed to settle for 30 min before the solution was transferred out of the tank and flushed through the DBF to T-21. This process was repeated twice more with NaOH; then, HNO_3 (2.5 L, 8 M) was added to dissolve the actinide hydroxides left on the DBF. This was transferred to T-54 followed by additional HNO_3 (1 M) flushes and two water (2 L) flushes. T-54 was then evaporated down to less than 5 L of solution. All of the solution in

T-21 was transferred to F-115 followed by two water (10 L) rinses. A flowchart of the process flow of OH-1 is shown in Figure 14.

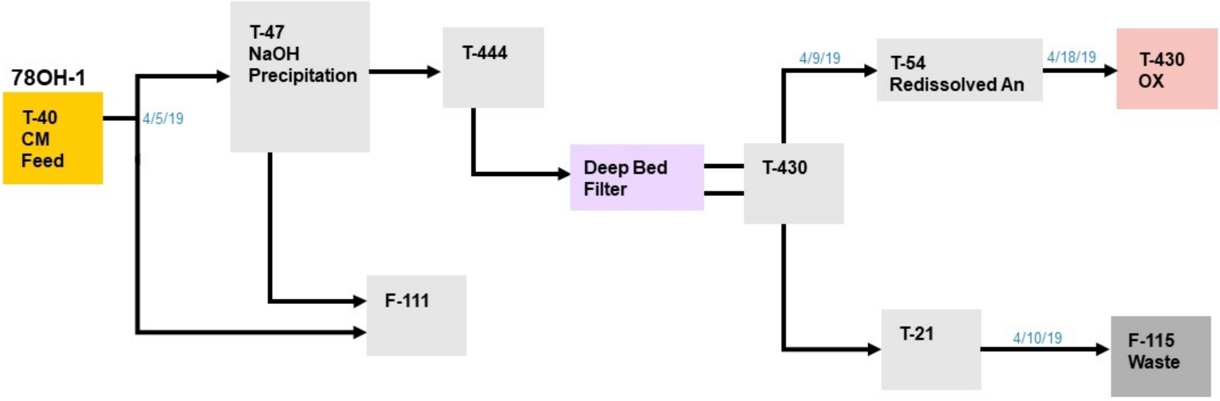


Figure 14. A flowchart showing a schematic of OH-1.

8.2 AMERICIUM/CURIUM HYDROXIDE PRECIPITATION [78OH-2]

April 10, 2019–April 16, 2019

Cleaning and recharging the DBF filter began on April 10. During this process a leak in the DBF was discovered; upon trying to fix it, the cable on the finger of the left manipulator broke. In parallel, T-40 was sparged for about 15 min before being transferred to T-47. This path was then washed with water, sparged for 30 min, and sampled as OHPF-056 (#3408). Next, NaOH (8 L, 10 M) was added to T-47 and the pathway was rinsed with water (2 L) twice, sparged for 30 min, and then left to settle for 2 h. Once the DBF was ready, T-47 was transferred to T-21 through the DBF. Sodium hydroxide (4.5 L, 2 M) was added to T-47 and the tank was sparged for 30 min before it was left to settle. T-47 was transferred to T-21 through the DBF. T-47 was then rinsed twice with NaOH (4.5 L, 2 M) followed by a water rinse which were added to T-21. Then, HNO_3 (2.5 L, 8 M) followed by two rinses with water (500 mL) were added to T-47 and sparged for 60 min. T-47 was then transferred through the DBF to T-54 to dissolve actinide hydroxides and convert them into their nitrate form. The path was rinsed with additional HNO_3 (2.5 L, 1 M) three more times. The following samples were sent for chemical analysis and results are shown in Table 18.

Table 18. Sample results taken during OH-1 and OH-2.

Tank	Sample ID	Date	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)
T-47	OHPF-055	4/8/2019		1.17	0.202	19.6	5.68	0.177
T-47	OHPF-056	4/11/2019	0.368	0.755	0.130	12.9	3.75	0.269
T-54	OHDP-034	4/14/2019	0.181	1.48	0.254	30.7	8.92	0.226
T-40	MSA-483	4/14/2019	0.001	0.0008	0.0001	0.0127	0.0037	
T-21	OHFL-075	4/14/2019	0.0122	0.005	0.0086	0.0356	0.0103	0.0092
T-47	MSA-484	4/14/2019	0.0005	0.0005		0.0020	0.0006	0.0014

Blanks in the table refer to either values smaller than 1×10^{-5} or the sample was not analyzed for a specific isotope.

T-54 was then set to evaporate down to 5 L. Follow-up operations continued by flushing numerous tanks, such as T-47, T-40, T-21 to F-115. A flowchart of the process flow of OH-2 is shown in Figure 15.

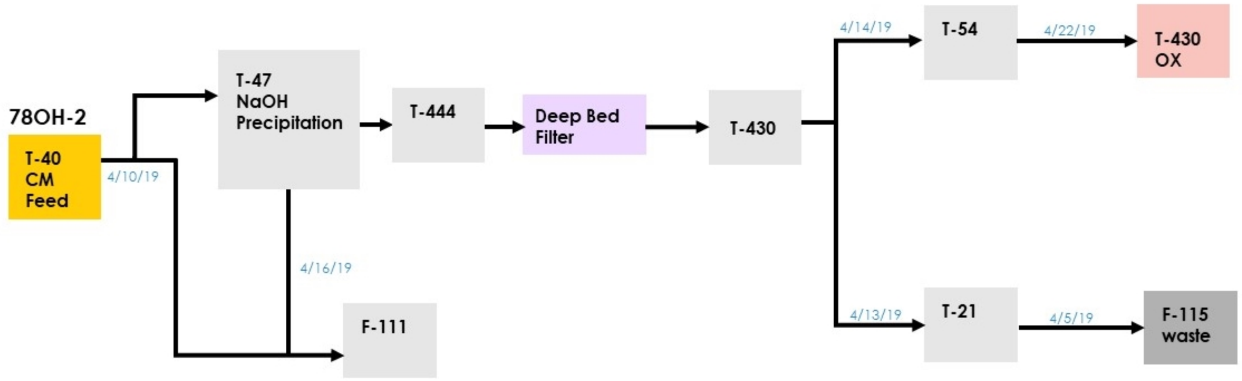


Figure 15. A flowchart showing a schematic of OH-2.

9. AMERICIUM/CURIUM OXALATE PRECIPITATION [78OX]

9.1 AMERICIUM/CURIUM OXALATE PRECIPITATION [78OX-1]

April 15, 2019–April 21, 2019

Reagent makeups and equipment preparation began on April 15th. T-54 was set to evaporate to 1 L and then cooled. Water (9 L) was added to T-54 and sparged for 1 h before a sample OXPF-446 #3352) was sent for chemical analysis. T-54 was in the process of being evaporated down to 0.5 L when it started to become clogged. The evaporation was terminated, and water (9 L) was added and sparged for 1 h. T-54 was evaporated down to 0.5 L; then, additional water (9 L) was added and OXPF-447 (#3401) was taken as a QC sample for analysis. The solution was sparged for 1 h and evaporated to 0.5 L again. The heating was shut down again because probes clogged. Additional water (10 L) was added and the tank was sparged for 1 h. The sample continued to evaporate down, and water was added until sample OXPF-448 (#3406) confirmed it had reached 0.6 M. Then, NH_4OH (146 mL) and water (800 mL) were added to T-54 and sparged for 30 min. T-54 was then evaporated to 4 L, but during evaporation, the probes clogged, and the heat was turned down to 75°C. The probes were blown out to clear them and then heating was resumed. The probes that clogged repeatedly were cleared to continue the solution evaporation down to 3.6 L. Water (400 mL) was added to T-54 and it was sparged for 30 min. Approximately 2 L of feed was sent to T-451 and flushed with water (0.166 L) three times. T-451 was set to heat to 70°C and oxalic acid (1.0 L, 0.8 M) was added. The solution was stirred for 15 min and was left to cool. T-451 was transferred to T-456, but after the addition of 300 mL, the discharge line became clogged. Vacuum was applied to free the plug and the rest of T-451 was flushed to T-456 with three water (500 mL) washes. Then, HNO_3 (375 mL, 15.8 M) was added to T-451 and it mixed for approximately 15 min to dissolve any precipitate. T-451 was then flushed to T-458 with two additions of water (300 mL). The solution in T-458 was washed back and forth two more times with water (300 mL) from T-458 to T-451, where it stayed.

