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Planning for Receipt of Mark-18A Recovered Nuclear Material at Oak Ridge National Laboratory

Sharon Robinson, Julie Ezold, John Neal, and Bradley Patton
Oak Ridge National Laboratory
P.O. Box 2008, Oak Ridge, TN 37831

ABSTRACT

Mark-18A high-burnup targets produced at the Savannah River Site contain rare isotopes that are of use for heavy isotope production and isotope dilution mass spectrometry. These targets will be processed at Savannah River National Laboratory to recover plutonium oxide and a mixture of curium, americium, and lanthanide oxides. A Type A shipping package has been designed, fabricated, and certified to transport the heavy curium in special form to Oak Ridge National Laboratory. This paper discusses the preparations underway to receive the heavy curium packages and transfer the material into hot cell processing and storage facilities for subsequent use for heavy actinide production and research and development applications.

INTRODUCTION

The US Department of Energy's Office of Nuclear Materials Integration established the Mark-18A (Mk-18A) Target Material Recovery Program in 2015 to preserve the world's supply of ^{244}Pu and heavy curium (≥50% ^{246}Cm and ^{248}Cm) [1]. High neutron irradiation of ^{242}Pu in the Mk-18A targets in the K Reactor at Savanah River Site (SRS) resulted in unique contents, generating significant quantities of ^{244}Pu, heavy curium, high-Z isotopes, and long-lived fission products isotopes. The isotopic composition of the 65 Mk-18A targets in storage at SRS is shown in Table 1 [2]. Of particular interest are the ^{244}Pu and heavy curium. The Mk-18A targets contain ~650 g of heavy curium, which is the majority of the nation's heavy curium inventory.

The heavy curium in the Mk-18A targets will be used as a long-term feedstock for the production of high-demand heavy actinides such as ^{252}Cf and ^{249}Bk. Californium-252 is a radioactive neutron source used in many industrial applications, including oil exploration applications; in process control systems in the cement, coal, and mineral analyzer industries; to start nuclear reactors; in nondestructive materials analyses; and in medical research. Heavy actinides produced as by-products in ^{252}Cf production are used in super heavy element research. For example, ^{249}Bk was used in the discovery of new element 117, recently named tennessine [3]. Although alternative feedstocks containing light curium are available, they are much less attractive than the heavy curium contained in the Mk-18A targets. Light curium is defined as the element curium predominantly containing the isotope ^{244}Cm (<50% ^{246}Cm).

Table 1. Composition of Mk-18A Targets (decayed to October 1, 2022)

Isotope	Mass (g)	Activity (Ci)
Actinides		
^{238}Pu	6.70E-01	1.14E+01
^{239}Pu	3.89E-01	2.41E-02
^{240}Pu	5.17E+02	1.19E+02
^{241}Pu	3.52E+00	3.87E+02
^{242}Pu	5.49E+01	2.15E-01
^{244}Pu	2.40E+01	4.32E-04
^{241}Am	2.26E+01	7.22E+01
^{243}Am	4.13E+01	7.87E+00
^{244}Cm	1.22E+02	9.95E+03
^{245}Cm	2.76E+01	4.70E+00
^{246}Cm	4.43E+02	1.37E+02
^{247}Cm	2.44E+01	2.27E-03
^{248}Cm	3.51E+01	1.48E-01
^{249}Bk	1.33E-14	2.12E-11
^{249}Cf	5.79E-01	2.37E+00
^{250}Cf	3.86E-02	4.25E+00
^{251}Cf	1.24E-01	1.99E-01
^{252}Cf	3.55E-05	1.92E-02
Total actinides	1.32E+06	3.01E+05

Isotope	Mass (g)	Activity (Ci)
Primary Fission Products		
^{137}Cs	5.02E+01	4.35E+03
^{137m}Ba	7.61E-06	4.11E+03
^{90}Y	3.47E-03	7.74E+02
^{90}Sr	5.64E+00	7.74E+02
^{154}Eu	1.15E-01	3.10E+01
^{85}Kr	4.54E-02	1.81E+01
^{151}Sm	1.60E-01	4.21E+00
^{155}Eu	2.18E-03	1.07E+00
^{121m}Sn	1.51E-02	1.01E+00
^{121}Sn	2.00E-06	7.87E-01
^{99}Tc	2.61E+01	4.46E-01
^{134}Cs	7.74E-05	1.00E-01
Transition Metals		
^{79}Se	1.24E-01	1.91E-03
^{93}Zr	2.81E+01	7.02E-02
^{99}Tc	2.61E+01	4.46E-01
^{107}Pd	7.54E+01	3.83E-02
^{126}Sn	3.44E+00	4.24E-02
^{129}I	1.13E+01	2.00E-03

The production of ^{252}Cf requires both a high-flux reactor and a unique feedstock. The heavy curium in the Mk-18A material is attractive feedstock for irradiation to produce heavy actinides (Fig. 1). Since ^{252}Cf has a short half-life (2.6 years), it decays at a rate of about 25% per year, and new supplies must be regularly manufactured to meet the needs of various user communities. In the process of producing ^{252}Cf, other heavy actinides are produced as by-products including ^{249}Bk, ^{254}Es, and $^{255/257}$Fm.

