Processing and Disposition of Special Actinide Target Materials – 13138

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ABSTRACT

The Department of Energy (DOE) manages an inventory of materials that contains a range of long-lived radioactive isotopes that were produced from the 1960s through the 1980s by irradiating targets in high-flux reactors at the Savannah River Site (SRS) to produce special heavy isotopes for DOE programmatic use, scientific research, and industrial and medical applications. Among the products were californium-252, heavy curium (including Cm-246 through Cm-248), and plutonium-242 and -244. Many of the isotopes are still in demand today, and they can be recovered from the remaining targets previously irradiated at SRS or produced from the recovered isotopes. Should the existing target materials be discarded, the plutonium (Pu) and curium (Cm) isotopes cannot be replaced readily with existing production sources.

Some of these targets are stored at SRS, while other target material is stored at Oak Ridge National Laboratory (ORNL) at several stages of processing. The materials cannot be stored in their present form indefinitely. Their long-term management involves processing items for beneficial use and/or for disposition, using storage and process facilities at SRS and ORNL. Evaluations are under way for disposition options for these materials, and demonstrations of improved flow sheets to process the materials are being conducted at ORNL and the Savannah River National Laboratory (SRNL). The disposition options and a management evaluation process have been developed. Processing demonstrations and evaluations for these unique materials are under way.

INTRODUCTION

Mk-42 and Mk-18A target materials are of special interest for their Pu-244 and heavy curium components. They are stored at ORNL and SRS, respectively. The inventory of isotopes of interest contained in each is given in Table I. Their status and unique features are described below.

TABLE I. Estimated inventory of Mk-42 and Mk-18A materials (decayed to October 1, 2011)

Item	Fission Products	Plutonium (grams)	Americium (grams)	Curium (grams)
Mk-42 (28 unprocessed target segments)	~20 kCi mixed	1244	295	72
Mk-42 (104 Am/Cm/Ln oxide capsules)	4,620 grams lanthanides only	79	484	189
Mk-18A (65 unprocessed assemblies)	~10 kCi mixed	426	34	659

Mk-42 Material

Mk-42 targets were fabricated from Pu-239 and irradiated at SRS to make Pu-242, Am-243, and Cm-244 primarily for defense applications. Ten of the Mk-42 targets were allocated for ORNL to recover Pu-242, Am-243, and Cm-244. These ten targets were shipped from SRS to the Pacific Northwest National Laboratory where they were cut into eight segments each, and a total of 80 segments were shipped to ORNL for processing. Fifty-two of the original 80 segments have been processed. Early in the program, the sponsor requested that ORNL recover only the Pu-242 and hold the americium/curium (Am/Cm) for later use. Based on that request, ORNL recovered and purified the Pu-242 (~2.2 kg have been shipped and another ~1 kg remains in unprocessed segments) but left the lanthanide (Ln) fission products– rare earth elements of atomic numbers 57–71– with the Am/Cm and solidified the resulting mixture to facilitate the Pu-242 recovery. This mixed Am/Cm fission product material was transferred to small, welded stainless steel (SS) capsules. The Am/Cm materials contain significant quantities (~5 kg total) of Ln fission products which must be removed to produce a relatively pure Am/Cm oxide form for future use as feedstock for heavy actinide production. Examples of the unprocessed target segments and capsules containing the Am/Cm/Ln oxides are shown in Fig. 1.

Mk-42 Segments Storage Canister

Am/Cm/Ln Oxide Capsule

Fig. 1. Mk-42 target segments and oxide capsules in storage at ORNL.

Mk-18A Material

Sixty-five Mk-18A targets were irradiated in a high neutron flux mode in the K-Reactor at SRS from August 1969 until November 1970, and then in a low flux mode until 1979. Upon removal from the reactor, the targets were placed in J-cans (Fig. 2) in water basin storage in the Receiving Basin for Offsite Fuels until 2001, when they were moved to their present storage location in L-Basin.