The second cycle of the oxalate precipitation started on April 19th. T-451 was stirred and NH_4OH (286 mL, 14 M) was added. The addition was rinsed with water (100 mL) and was allowed to mix for 15 min before it was sampled as OXPF-449 (#3911). Results indicated that the molarity of the solution was 1.65 M and therefore it was advised to mix and resample. Analysis of the next sample, OXPF-450 (#3919), showed that the molarity was now 1.41 M. The solution was adjusted by adding NH_4OH (100 mL, 14 M) and water (100 mL), and the tank heated to 65°C. Once at temperature, oxalic acid (1 L, 0.8 M) was added to T-451 followed by the wash solution in M-450 (400 mL) and water (300 mL). The tank continued to mix at temperature for 30 min before it cooled. T-451 was transferred to T-458 along with three water (375 mL) rinses and the T-458 was mixed for 15 min. T-451 was rinsed with additional HNO_3 (1 L, 8 M) and water (2 L), which was then pumped to T-43, which was used to collect the Cm product. Solution was then pulled from T-458 back to T-451 in batch 1, which contained 2.55 L. The tank was mixed with NH_4OH (65 mL, concentrated) followed by water (1.56 L), which was added to T-451 and mixed for 10 min until it was transferred and filtered into T-40. The second batch (2.55 L) of solution was transferred from T-458 to T-451, where it was mixed. Then, NH_4OH (65 mL, concentrated) and water (1.56 L) were added to T-451, where it mixed for another 10 min; then, the solution was transferred and filtered into T-40. This was repeated with the third (1.0 L from T-458 and 1.6 L from T-456), fourth (2.6 L from T-456), and fifth (1.7 L from T-456) batches. Refiltration and dissolution began by adding HNO_3 (500 mL) to T-456, which was transferred to T-451. Acid was then pumped from T-451 accidentally to T-457 instead of T-458. The acid was then pumped from T-457 to T-458, where it was sampled as OXDP-772 (#3967, Table 19) before it was set to evaporate down to 15 L. A flowchart of the process flow of OX-1 is shown in Figure 16.

Table 19. Sample results from OX-1.

Tank	Sample ID	Date	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)
T-458	OXDP-772	4/20/2019	0.0101	0.0422	0.0073	0.637	0.185

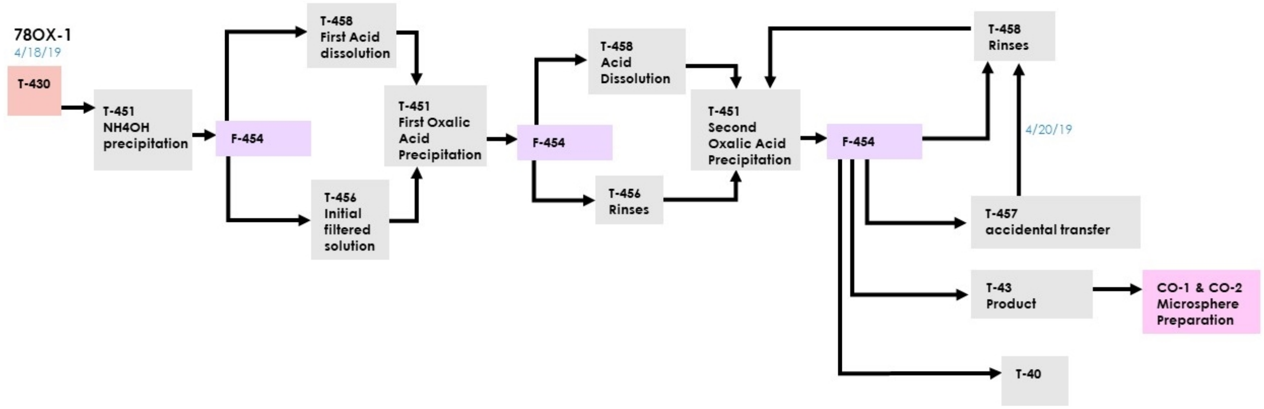


Figure 16. A flowchart showing the process flow of OX-1.

9.2 AMERICIUM/CURIUM OXALATE PRECIPITATION [78OX-2]

April 22, 2019–April 25, 2019

The feed transfer of T-54 to T-451 began on April 22 by transferring 1 L of feed and four water (0.413 L) rinses for a total of 2.8 L. T-451 was heated to 70°C and oxalic acid (1.0 L, 0.8 M) was added followed by wash solution (400 mL). The tank mixed for 15 min before the heat was turned off. T-451 was then pumped to T-456, bypassing F-454, and was rinsed three times with water (500 mL). Then, HNO_3 (1.25 L, 6 M) was added to T-451, where it was mixed for 15 min before it was slowly transferred to T-458 through the filter F-454. Two water (300 mL) washes followed. Solution was then washed back and forth from T-458 to T-451 by water, and finally residing in T451. T-451 was mixed while NH_4OH

(420 mL, 14 M) was added followed by water (100 mL). The combination was mixed for 15 min and then a sample was taken to confirm the correct acid range OXPF-451 (#3931). T-451 was then heated to 70°C for 30 min before oxalic acid (1.0 L, 0.8 M) was added. Wash solution was added, and the mixture continued to heat for another 15 min before it was pumped to T-458 through filter F-454. Water was used to wash the path before HNO_3 (275 mL, 15.8 M) and water (275 mL) were added to T-451. The acid was allowed to slowly drain through F-454 to T-43. Additional HNO_3 (1 L, 8 M) and water (2 L) were separately added and washed to T-43. T-43 was then sampled as OXDP-773A, B, and C (Table 20) and set to evaporate to 5 L.

Table 20. Sample results taken during OX-2.

Tank	Sample ID	Date	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)
T-43	OXDP-773A	4/23/2019	0.398	1.82	0.313	28.5	8.26	0.0303
T-43	OXDP-773B	4/23/2019	0.398	2.06	0.354	31.0	9.00	0.0437
T-43	OXDP-773C	4/23/2019	0.398	2.00	0.343	31.1	9.03	0.0335
T-458	OXDP-774	4/24/2019	0.009	0.03	0.006	0.501	0.145	

Blanks in the table refer to either values smaller than 1×10^{-5} or the sample was not analyzed for a specific isotope.

Refiltration of oxalate precipitation run filtrates continued by pulling five batches from T-458 to T-451. Each batch was precipitated by concentrated NH_4OH (65–45 mL) and washed with water before being filtered through F-454 to T-40. Then, HNO_3 (500 mL, 15.8 M) was added to T-456, where it was sparged for 15 min before it was transferred to T-451 followed by water (400 mL). The solution was then slowly pumped to T-458 while stopping in three 5-min intervals to let the acid sit on the filter (F-454). T-458 was sparged for 30 min before it was sampled as OXDP-774 (#3923). Meanwhile, T-40 was set to evaporate to 15 L and was sampled for acidity as OXWR-218 (#3418).

Feed preparation for microspheres began by heating T-43 to 70°C while sparging until it boiled. This operation was put on hold several times because of "rabbit" transfers. A flowchart of the process flow of OX-2 is shown in Figure 17.

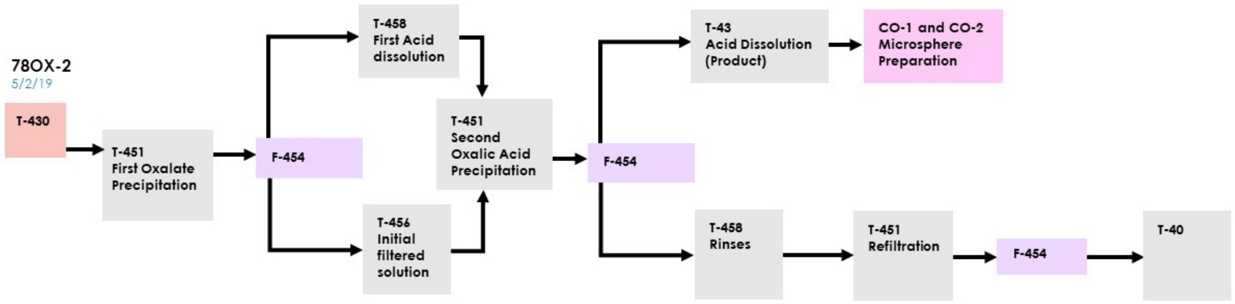


Figure 17. A flowchart showing the process flow of OX-2.

10. AMERICIUM/CURIUM MICROSPHERE PREPARATION [78CO]

The recovered Am and Cm is saved for future use in Cm target fabrication. Before this material can be stored it is made into AnO_x ($x = 1.5$ or 2) microspheres. These microspheres are synthesized by loading Am and Cm onto a Dowex 50W-8X cation exchange column where the resin is burned in air at 1,050°C. This allows for the formation of uniform spherical An oxide particles called microspheres [6].