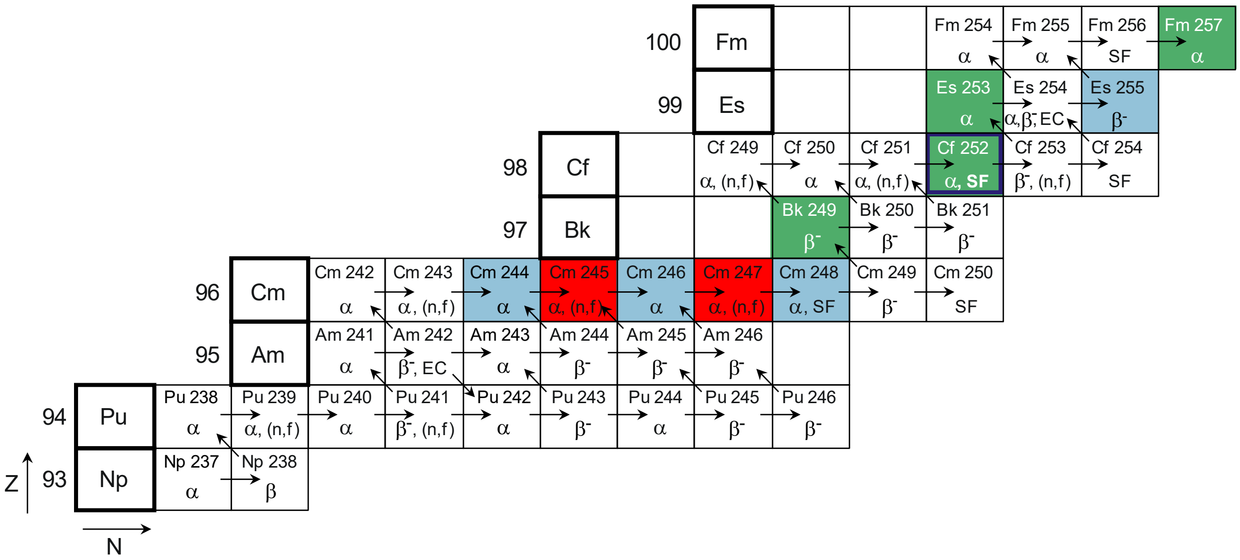

Fig. 1. Production of heavy actinides from plutonium and heavy curium

Heavy and light curium feedstock available for ^{252}Cf production originated from the recovery of curium from plutonium targets, which were irradiated in reactors located at SRS. Heavy curium was recovered at ORNL from Mk-18A targets in the 1970s and has been used as the primary feedstock for heavy actinide production at ORNL since it became available [4]. However, the inventory of recovered heavy curium has been reduced to the point that light curium is being mixed with heavy curium to provide sufficient feedstock for recent ^{252}Cf production campaigns [5].

Transplutonium isotopes are produced by irradiation in nuclear reactors where they are exposed to intense thermal neutron flux. The feed material is transmuted into heavier isotopes by a series of neutron captures and beta decays (Fig. 1). In the production of transplutonium isotopes, fission after neutron capture is considered a fission loss because it is not available for further transmutation. The production of transplutonium isotopes requires many neutron absorptions/beta decays, which result in large fission losses. Feedstock makeup has a significant impact on the efficiency of producing transplutonium isotopes, as shown in Figs. 2 and 3 [6].

The long irradiation pathway of neutron absorption/beta decay results in large fission losses, particularly during the production of heavy curium from ^{242}Pu. Figure 2 shows fission losses that occur during thermal neutron irradiation of ^{242}Pu in producing ^{252}Cf as a function of feedstock. There is a 99% fission loss in producing ^{252}Cf from unirradiated ^{242}Pu, as was done with original feedstock material in the 1960s. If light curium is used as the feedstock for ^{252}Cf production, a fraction of the ^{242}Pu has previously been transmuted to americium and ^{244}Cm, and further irradiation to produce ^{252}Cf results in approximately 98% fission losses. In the case of a heavy curium feedstock, previous irradiations have transmuted much of the ^{242}Pu to $^{246\text{–}248}$Cm, and the fission losses in producing ^{252}Cf are only about 11%.