10.1 MICROSPHERE PREPARATION [78CO-1]

April 25, 2019–May 20, 2019

Feed preparation continued by evaporating T-43 at 119°C for 5 h. Then, HNO_3 (8 L, 0.01 M) was added to T-43, sparged for 10 min, and then heated to 80°C–100°C for 30 min. A sample was taken as COAF-179 (#3449) of T-43 (Table 21). Additional acid adjustments were made with HNO_3 (80 mL, 15.8 M) and another sample was taken as COAF-180 (#3456) (Table 21). T-43 was evaporated down to 6 L and then cooled to room temperature. A new quartz column and resin were prepared and were leak-checked in Cub 4. Meanwhile, T-43 was heated to 75°C and cooled once it had evaporated to 20% of the starting volume. T-43 was sparged for 30 min; then, about 2.28 L was pumped from T-43 through the column C-448 to T-45. The discharge was switched from T-45 to T-43 just prior to Cm being eluted. An additional 2.93 L of feed was pumped through C-448 to T-43 rinsing three times with water (200 mL). The resin was pumped dry for 1 h; then, HNO_3 (500 mL, 8 *M*) was added to T-45 along with water (1 L) and then was sparged for 30 min and sampled as CORF-504 (#3346) (Table 21). Meanwhile, T-43 was evaporated down to about 2.5 L. While the column was being moved from LR4 into the furnace, the column holder broke. As a result, this operation was put on hold on April 30. A test run was successfully performed with a new column holder on May 15. The empty column test was removed from the furnace and the previously loaded column was placed in the new holder. The column was lowered into the furnace, and the furnace was started on May 17. While the furnace was heating, air was purged through the system until the temperature reached 850°C; then, the air was turned off and $Ar/H_2(4\%)$ was turned on for the hold at 850°C. During this operation, it was noticed that the $Ar/H_2(4\%)$ line was almost broken in half. Yellow tape was used to temporarily hold the line together for the rest of the temperature hold at 850°C. The furnace then continued heating to 1,050°C in air and held temperature for 16 h. Once cooled to 200°C, the air was turned off and the column was left to cool to room temperature. On May 20, the oxide as screened and weighed on a balance to be 19.12 g with a tap density of 8.4 mL. The oxide was then placed into a Twist-Lock Container "II" for storage.

Table 21. Samples taken during CO-1.

Tank	Sample ID	Date	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)
T-43	COAF-179	4/27/2019	0.205	1.39	0.239	25.2	7.33
T-43	COAF-180	4/28/2019	0.796	1.82	0.313	29.4	8.54
T-45	COAF-504	5/1/2019	0.016	0.005	0.0009	0.0045	0.0013

10.2 MICROSPHERE PREPARATION [78CO-2]

May 2, 2019–May 26, 2019

Equipment and leak checks began on May 2nd. Meanwhile, T-43 was diluted with water (6.5 L) and sparged for 1 h before it was sampled as COAF-181 (#3358). T-43 was then set to evaporate down to 1.031 L.

Feed transfer and resin loading began on May 16th by loading a new column with fresh resin. The column was leak-checked and washed with water before the feed (1.14 L) was loaded onto the column via T-444 and collected in T-45. The discharge was switched to T-43 just prior to Cm breakthrough and the rest of the feed was loaded. The column was washed with water (200 mL) three times before it was pumped dry for 1 h. Both T-43 and T-45 were flushed with HNO_3 and water before they were sampled for analysis as CORF-505 (#3386) and CORF-506 (#3447), respectively (Table 22). On May 20, the column was loaded into the furnace using the new column holder. It was heated to 850°C in air before it was switched to

Ar/H_2(4%) and held for 4 h. The gas was then switched to air for another 4 h before the furnace was heated to 1,050°C in air and held for 16 h before cooling. The oxide was weighed out on May 23 to be 18.99 g with a tap density of 8.8 mL. The oxide was screened (sieved) and accidently added to the container holding CO-1. The oxide was then rescreened and weighed to be 37.92 g before it was placed back into Container "II."

Table 22. Samples taken during CO-2.

Tank	Sample ID	Date	Pu (g)	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)
T-45	COAF-505	5/17/2019	0.0168	0.0054	0.0009	0.0017	0.0005
T-43	COAF-506	5/17/2019	0.0214	0.0066	0.0011	1.04	0.303

Flushing of CO equipment was performed by back-flushing water from T-43 back through T-444 and then back to T-43 again. T-43 was then set to evaporate for freeboard. The route was then rinsed back and forth again with HNO_3 (0.5 L, 1 M) and water before it was set to evaporate to less than 5 L. The material in T-43 was considered rework and was transferred to T-40 with water (500 mL) followed by HNO_3 (4 L, 2 M) and two water rinses (4 L). T-40 was set to evaporate to less than 5 L. A flowchart of the process flow of CO-1 and CO-2 is shown in Figure 18.

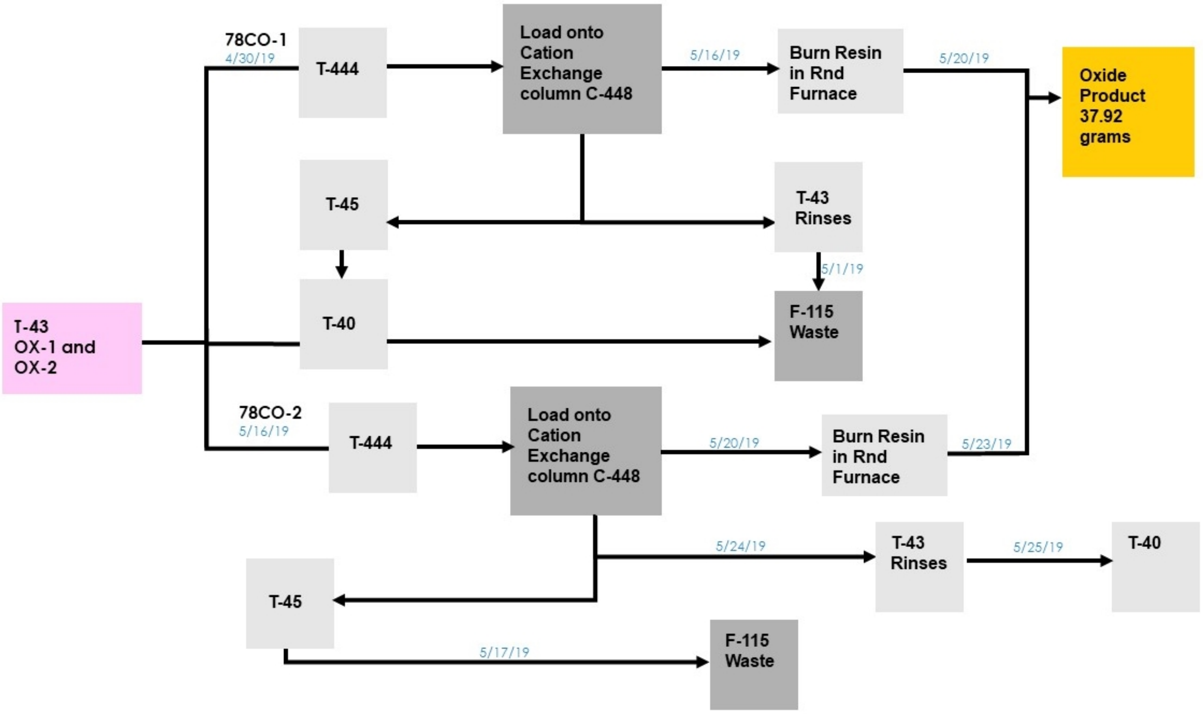


Figure 18. A flowchart showing the process flow of CO-1 and -2.

11. C78 REWORK [78]

11.1 CLEANEX REWORK [CY19F115CL-1A]

March 6, 2019–March 11, 2019

Extractant (30 mL) was added to F-115 with two water rinses (2.5 L). F-115 was sparged with air for 1 h and was sampled as SFW-942 (#3284) to confirm an acidity of 0.31 M. Additional NaOH (14.5 L, 19 M) was added to F-115 to adjust the molarity to 0.1 M. Another acidity adjustment was made by adding NaOH (14.4 L, 19 M) with two water (2.5 L) washes. F-115 sparged for 1 h before a sample was taken as SFW-943 (#3307) and had a molarity of 0.12. NaOH (4.92 L, 19 M) and water (5 L) were added to F-115. The tank sparged for 1 h and another sample (SFW-944) was sent for analysis of acid concentration. Two more additions of NaOH (2.27 L, 19 M) and water (2.5 L) were added to F-115. The tank was sparged for 1 h and sampled as SFW-945 (#3323) to give a molarity of 0.023 M. One more addition of NaOH (0.56 L, 19 M) and two water rinses (2.5 L) were added to F-115, sparged, and sampled as SWF-946. Sample results met disposal limits. F-115 was flushed to F-126 on March 11.

11.2 CLEANEX REWORK [CY19T23CL-1A]

February 20, 2019–March 5, 2019

Rework began on February 20 by making HDEHP solutions. T-23 was sparged with air for 30 min and left to settle for 15 min before it was sampled as CLER-376 (#3183). Sample results conveyed an acid concentration of 0.46 M. To adjust the acid concentration, NaOH (5.47 L, 5.15 M) was added to T-23. The tank was sparged for 1 h before a sample was taken as CLER-377 (#3215). Acidity was confirmed to be 0.12 M and another adjustment to 0.05 M was performed using NaOH (1.15 L, 5.15 M) before it was sparged and resampled as CLER-378 (#3122). The next acid adjustment was made to 0.03 M using NaOH (808 mL, 5.15 M) followed by two water (250 mL) flushes. T-23 was sparged with air for 1 h and sampled as CLER-379 (#3213) and confirmed a normality of 0.036. Because of the high activity of the sample, an additional acid adjustment to 0.02 N was made using two additional NaOH (260 mL, 5.45 M) washes and two rinses with water (250 mL). It was then sampled as CLER-380 (#3196). T-23 (91 L) was transferred to T-21 and flushed with water (1 L) on February 28. T-21 was then reacidified using HCl (5.8 L, 8 M) and two flushes of water (1 L). The tank was sparged with air for 1 h before a sample was taken as CLER-381(#3209).

On March 5, T-21 was jetted to F-115. T-21, rinsed twice with water (10 L) and sparged for 10 min between transfers to F-115. HCl (5 L, 2 M) was added to T-23 with two water (1 L) rinses.

11.3 CLEANEX REWORK [CY19T23CL-1B]

March 25, 2019–April 2, 2019

T-23 was sparged with air for 30 min and sampled as CLER-383 (#3321) to give a molarity of 0.30 M. To adjust acidity to 0.1 M, NaOH (2.64 L, 5.45 M) and two water (2.5 L) rinses were added to T-23. The tank was sparged for 1 h and sampled as CLER-384 (bottle #3282) to yield an acidity of 0.13 M. Additional NaOH (1.10 L, 5.45 M) and water (5 L) were added to T-23, sparged, and sampled as CLER-385 (#3312). To reduce acidity, more NaOH (0.4 L, 5.45 M) was added to T-23 and sampled as CLER-386 (#3320). Acidity was confirmed to be 0.026 M and the gross alpha was 611 counts/s. The aqueous phase of T-23 was transferred to T-21 via T-607 on April 1. T-21 was then acidified by adding HCl (3.27 L, 12 M) and water (2 L). T-21 was then sparged with air for 1 h and sampled as CLER-387 (#3338). On April 2, T-21 was flushed to F-115 with two flushes of water (10 L).

Then, HNO_3 (5 L, 2 M) and two flushes of water (1 L) were added to T-23, which was sparged with air for 1 h.

11.4 CLEANEX REWORK [CY19F115CL-1B]

April 24, 2019–April 30, 2019

F-115 was sparged for 1 h with air before it was left to settle for about 10 min. A sample for analytical analysis was taken as SFW-947 (#3398). To reacidify F-115, HNO_3 was added from MUA-P10-004 (3.5 L, 8 M), MUA-P25-019 (25 L, 5.5 M), and an additional 209 mL of concentrated HNO_3. The route was flushed twice with water (5 L) before additional HNO_3 (5 L, 1 M) was added through C4C601 and again with two water (2.5 L) rinses. F-115 was then sparged with air for 1 h and was left to settle for 1 h. An attempt was made to collect a sample, but no phase separation was visible. The tank was left to settle for another 1 h before a sample was taken as SFW-948 (#3348) which indicated a molarity of 0.03 M. The tank continued to be acidified by adding HNO_3 (9.77 L, 15.8 M) and two water rinses (2.5 L). The tank was sparged and sampled as SFW-949 (#3453) to yield an acid concentration of 0.12 M. NaOH (4.65 L, 19 M) and two water (2.5 L) rinses were added to F-115. The tank was sparged and sampled as SFW-950 (#3396) followed by another base adjustment with NaOH (1.94 L, 19 M) and water (5 L), and it was sampled as SFW-951 (#3392). On April 30, F-115 was jetted to F-126, stopping once F-126 reached 3,100 L.

11.5 CLEANEX REWORK [CY19T23CL-1C]

May 9, 2019–May 16, 2019

On May 9, T-66 was flushed to T-23 via T-607 with three additions of water (500 mL). The following bottles were also transferred to T-23, washing each bottle twice with water: C78-SS-1 Strip #3, C78-SS-2 Strip #1, C78-SS-1 EXT, C78-SS-1 Scrub #2, C78-SS-2 Strip #2, C78-SS-2 Scrub #2, C78-SS-2 Scrub #1, C78-SS-1 Strip #1, C78-SS-2 Strip #2, C78-SS-1 Strip #3, and C78-SS-1 Scrub #1. After all of the additions, T-23 was sparged for 30 min and was left to settle for 15 min before a sample CLER-388 (#3349) was collected. pH adjustments began by adding NaOH (3.35 L, 5.53 M) and two water (250 mL) rinses. The tank was sparged and a sample was collected as CLER-389 (#3898) to check acidity. Additional adjustments were made by adding more NaOH (2.42 L, 5.45 M), which was collected as sample CLER-390 (#3886), followed by another addition of NaOH (396 mL, 5.45 M) sampled as CLER-391 (#3424). The next adjustment was to 0.02 M by adding NaOH (217 mL, 5.45 M), which was sampled as CLER-392 (#3374) and was reported to be in good range of 0.02 M. T-23 (94 L) was then transferred to T-21 on May 14 followed by water (500 mL). T-21 was then acidified using HNO_3 (3.045 L, 15.8 M) and was sampled as CLER-393 (#3410). T-21 was then jetted to F-115 and rinsed twice with water (10 L).

11.6 CLEANEX REWORK [CY19T23CL-1D]

May 21, 2019–May 28, 2019

T-23 was sparged for 30 min until a sample was submitted as CLER-394 (#3884). pH adjustments began by adding NaOH (2.45 L, 5.45 M) and water (5 L); the tank was sparged and sampled as CLER-395 (#3880). The next adjustment was made by adding NaOH (2.34 L, 5.45 M) and was sampled as CLER-396 (#3887). On May 25, T-23 was transferred to T-21. To acidify T-21, HNO_3 (1.96 L, 15.8 M) was added and sampled as CLER-397 (#3888), and confirmed it was within limits to dispose of. T-21 was jetted to F-115 with water (20 L).

11.7 CLEANEX REWORK [CY19T23CL-1E]

May 29, 2019–June 21, 2019

T-23 was sparged for 30 min and sampled as CLER-398 (#3560). T-23 was adjusted with NaOH (2 L, 5.45 M) sparged with air for 1 h and sampled as CLER-399 (#3556). An additional adjustment was made with NaOH (330 mL, 5 M) and was sparged and sampled as CLER-400 (#3555). The expected sample activities were not met. Additional NaOH (0.175 L, 5 M) was added with two water rinses (250 mL) and the tank was sparged and sampled as CLER-401 (#3516) and activity limits were not met. Therefore, additional NaOH (0.205 L, 5.55 M) and water (5 L) were added and sparged before it was sampled again as CLER-402 (#3875). Activity limits were met. On June 6, the T-23 aqueous phase was transferred to T-21 and the organic phase was transferred to T-72. T-21 was then sparged for about 30 min before it was sent for chemical analysis as CLSR-946 (#3462). Meanwhile, T-72 was stripped and sent to T-40 starting on June 9.

11.8 CLEANEX REWORK [CY19T115CL-1C]

June 3, 2019–June 10, 2019

F-115 was sparged with air for 1 h and sampled as SFW-952 for an acidity of 0.19 M. To adjust the acid concentration, NaOH (10.15 L, 19 M) was added and a sample was taken as SFW-953 (#3552). Additional NaOH (3.45 L, 19 M) was added and F-115 was sparged before a sample was taken as SFW-954 (#3430) to assess acidity and activity. A sparge and resample was performed as SFW-955(#3528). On June 10, about 1,290 L of solution was transferred from F-115 to F-126.