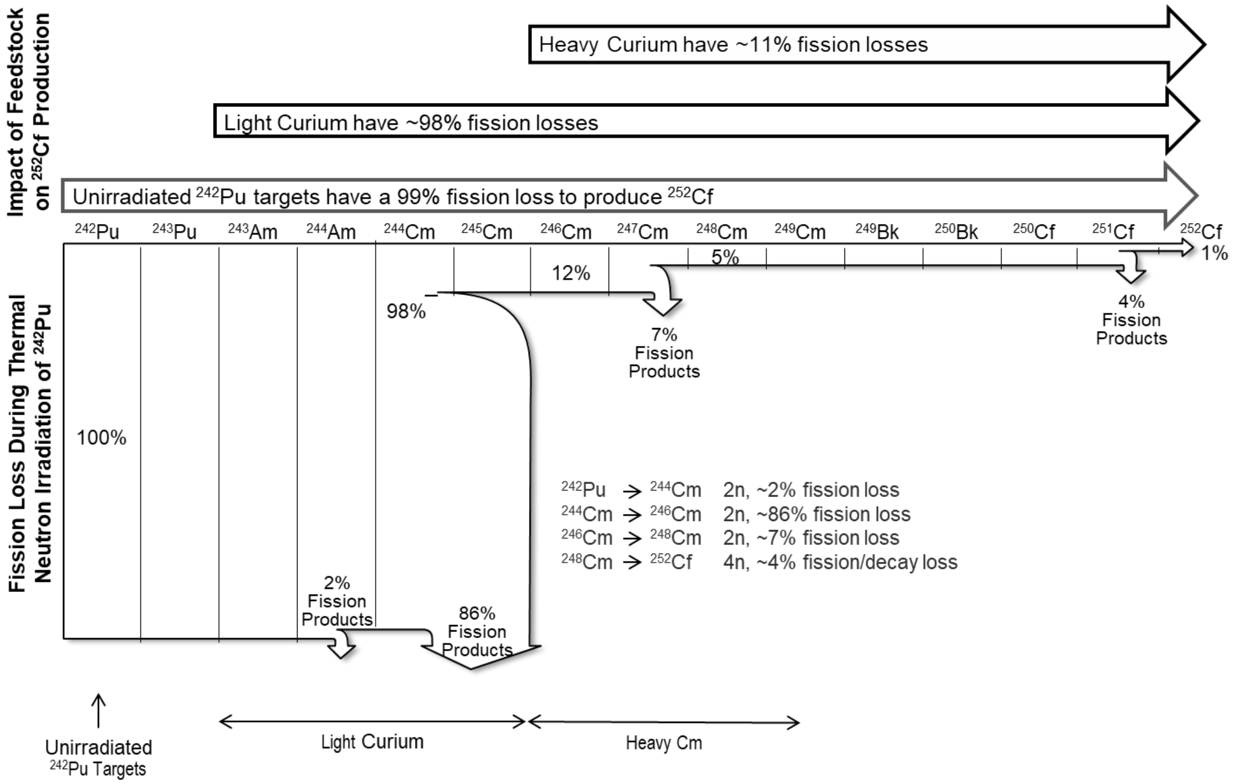

Fig. 2. Fission losses in production of ^{252}Cf

The influence of feedstock composition on ^{252}Cf production is illustrated in Fig. 3. The long irradiation pathway of neutron absorption/beta decay for production of ^{252}Cf from ^{242}Pu results in low production yields and requires more irradiation cycles in ORNL's High Flux Isotope Reaction (HFIR). Using ^{242}Pu as the feedstock in 1969 required irradiation of many targets for up to nine cycles in HFIR to produce ^{252}Cf. With heavy curium, much of the ^{242}Pu–^{245}Cm is burned out during previous irradiations before it is used as a feedstock. As such, heavy Cm can be transmuted to produce two to three orders-of-magnitude larger amounts of transplutonium isotopes with fewer irradiation cycles in HFIR than that required for ^{242}Pu or light curium. The use of heavy curium rather than light curium as a feedstock is estimated to result in 50% fewer targets and 50% shorter irradiation periods to produce the same amount of ^{252}Cf. Therefore, the heavy curium in the Mk-18A targets is a preferred feedstock for ^{252}Cf and heavy actinides.