12. DISCUSSION

TCOMP and ORIGEN codes were used to predict isotope yields before and after irradiation and are shown in Table 23 shaded in purple. In comparing the calculated values with the recovered values, most estimations seem to fall within an acceptable range. A direct comparison without process hold-up is not possible because of limitations of measuring and assaying the targets before dissolution. In general, in comparing TCOMP and ORIGEN with the 78CL feed material, the amount of Cm was underestimated and the amounts of Bk, Cf, and Es were slightly overestimated. However, when decay and process holdups are taken into account, these numbers provide a good estimate of the isotopes that were produced in the HFIR.

Historical campaign data were used to predict the isotope quantities for C78 (see Appendix A). Based on this, it was predicted that chemical processing would yield about 1 g of Am, 25 g of Cm, 12 mg of ^{249}Bk, 85 mg of ^{252}Cf, and 1 µg of ^{254}Es. Though most of these estimates seem reasonable, the ^{252}Cf was roughly 10 mg overestimated because only 73.3 mg was available for the nut package feed. Overall, TCOMP and historical data give a good estimation of isotope yield produced in the HFIR and the final yield of isotopes post-processing. In general, based on the TCOMP estimates after separation, this campaign had a final Cf-252 yield of roughly 78%.

Tracking process hold-ups throughout the campaign can be done by comparing feed solutions to product solutions as seen in Table 24. The largest mass differences of Cf are attributed to the 78CM LiCl anion exchange process and the 78CX cation exchange process which account for roughly 15 mg of Cf-252 with 6 mg from the 78CM process and 9 mg from 78CX. Though these processes have proven to be robust since their implementation in the 1960's, very little has been done since then to improve on their efficiency. Small scale studies on alternative processing methods by Laetitia Delmau have shown promising results [8]. These new methods will be tested on C79 material for further assessment of their viability in our processes. Future campaigns will continue to track differences and provide insights into how to make process improvements and how to improve our modeling predictions.

Table 23. A comparison of TCOMP and ORIGEN calculations for the C78 irritated targets.

Analysis method or code[+]	Decay date	Am (g)	Cm (g)	^{244}Cm (g)	^{246}Cm (g)	^{248}Cm (g)	^{249}Bk (mg)	^{252}Cf (mg)	^{253}Es (µg)	^{254}Es (µg)
TCOMP	12/27/2018	0.355	24.03	6.51	13.95	3.12	18.67	99.59	206	2
DSDP-621A-C[*] (78CL feed)	1/24/2019	0.36	26.4	7.38	15.27	3.31	16.0	91.7	94.2	
ORIGEN	12/27/2018	0.363	24.1	6.54	14.0	3.13	20.5	98.4	203	3.44
TCOMP (after separation)	3/27/2019	0.355	23.97	6.45	13.95	3.12	15.36	93.34	15	2
CXCF-828 (nut package feed)	3/29/2019							73.3		
COAF-180 (Cm for C79)	4/28/2019	1.82	29.4	8.54	16.6	3.65				

[+] ORIGEN is the Oak Ridge Isotopic Generation and Depletion Code [9], and TCOMP is the Transmutation Computation Code [10], [11]. 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.

[*] Represents an average where multiple samples were taken of the same product stream.

Blanks in the table refer to either values smaller than 1×10^{-5} or the sample was not analyzed for a specific isotope.

Table 24. Major products from each processing step in C78.

Process	Sample code	Date	Am (g)	^{241}Am (g)	Cm (g)	^{244}Cm (g)	^{252}Cf (mg)	^{249}Bk (mg)	^{253}Es (µg)
CL feed	DSDP-621A-C[*]	1/24/2019	0.36	0.049	26.4	7.38	93.2	15.8	94.2
CM feed	CLCP-452A-C[*]	2/8/2019	1.61	0.496	32.8	9.51	92.1		62.35
CM-3 Product	CMBP-166	2/19/2019	0.257	0.044	0.107	0.031	85.9	10.9	39.7
CM-3-1 LiOH Product	CMBP-167A-B[*]	2/26/2019	0.055	0.0095	0.079	0.023	63.1	11.15	26.4
CM-3-2 LiOH Product	CMBP-169A-B[*]	3/7/2019	0.019	0.003	0.027	0.008	19.6	3.16	3.7
CX Cf product (nut package feed)	CXCF-828	3/29/2019			0.0034		73.3		
BX feed	BXBK-084	3/31/2019	0.001	0.0003	0.0003		0.913	13.4	
D-110 nut package	C78CFD110	4/1/2019					25.824		
D-111 nut package	C78CFD111	4/1/2019					27.557		
D-112 nut package	C78CFD112	4/1/2019					19.639		
OH product	OHDP-034	4/14/2019	1.48	0.254	30.7	8.92	0.226		
Oxalate product	OXDP-773A-C[*]	4/23/2019	1.96	0.336	30.2	8.76	0.036		
BX product	BXBK-085	4/26/2019						13.8	
Microsphere feed	COAF-180	4/28/2019	1.82	0.313	29.4	8.54			

13. REFERENCES

1. Mosley, W. C., Smith, P. K., and McBeath, P. E. 1975. "Neutron Sources of Palladium ^{252}Cf Oxide Cermet Wire." *Proceedings of the American Nuclear Society National Topical Meeting*, Austin, Texas, September 11–13, 1972.
2. King, L. J.; Bigelow, J. E.; Collins, E. D. 1981. "Experience in the Separation and Purification of Transplutonium Elements in the Transuranium Processing Plant at Oak Ridge National Laboratory" Chapter 8 of ACS Symposium Series 161.

3. Bigelow, J. E.; Collins, E. D.; King, J. L. "The Cleanex Process: A Versatile Solvent Extraction Process", ACS Symposium Series 117, 1980, 147-155.
4. Collins, E. D., et al. "Multigram Group Separation of Actinide and Lanthanide Elements by LiCl-Based anion Exchange", Chapter 9, ACS Symposium Series 161, 1981, 147-160.
5. Benker, D. E. et al, "Chromatographic Cation Exchange Separation of Decigram Quantities of Californium and Other Transplutonium Elements", ACS Symposium Series 161, Chapter 9, 1980, 161-171.
6. Chattin, F. R. et al, "Preparation of Curium-Americium Oxide Microspheres by Resin-Bead Loading", ACS Symposium Series 161, Chapter 11, 1980, 173-185.
7. Cross, J. N., Su, J., Batista, E. R., Cary, S. K., Evans, W. J., Kozimor, S. A., Mocko, V., Scott, B. L., Stein, B. W., Windorff, C. J., and Yang, P. 2017. "Covalency in Americium(III) Hexachloride." Journal of the American Chemical Society 139 (25), 8667–8677.
8. Lemonick, S. Chemical & Engineering News. Cleaning up and simplifying californium production https://cen.acs.org/physical-chemistry/nuclear-chemistry/Cleaning-simplifying-californium-production/97/i34 (accessed December 30, 2020).
9. Croff, A. G. 1983. "ORIGEN2: A versatile computer code for calculating the nuclide compositions and characteristics of nuclear materials." *Nuclear Technology* 62, 335–352.
10. Hogle, S., Maldonado, G. I., Gauld, I., and Ezold, J. G. 2010. "Calculating transcurium production yields at the High Flux Isotope Reactor." *Transactions of the American Nuclear Society* 103, 766–767.
11. Bigelow, J. E., et al. 1981. "Production of transplutonium elements in the High Flux Isotope Reactor." In *Transplutonium elements—Production and recovery*, J. Navratil et al., eds., ACS Symposium Series. Washington DC: American Chemical Society.

APPENDIX A. C78 PLAN

Date: December 27, 2018

To: Distribution

From: D. E. Benker

Subject: **Plans for Campaign 78: Processing of Four AmCm Targets plus Rework Materials**

A group of four irradiated targets were discharged from HFIR on September 28, 2018 after cycle 482. Chemical processing is scheduled to begin on January 21, 2019 and Table 1 lists the material estimated in the targets plus rework materials that will be processed in Campaign 78. A tentative schedule for the various processing steps is shown in Table 2. The time scheduled for the various steps include 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 Appendix 1. The goal is to complete the processing steps in a timely manner to replenish the californium stock for the Isotope Sales Program, provide berkelium and einsteinium products for science projects, convert the recovered americium-curium into suitable oxide for fabrication of new targets for HFIR irradiation, and allow the quick resumption of other scheduled work. As final products, we hope to recover about, 1 g of americium, 25 g of curium, 12 mg of ^{249}Bk, 85 mg of ^{252}Cf, and 1 µg of ^{254}Es.