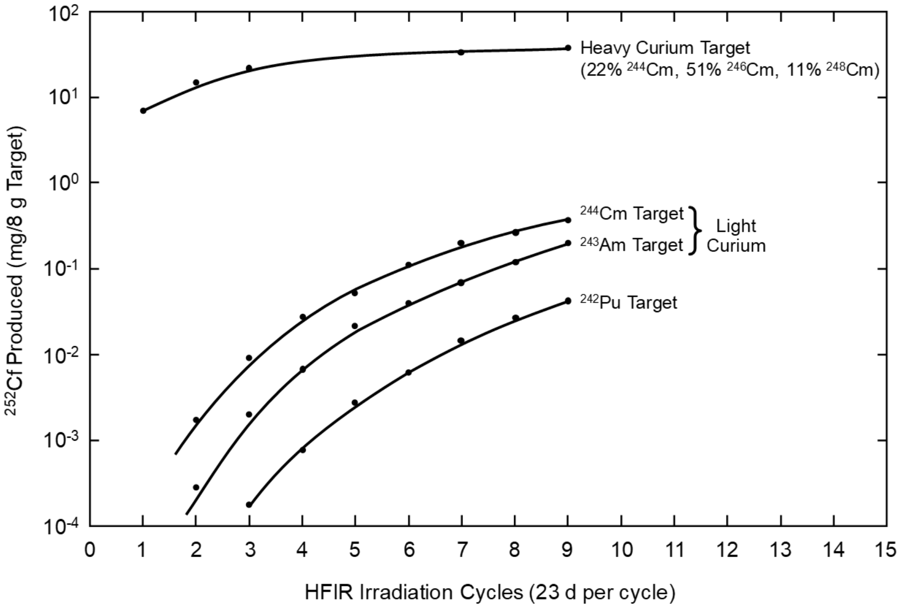

Fig. 3. Impact of feedstock on ^{252}Cf production

TRANSPORT, RECEIPT, AND STAGING OF MK-18A MATERIAL

Heavy curium will be recovered from the Mk-18A targets at Savannah River National Laboratory (SRNL) and will be shipped to ORNL for use in heavy actinide production. The shipping, handling, and staging plans are shown in Fig. 4.

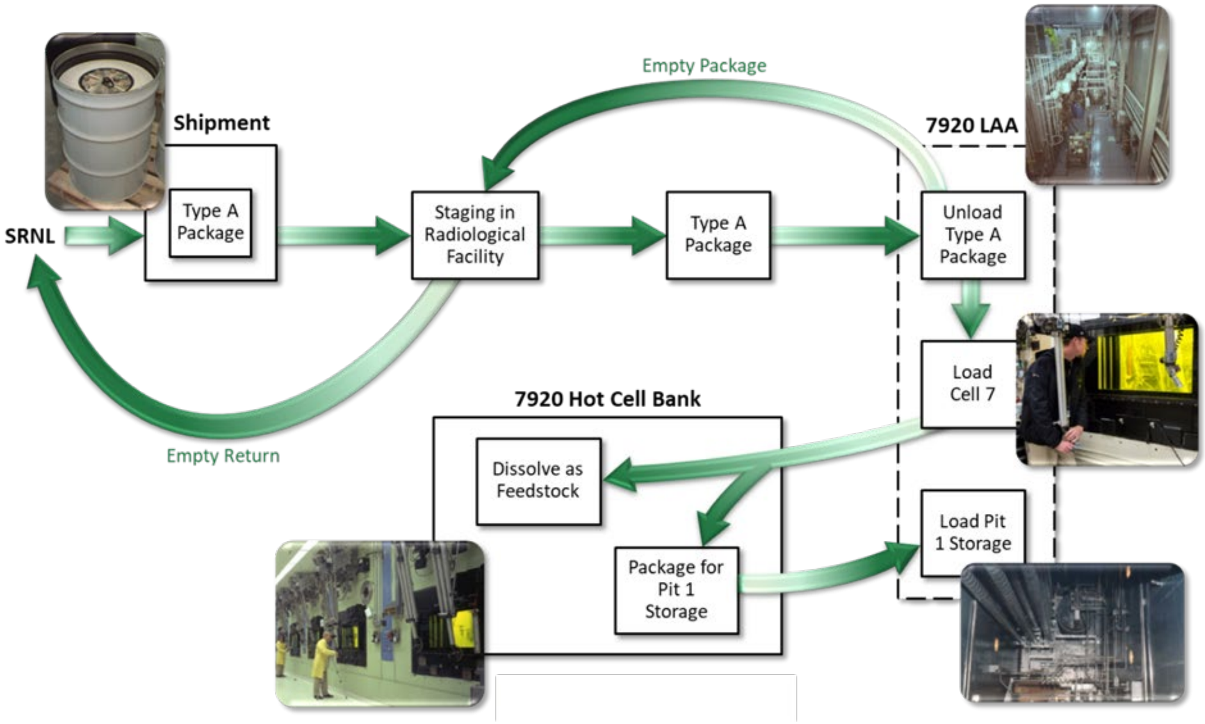

Fig. 4. Shipping, handling, and staging plan for Mk-18A heavy curium

The heavy curium can be shipped in Type A packaging when it is in special form. A Mk-18A 7A shipping container has been developed for shipping the Mk-18A material. It is a modification of the Waste Isolation Pilot Plant Nuclear Waste Partnership S-300 7A packaging that contains both gamma and neutron shielding. It has been tested to meet 49 CFR 178.350 requirements for shipping Type A quantities of material in normal form. A conceptual drawing and photographs of the Mk-18A 7A shipping container are shown in Fig. 5. It is a 208 L (55 gal) drum that contains ~6 cm (2.4 in.) of lead shielding and ~13.7 cm (5.4 in.) of high density polyethylene (HDPE) shielding plus a 1.3 cm (0.5 in.) HDPE removable sleeve located inside the lead shielding.

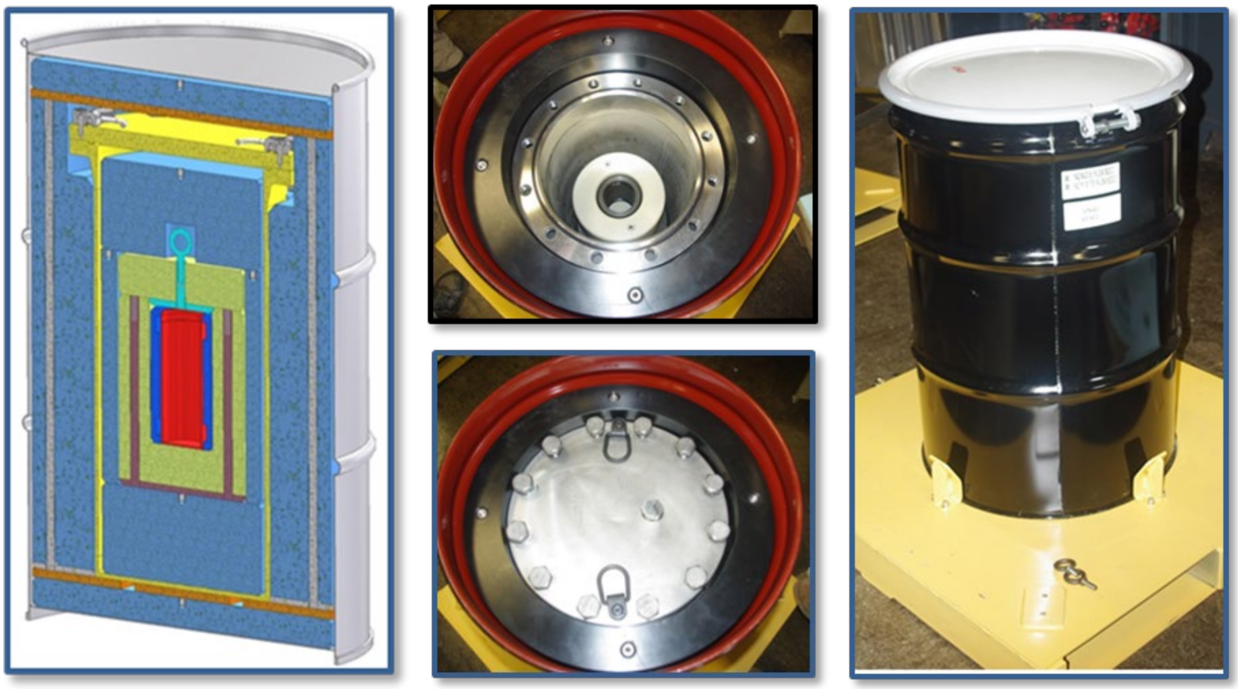

Fig. 5. Mk-18A 7A Type A shipping container

The cavity of the new container is ~11.4 cm (4.5 in.) diameter (without the HDPE sleeve) and 22 cm (8.5 in.) high. Without the HDPE sleeve, it will accommodate a 10 cm (4 in.) diameter food pack can, typically a thin-walled tinned carbon steel container that is double-crimped metal-to-metal closure used in the food industry and for nuclear materials transport. It will accommodate a 6.4 cm (2.5 in.) diameter by 18 cm (7 in.) tall special form capsule (SFC) with the HDPE sleeve in place. It will reduce a radiation dose of ~5,000 R/h on the surface of the SFC to below dose limits for the Type A package for exclusive use in domestic ground shipments (1,000 mR/h on the package surface, 200 mR/h on the outer surfaces of the vehicle, and 10 mR/h at 2 m for the lateral surfaces of the vehicle).