Table 1. Feed Material for Campaign 78, adjusted to February 1, 2017

Item	^{131}I, Ci[a]	^{241}Am, g	^{243}Am, g	Cm, g	^{244}Cm, g	^{249}Bk, mg	^{252}Cf, mg	^{254}Es, µg
AmCm Targets[b]	0.2		0.3	24.0	6.5	17	97	2.2
Target Fabrication Rework[c]		0.1	0.3	5.0	1.6			
Campaign 77 Rework		0.2	0.9	4.9	1.5		7	
Total	0.2	0.3	1.5	33.9	9.6	17	114	2.2

[a] It is estimated that ~0.002 Ci will be generated per day from the fissions of ^{252}Cf until the ^{252}Cf is encapsulated for transfer to Building 7930.

[b] S-53, S-54, S-55, and S-57.

[c] Thirty reject pellets

Table 2: Schedule for Campaign 78

Date	Process	Purpose
01/21/19 to 01/27/19	Target Dissolution (78DS-2)	Separate the actinides from the Al target material and place in nitric acid solution
01/27/19 to 02/10/19	Cleanex Batch Solvent Extractions (78CL-1)	Remove chemical impurities, most fission products, and plutonium
02/10/19 to 02/26/19	LiCl Anion Exchange (78CM-1 & 2)	Separate AmCm, transcurium elements, and residual lanthanides
02/26/19 to 03/01/19	LiOH Precipitation (78CM-2)	Convert transcurium product from LiCl solution to small volume of dilute HNO_3 solution
03/01/19 to 03/08/17	High-Pressure Ion Exchange (78CX-1 & 2)	Separate Es, Cf, Bk, and AmCm into individual product fractions
03/11/17 to 03/20/19	Cf Package Loading (78CF-1, 2, & 3)	Load Cf onto storage packages suitable for transfer to Building 7930
03/08/19 to 03/25/19	Berkex (78BX-1)	Remove residual Cf so that the Bk product can go to the alpha laboratories unshielded.
03/21/19 to 03/28/19	Cf Rework Consolidation	Decontaminate equipment and consolidate various rework solutions for recovery in Cleanex stage (T-23).
03/25/19 to 04/12/19	Hydroxide Precipitation (78OH-1)	Conversion of AmCm product from LiCl solution to a HNO_3 solution appropriate for combining with 76 AmCm product.
04/12/19 to 04/19/19	Oxalate precipitation (78-OX-1, & 2)	Final purification of the AmCm.
04/19/19 to 05/03/19	AmCm oxide conversion (78-CO-1, 2, & 3).	Load AmCm on Dowex resin and calcine resin to make oxide product
05/03/18 to 05/08/19	Rework roundup	Equipment cleanup
05/03/1 to 05/31/19	F-115 & T-23 Recovery	Recovery of trace actinides from low level process streams.

Appendix 1: Description of Processing Steps for Campaign 78

Dissolution of Scrap aluminum (78DS-1)

The coolant flow-shrouds from the four 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 6.2 L of 2.1 *M* $NaNO_3$. The dissolver will be heated to 92°C and 2.6 L of 10 *M* NaOH will be metered to the dissolver (<2.5 L/h) to affect 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 complete 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 (78DS-2)

After the scrap aluminum dissolution is completed, 11.3 L of 2.1 *M* $NaNO_3$ will be added to T-70 and the four irradiated targets will be charged for the caustic dissolution. In addition to the irradiated targets, reject Al-Cm rework from Campaign 77 will also be added. The dissolver solution will be heated to 92°C and 5.7 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 complete 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, 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 77 rework materials. T-70 and F-70 will be flushed to T-72 with 2 × 3 L of 4.0 *M* HNO_3, 2 × 3 L of 1.0 *M* HNO_3, and 2 × 3 L of 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 of 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 of water.

Purification of the transplutonium elements using batch solvent extraction (78CL-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, 1-5% of the solution will be diverted to a separate separation experiment that will be conducted in parallel. The product from this experiment is expected to be added to the Cleanex product. The remaining solution in T-72 will then 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 enough volume to cover the specific gravity 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 0.1 M sodium hypochlorite by addition of NaOH (5 M) and sodium hypochlorite (2.5 L of 5 wt. % available chlorine); the solution is then digested at 80°C for 30 minutes.

The adjusted feed solution in T-72 will be contacted with 20 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 <0.10-g ^{244}Cm (<1% 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 Alamine 336 to hold iron impurity 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. The product from the small-scale experiment, will be returned as an HCl solution and added to the Cleanex product.

Separation of lanthanides, americium-curium, and transcurium elements using LiCl chromatographic anion exchange (78CM-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. 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 a floor filter, and T-43 will be flushed to T-65 with 4 × 750 mL of 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 may also be changed depending on its condition. 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 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 78CM-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 of 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 of 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 transferred to Cub 4 for the start of the cation ion exchange (CX) runs.

Separation of actinides using AHIB (α-hydroxyisobutyrate) cation exchange (78CX-1 and 2)
The combined transcurium element product from the 78CM 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 concentrations and pH through the columns. The complexing strength for the elements is 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 determine which fractions contain Es, 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 runs (two to three) 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 of 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 of water, 300 ml of 0.3 *M* NH_4NO_3, and 80 mL of 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 typical elution sequence is (1) 220 mL of 0.25 *M* AHIB-pH 3.9 to elute the actinides off the loading column and onto the separation column, (2) 1.5 L of 0.25 *M* AHIB-pH 4.2 to elute the einsteinium and californium, (3) 700 mL of 0.25 *M* AHIB-pH 4.6 to elute the berkelium, (4) 450 mL of 0.50 *M* AHIB-pH 4.8 to elute americium-curium, and (5) 450 mL of water to flush the system free of any remaining reagent solutions. Approximately 10-12 cuts of varying volumes 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., Es, Cf, Bk, and AmCm product fractions or recycle.

The remaining 40% of the feed plus recycle fractions will be used in the second run. The run conditions and product handling will be like the first run. Depending on the quality of the Es/Cf/Bk separations, a third recycle run may be added. For the Es/Cf separation, customized lab-scale equipment may be used for the third cycle to remove californium.

Loading Cf onto storage/transfer packages (78CF-1 through 3)
The californium fractions from the CX runs will be composited and prepared for loading onto 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 Swagelok nut package for later transfer via the pneumatic transfer line to Building 7930 for further processing.

Berkelium purification using batch solvent extraction (78BX-1)
The berkelium product fractions from 78CX 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 transferred 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 finally removed with 500 mL of 8 *M* HNO_3, which is the desired feed acidity for Berkex.

The berkelium feed solution will then be adjusted to 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 of 8.0 *M* HNO_3–0.3 *M* $NaBrO_3$. Finally, the organic will be stripped with 2 × 280 mL of 8.0 *M* HNO_3–1 *M* H_2O_2. If a sufficient decontamination is achieved 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.

Removal of LiCl from the americium-curium product (78OH-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 concentrated and stored or used to make new targets. 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 that could be done if there are problems with the precipitation equipment, 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 is 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. If the deep-bed filter is filled with precipitate before the filtration or later flushing is completed, the inlet line will be back-flushed to T-47, and the filter flushed forward to T-20 with caustic and water. The filter discharge will be routed to T-54 and the precipitate dissolved with acid to T-54. The routes are reset to the original configuration and the filtration from T-47 to T-20 continued until all the filtrate and programmed flushes are completed.

The precipitate in T-47 will be washed with 3 × 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 × 2.5 L of 1 *M* HNO_3 and 3 × 2.0 L of water to T-54. T-54 must be boiled periodically to keep the volume <17 L. 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.

78-OX-1 and 2

The combined americium-curium product in T-54 will be adjusted to 4 L of 1 *M* HNO_3 for oxalate precipitation feed. One-half of the feed (16-g actinides) will be processed in each of two, 2-cycle oxalate precipitation runs. The oxalate product will be collected in T-43 for oxalate destruction and feed adjustment for the curium microsphere processing and the rework solutions will be collected in T-40 for subsequent oxalate destruction and recovery.

Approximately 2 L of feed solution will be transferred to T-451 via T-430 for the first oxalate precipitation run. T-451 will be heated to 60-65°C and 1.0 L of 0.8 *M* oxalic acid will be metered (~200 mL/min) from M-450 to T-451 while stirring the solution. A 400 mL wash (0.2 *M* oxalic acid - 0.5 *M* HNO_3) will be added to M-450 to clear the addition line to T-451. After 15 min at 60°C with stirring, T-451 will be cooled to the RCW temperature. The precipitate will be filtered through F-454 to T-456 and T-451 will be flushed with 3 × 500 mL of wash solution. The collected precipitate will be dissolved to T-458 with 750 mL of 8.0 *M* HNO_3 plus 2 × 300 mL water flushes. The solution in T-458 will then be transferred back to T-451 plus 2 × 300 mL water flushes for the second cycle precipitation.

For the second cycle precipitation, a calculated amount of NH_4OH will be added to T-451 to neutralize the excess acid and a sample will be pulled for analysis to determine the acid concentration. A final acid adjustment will be made if necessary and 1.0L of 0.8 *M* oxalic acid will be added to T-451 to precipitate the actinides again. The oxalic acid will be flushed in with 400 mL wash solution. The precipitate will be filtered via F-454 to T-458 and washed with 3 × 500 mL of wash solution. The precipitate will be dissolved to T-43 with 750 mL of 8.0 *M* HNO_3 and flushed with 1.0L of 8.0 *M* HNO_3 plus 2 L of water.

The accumulated filtrates will be treated and re-filtered to recover additional actinides for recycle to the next run. The filtrates collected in T-456 and T-458 will be transferred back to T-451 in several batches. Each batch will be adjusted with water and NH_4OH to adjust the oxalate and HNO_3 concentrations to conditions (~0.1 *M* each) where the actinides are less soluble than in the first-cycle conditions (0.2 *M* oxalate and 0.5 *M* HNO_3). The solution will be filtered through F-454 to the final filtrate tank (T-40). After all the batch filtrations have been done, 500 mL of conc. HNO_3 will be added to T-456, sparged and transferred to T-451 followed by a 400 mL water flush. The acid solution will then be transferred to T-458 via F-454 to dissolve any collected precipitate (plus 2 × 200 mL water flushes). Since the conditions used in this refiltration of the filtrates will also recover more impurities it is not considered a final product. This acid solution will be held in T-458 to be used to dissolve the first-cycle precipitate in the following run which receives another cycle of purification.

All the subsequent runs will follow the same processing steps with the americium-curium-lanthanide product collected in T-43 and the final filtrates collected in T-40. For the last run, the re-precipitation of the filtrates will be done to help minimize the amount of actinides in the filtrate and potentially allowing the disposal of the filtrates to waste. At the end of the run, extensive flushing of the rack (external and internal) and of Cubicle 4 will be made and the flushes will be collected in T-40. Once all the rework and waste streams that may contain oxalate have been collected, the solution will be digested in strong HNO_3 to destroy any leftover oxalate reagent and the actinides will be collected in the final rework recovery or discard to waste.

78-CO-1, 2, and 3

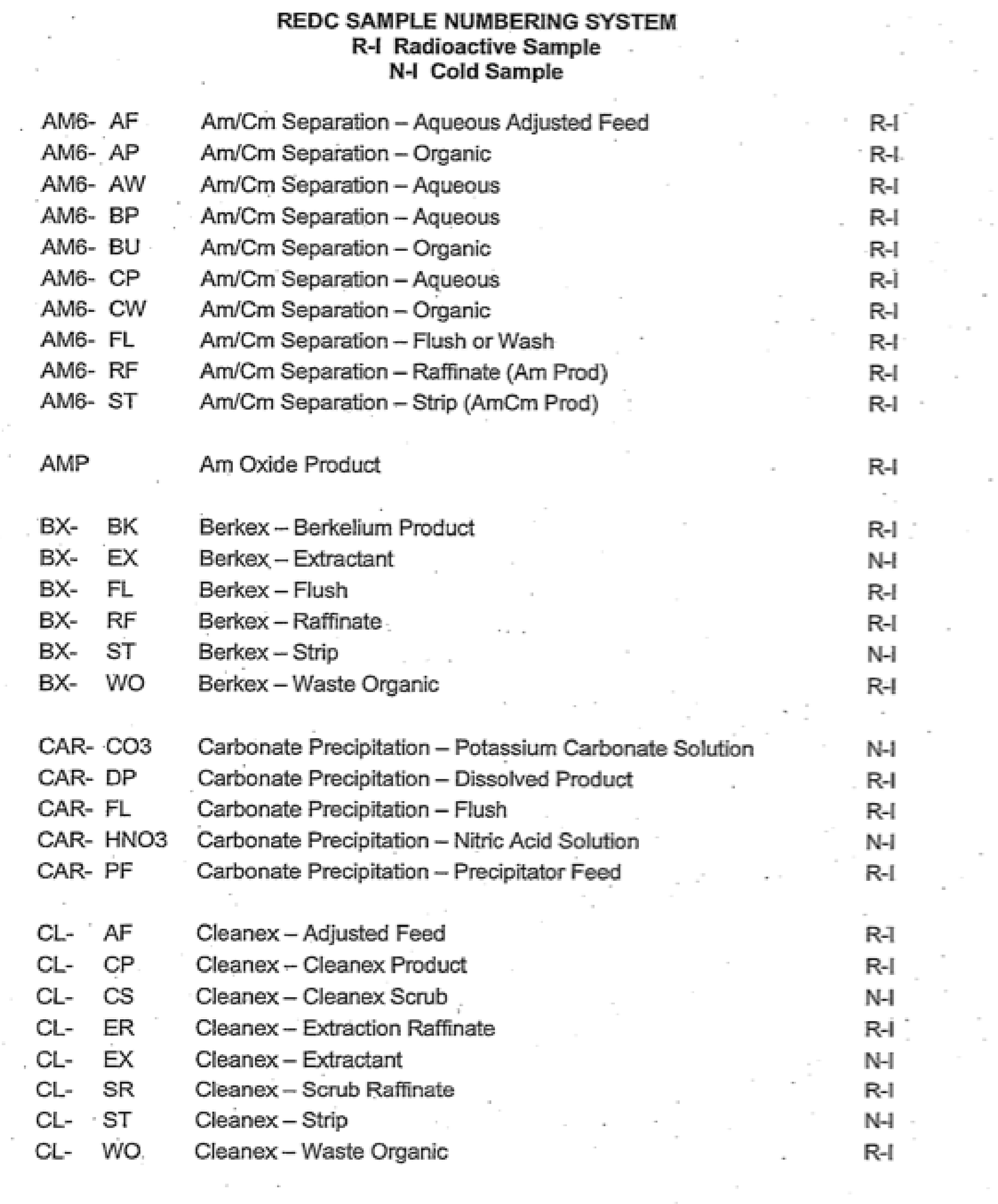
The product from the oxalate precipitation runs in T-43 will be adjusted to 5 to 10 g/L actinides and 0.25 *N* HNO_3. The adjusted feed will be processed in three batches to prepare microspheres for fabrication into recycle targets for irradiation at HFIR. A batch of feed solution will be passed through a fused silica glass column containing 120-140 mL of Dowex 50W-X8 resin, 60-80 µm, to sorb the actinides. The raffinate will be collected into T-45, sampled for curium content, and transferred to waste or rework. The resin will be loaded to breakthrough for each of the runs. When the loading band reaches within an inch of the bottom of the column the output is switched back to the feed tank. Once the column is fully saturated it is washed with 2 x 200 mL of water. On the final run, the amount of resin will be adjusted to sorb about 80% of the remaining actinide content. The 80% figure is used to help ensure that the resin is loaded to saturation by the actinides in the hope that the actinides will displace weakly loaded impurities off the column and make a purer product.

The loaded resin in the silica column will be transferred to the round furnace on the left rack of Cub 4 for pyrolysis of the resin matrix. The resin will be dried for 1.5 h at 150°C in the round furnace followed by an additional hour at 150°C with an air purge. The temperature will then be increased to 850°C and held for 4 h with an air purge; 4 h with a 4% H_2-Ar purge; and 4 h with an air purge. While continuing the air purge, the furnace will then be heated to 1050°C and held for 16 hr. The oxide product will then be screened (212 µm openings), weighed, sampled, measured for tap density, and the ^{244}Cm content measured by calorimetry. The curium oxide microsphere product will then be transferred to the Target Fabrication group for making targets for irradiation in HFIR. The feed heel and any dissolved oxide from the handling equipment should be considered clean and not be sent to the T-23 recovery stage (although they may be added to the final strip product from T-23 for storage).