The Mk-18A 7A shipping container weighs ~420 kg (930 lb). A metal pallet has been designed for movement of the loaded container. The drum is secured to the pallet with a custom rolled steel band bolted together and clamped to the side of the drum (Fig. 6) for movement with a folk lift or a crane. It is envisioned that the drum will stay attached to the pallet during transport, but this will not be a requirement.

Fig. 6. Pallet for Mk-18A 7A shipping container

The heavy curium will be shipped from SRNL to ORNL in the Mk-18A 7A shipping container in special form. Multiple Type A containers will be shipped during a single transport. They will be temporarily staged in a radiological facility adjacent to the ORNL hot cell facility and transferred individually into the Radiochemical Engineering Development Center where the SFC will be removed from the Type A shipping container in a limited access area and transferred into the hot cells.

A semiremotely operated transfer shield has been designed to transfer the SFC from the Mk-18A 7A shipping container into the hot cell as shown in Fig. 7. The transfer carrier will mate to both the Type A shipping container and the transfer opening located on the top of the hot cell. The shielded retrieval system will be lowered into the Type A container where a specially designed gripper will attach to the SFC basket and lift it into the transfer carrier. The transfer carrier will be moved to mate with the hot cell transfer opening, and the SFC basket will be lowered into hot cell where the material will be stored until used as heavy actinide production feedstock. The Type A container will be returned to SRNL for reuse.

The transfer shield has 20 cm (7.7 in.) of HDPE shielding and 2.8 cm (1.1 in.) lead shielding. It uses a specially designed shielded retrieval system with a state-of-the-art gripper mechanism capable of lifting up to 90 kg (200 lb). The schematics of the major components of the system are shown in Fig. 8. The overall weight of the unit will be ~635 kg (1,400 lb).

Once the SFC capsules are transferred into the hot cells, the heavy curium will be repackaged and stored in the building's storage pit for future use in ^{252}Cf production campaigns.

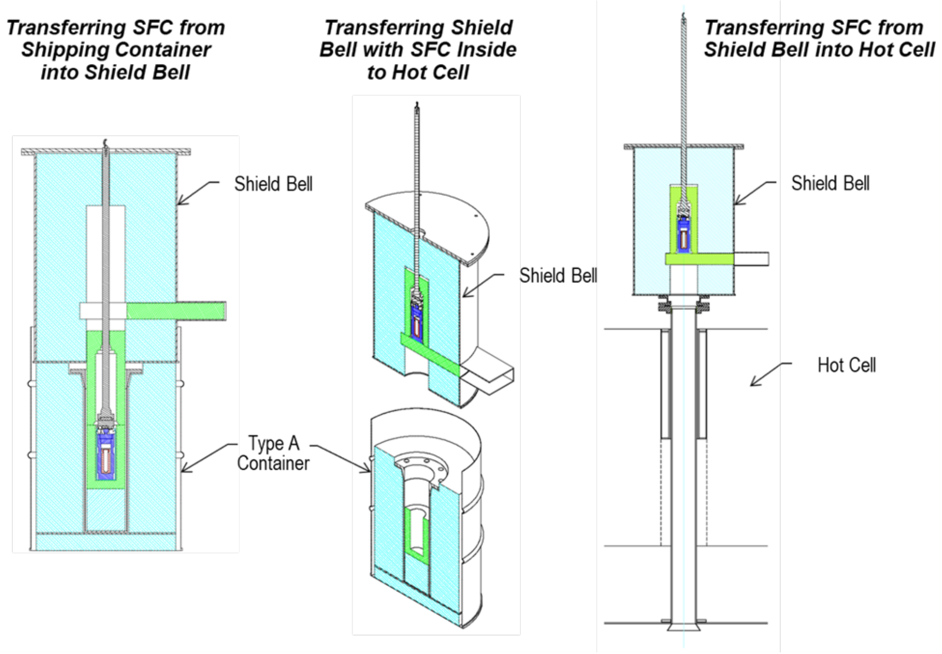

Fig. 7. Concept for transferring special form capsule from shipping container into the hot cell