Rework Collections (CY19F115CL-1 & CY19T23CL-1)
During the campaign, various rework solutions will be collected and processed in T-23 Cleanex extraction stage. The recovered actinides will be processed, cleaned up, and stored.

APPENDIX B. LIST OF SAMPLE CODES

Note: Some codes listed here may not appear in this report.



REDC SAMPLE NUMBERING SYSTEM
R-I Radioactive Sample
N-I Cold Sample

AM6-	AF	Am/Cm Separation – Aqueous Adjusted Feed	R-I
AM6-	AP	Am/Cm Separation – Organic	R-I
AM6-	AW	Am/Cm Separation – Aqueous	R-I
AM6-	BP	Am/Cm Separation – Aqueous	R-I
AM6-	BU	Am/Cm Separation – Organic	R-I
AM6-	CP	Am/Cm Separation – Aqueous	R-I
AM6-	CW	Am/Cm Separation – Organic	R-I
AM6-	FL	Am/Cm Separation – Flush or Wash	R-I
AM6-	RF	Am/Cm Separation – Raffinate (Am Prod)	R-I
AM6-	ST	Am/Cm Separation – Strip (AmCm Prod)	R-I
AMP		Am Oxide Product	R-I
BX-	BK	Berkex – Berkelium Product	R-I
BX-	EX	Berkex – Extractant	N-I
BX-	FL	Berkex – Flush	R-I
BX-	RF	Berkex – Raffinate	R-I
BX-	ST	Berkex – Strip	N-I
BX-	WO	Berkex – Waste Organic	R-I
CAR-	CO3	Carbonate Precipitation – Potassium Carbonate Solution	N-I
CAR-	DP	Carbonate Precipitation – Dissolved Product	R-I
CAR-	FL	Carbonate Precipitation – Flush	R-I
CAR-	HNO3	Carbonate Precipitation – Nitric Acid Solution	N-I
CAR-	PF	Carbonate Precipitation – Precipitator Feed	R-I
CL-	AF	Cleanex – Adjusted Feed	R-I
CL-	CP	Cleanex – Cleanex Product	R-I
CL-	CS	Cleanex – Cleanex Scrub	N-I
CL-	ER	Cleanex – Extraction Raffinate	R-I
CL-	EX	Cleanex – Extractant	N-I
CL-	SR	Cleanex – Scrub Raffinate	R-I
CL-	ST	Cleanex – Strip	N-I
CL-	WO	Cleanex – Waste Organic	R-I

CMP		Curium Oxide Product	R-I
CM-	AE	LiC1 AIX – Americium/Curium Elutriant	N-I
CM-	AF	LiC1 AIX – Adjusted Feed	R-I
CM-	AP	LiC1 AIX – Americium/Curium Product	R-I
CM-	BE	LiC1 AIX – Berkelium Elutriant (reserved, not yet used)	N-I
CM-	BP	LiC1 AIX – Transcurium Elutriant	R-I
CM-	CE	LiC1 AIX – Californium Elutriant	N-I
CM-	CF	LiC1 AIX – Column Flush	R-I
CM-	CR	LiC1 AIX – Column Raffinate	R-I
CM-	RE	LiC1 AIX – Rare-Earth Elutriant	N-I
CM-	SF	LiC1 AIX – Synthetic Feed	N-I

COG2		Cell Off-Gas – Charcoal Trap Sampler for ^{131}I	R-I

CO-	AF	Curium Oxide – Adjusted Feed	R-I
CO-	RF	Curium Oxide – Raffinate	R-I

CSA		Cold Sample Aqueous	N-I
CSO		Cold Sample Organic	N-I

CX-	AF	But Column – Adjusted Feed	R-I
CX-	AM	But Column – Americium Product	R-I
CX-	BK	But Column – Berkelium Product	R-I
CX-	CF	But Column – Californium Product	R-I
CX-	CM	But Column – Curium Product	R-I
CX-	ER	But Column – Elution Raffinate	R-I
CX-	ES	But Column – Einsteinium Product	R-I
CX-	FM	But Column – Fermium Product	R-I
CX-	LR	But Column – Loading Raffinate	R-I
CX-	SF	But Column – System Flush	R-I

DS-	DF	Dissolution – Dissolver Flush	R-I
DS-	DP	Dissolution – Dissolver Product	R-I
DS-	DS	Dissolution – Dissolvent Solution (reserved, not yet used)	N-I
DS-	JD	Dissolution – Jacket Dissolvent Solution	N-I
DS-	JW	Dissolution – Jacket Waste Solution	R-I

DS-	IC	Dissolver Off-Gas Iodine Retention System – Condensate	R-I
DS-	ICS	Dissolver Off-Gas Iodine Retention System – Caustic Scrubber Pot and/or Neutralized Scrubber-Column Effluent	R-I
DS-	IE	Dissolver Off-Gas Iodine Retention System – Analysis Stage Downstream of Scrubber Column	R-I
DS-	IF	Dissolver Off-Gas Iodine Retention System – Dissolver Solution	R-I
DS-	IS	Dissolver Off-Gas Iodine Retention System – Scrubber Effluent	R-I
MSA		Miscellaneous Sample Aqueous	R-I
MSO		Miscellaneous Sample Organic	R-I
OH-	DP	Hydroxide Precipitation – Dissolved Product	R-I
OH-	FL	Hydroxide Precipitation – Flush or Wash	R-I
OH-	PF	Hydroxide Precipitation – Precipitator Feed	R-I
OX-	DP	Oxalate Precipitation – Dissolved Product	R-I
OX-	DS	Oxalate Precipitation – Dissolvent (reserved, not yet used)	N-I
OX-	FL	Oxalate Precipitation – Flush	R-I
OX-	PF	Oxalate Precipitation – Precipitator Feed	R-I
OX-	PT	Oxalate Precipitation – Precipitant (reserved, not yet used)	N-I
OX-	WR	Oxalate Precipitation – Waste Raffinate	R-I
PC		Process Condensate (T-34)	R-I
PU-	AF	Pubex – Adjusted Feed	R-I
PU-	CF	Pubex – Nitric Acid Flush	N-I
PU-	CP	Pubex – Curium (or Transplutonium) Product	R-I
PU-	CF	Pubex – Curium Wash – Strip	N-I
PU-	EX	Pubex – Pubex Extractant	N-I
PU-	IS	Pubex – Iron Strip Solution	N-I
PU-	PF	Pubex – Pubex Flush	R-I
PU-	PP	Pubex – Plutonium Product	R-I
PU-	ST	Pubex – Pubex Strip	N-I
PU-	WO	Pubex – Waste Organic	R-I
PUP		Plutonium Oxide Product	R-I
PVC		Process Vacuum Condensate (T-66, T-79)	R-I

RE-	AE	LiC1 AIX – Americium/Curium Elutriant	N-I
RE-	AF	LiC1 AIX – Adjusted Feed	R-I
RE-	AP	LiC1 AIX – Americium/Curium Product	R-I
RE-	BP	LiC1 AIX – Transcurium Product	R-I
RE-	CE	LiC1 AIX – Californium Elutriant	N-I
RE-	CF	LiC1 AIX – Column Flush	R-I
RE-	CR	LiC1 AIX – Column Raffinate	R-I
RE-	RE	LiC1 AIX – Rare-Earth Elutriant	N-I
RE-	SF	LiC1 AIX – Synthetic Feed	N-I
SFL		Off-Gas Scrubber (L-131) Feed Liquor	N-I
SFW		Acidified Plant Waste (F-115)	R-I
SLW		Off-Gas Scrubber (L-131) Liquor Waste	R-I
SP		Aqueous	R-I
SPW		Neutralized Plant Waste (F-126)	R-I
ST-	CM	Storage – Stored Curium (and/or Americium) Product	R-I
TR-	AF	Tramex – Adjusted Feed	R-I
TR-	ER	Tramex – Extraction Raffinate	R-I
TR-	SR	Tramex – Scrub Raffinate	R-I
TR-	ST	Tramex – Tramex Strip	N-I
TR-	TF	Tramex – Tramex Flush	R-I
TR-	TP	Tramex – Tramex Product	R-I
TR-	TS	Tramex – Tramex Scrub	N-I
TR-	TW	Tramex – Tramex Waste Organic	R-I
TR-	TX	Tramex – Tramex Extractant	N-I
VOG1		Vessel Off-Gas Iodine Retention System – Numbers Refer to Specific Sample Points on the Retention System	R-I
VOG3			R-I
VOG4			R-I
VOG6			R-I