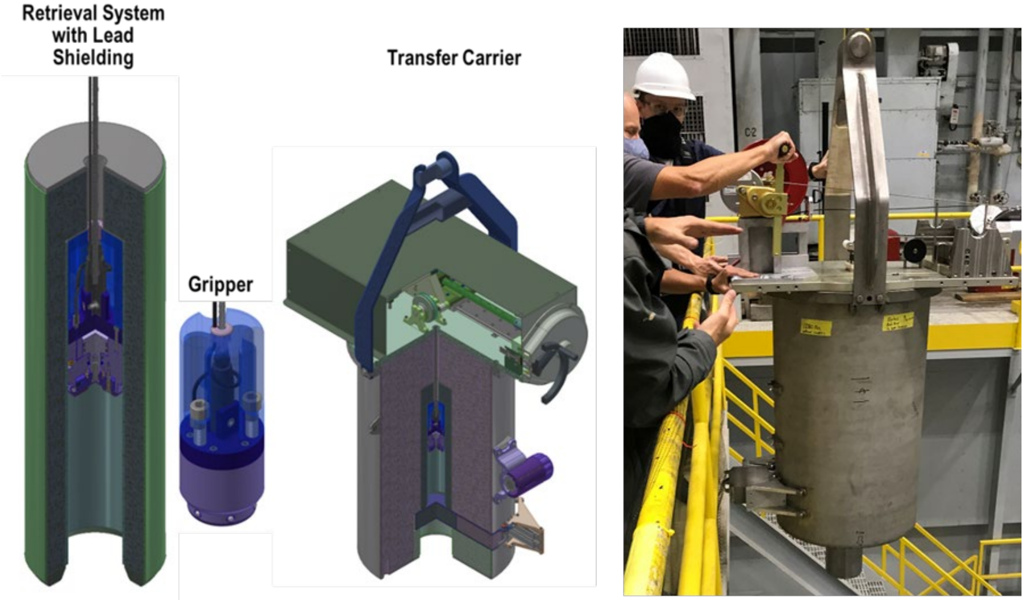

Fig. 8. Transfer shield design concept

PRODUCTION OF ^{252}Cf

A basic flowsheet for production of transplutonium elements is shown in Fig. 9 [6]. In general, the feed material (Pu, Am, Cm, or a combination of these elements) is prepared into Al-clad, Al-matrix cermet (a ceramic/metal composite material) targets. The targets are irradiated in HFIR, where the Pu/Am/Cm is transmuted to heavier isotopes in a series of neutron captures and beta decays to produce ^{249}Bk, ^{252}Cf, ^{253}Es, and ^{257}Fm. After irradiation, the Pu/Am/Cm targets can be allowed to decay for several months to eliminate the short-lived fission and activation products. The targets are then transferred to the Radiochemical Engineering Development Center for disassembly, chemical separation, purification, and packaging of the transplutonium products for distribution to users. A fraction of the Pu/Am/Cm starting material remains at the end of the process and can be recycled for the next campaign.

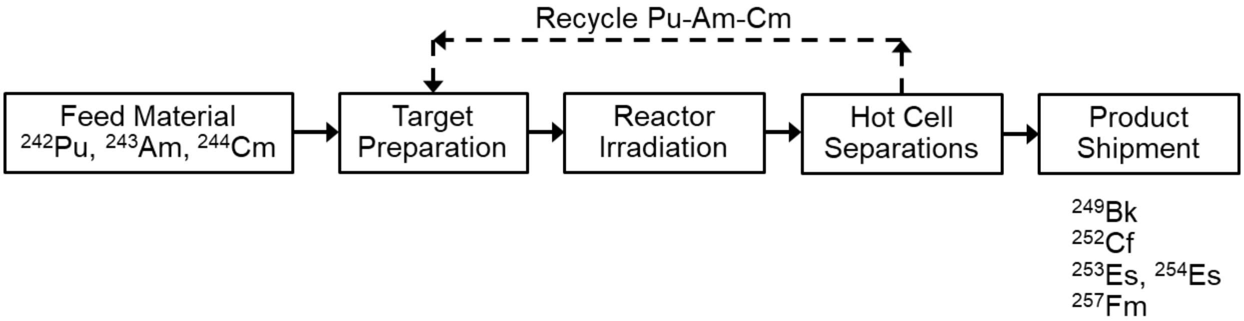

Fig. 9. Simplified flowsheet for production of transplutonium isotopes

Figure 10 illustrates key steps in californium processing that are performed at ORNL. This shows the cycle used to prepare and irradiate feedstock, recover californium, and reprocess unconsumed material. A flow diagram for the source material production process is shown below the separations and purification flow diagram in Fig. 11.

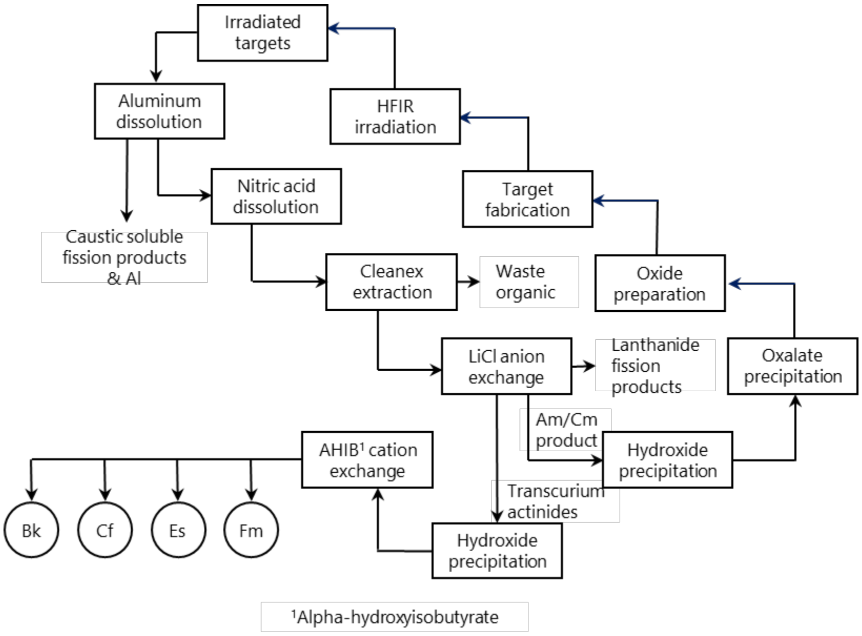

Fig.10. Irradiated target processing—separation and purification

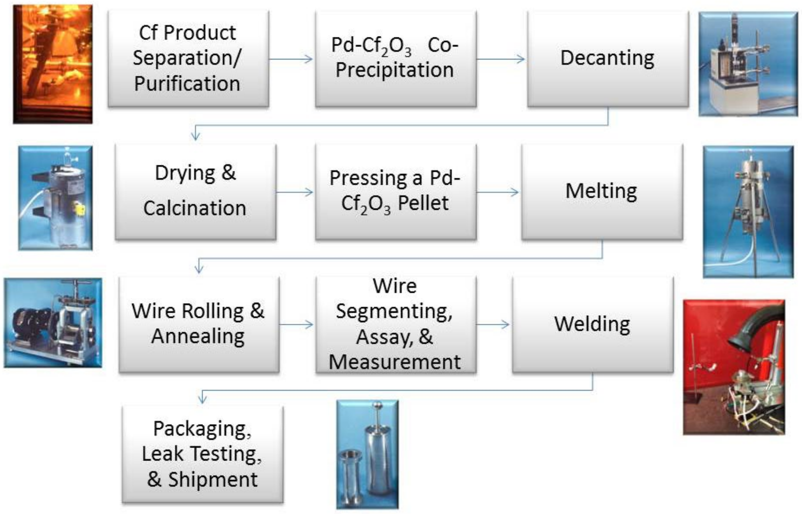

Fig. 11. Californium product for shipment to customers

SUMMARY AND CONCLUSIONS

The Mk-18A Target Material Recovery Program will retrieve Mk-18A targets from wet storage at L-Basin at the SRS and process them at SRNL to recover a plutonium oxide stream and a heavy curium oxide material that will contain the americium, curium, and lanthanides. ORNL is preparing for the transport of the materials from SRNL and receipt and staging of the materials. ORNL has developed, assembled, and certified a 208 L (55 gal) Type A shipping container for transport of heavy curium material. A transfer shield has been designed and constructed to transfer the material from the Type A shipping container into the ORNL hot cells where it will be stored until it is used as feedstock for heavy actinide production. ORNL expects to begin receiving Mk-18A material in approximately 2024.

ACKNOWLEDGMENTS

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REFERENCES

1. S. M. Robinson et al., *Recovery of Mark-18A (Mk-18A) Target Materials: Program Management Plan*, ORNL/TM-2014/314, Oak Ridge National Laboratory, Oak Ridge, TN, 2014.
2. B. D. Patton et al., Special Actinide Recovery from Mark-18A Target Material, INMM 58th Annual Meeting, Indian Wells, CA, 2017.
3. U. T. Oganessian et al., "Synthesis of a New Element with Atomic Number Z = 117," *Physical Review Letters, American Physical Society*, 104, 14 (April 2010).
4. S. M. Robinson et al., *Mark-18A (Mk-18A) Target Processing at Oak Ridge National Laboratory*, ORNL/TM-2015/577R1, Oak Ridge National Laboratory, Oak Ridge, TN, 2016.
5. S. Hogle et al., "Sensitivity Studies and Experimental Evaluation for Optimizing Transcurium Isotope Production," *Nuclear Science and Engineering*, 185(3): 473–483 (March 2017).
6. S. M. Robinson et al., "Production of Cf-252 and Other Transplutonium Isotopes at Oak Ridge National Laboratory," *Radiochimica Acta* (July 2020).