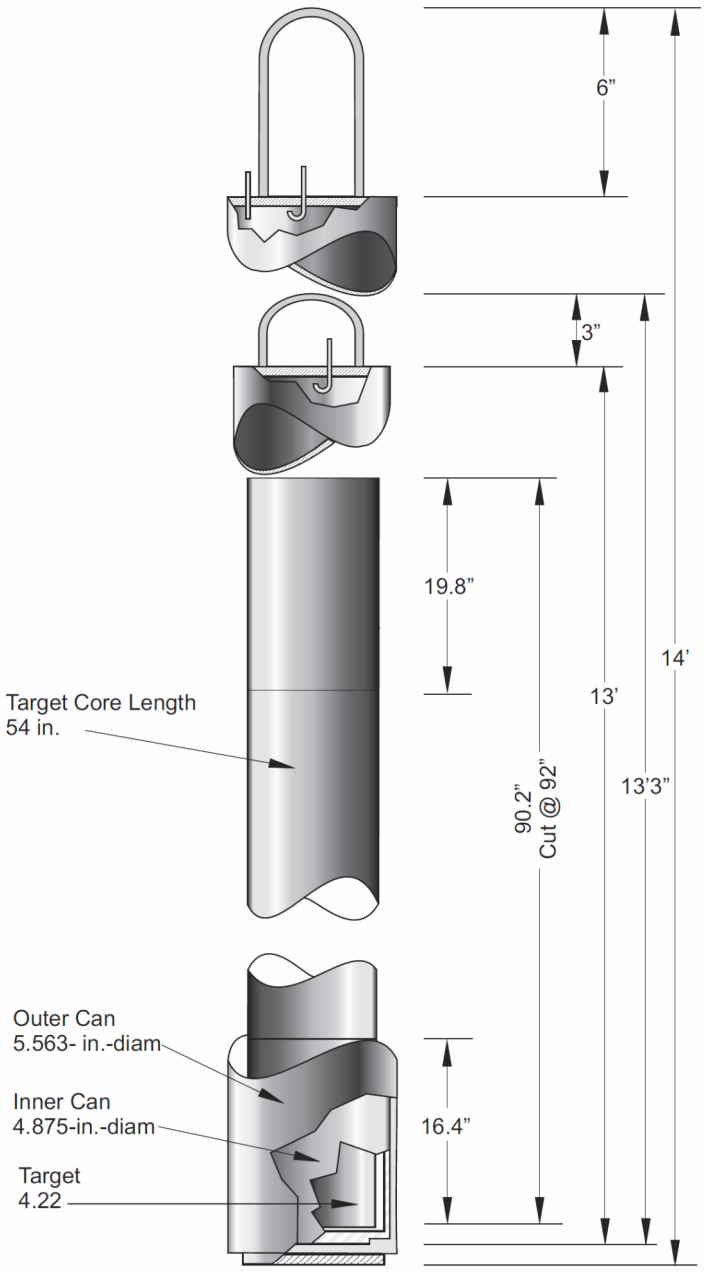

Fig. 2. Mk-18A target stored in J-cans in L-Basin at SRS.

The high neutron irradiation resulted in Mk-18A targets with very unique contents. In particular, they contain Pu-244 and heavy curium (including Cm-246 through Cm-248). Plutonium-244 is an extremely rare and long-lived isotope of plutonium that is not produced in either commercial fuel irradiation or weapons-grade plutonium production. The majority (>90%) of the existing global inventory of Pu-244 is contained in the 65 Mk-18A targets stored at the SRS. The total inventory in the Mk-18A targets is about 20 g of Pu-244 in several hundred grams of plutonium. It is ideal for precise radiochemical analyses measuring plutonium material properties and isotopic concentrations in items containing plutonium. The isotope dilution mass spectrometry is about ten times more sensitive using Pu-244 rather than Pu-242, which is commonly available as a spike for determining plutonium isotopic content. The isotope can also be irradiated in small quantities to produce super heavy elements. Since Pu-244 is much less hazardous than other isotopes of plutonium (a factor of 3,000 less of an inhalation hazard than Pu-239), it can also be used to conduct basic research studies on plutonium. The heavy curium in the Mk-18A targets is an attractive long-term feedstock for Cf-252 production in the High Flux Isotope Reactor (HFIR) at ORNL. The Mk-18A targets contain ~80% of the national heavy curium inventory. Although Pu-244 would be prohibitively difficult and expensive to produce in existing reactors and has a number of unique and important uses, there is currently no sponsor support to recover the material.

DESCRIPTION

ORNL and SRS have been tasked by DOE Office of Nuclear Materials Integration (NA-73) to identify and evaluate disposition options for the unprocessed Mk-42 segments and the Mk-42 Am/Cm/Ln oxide mixture as well as the Mk-18A targets. The objective of the study is to identify feasible disposition options, develop preliminary cost estimates for viable options, evaluate each option versus programmatic needs, and recommend dispositioning options for each material. Evaluations are under way for disposition options for these materials, and demonstrations of improved flow sheets developed to reduce the costs to process the materials are being conducted.

Disposition Options

The options being evaluated for the Mk-42 materials include (1) return to SRS for storage and disposition with similar material as waste; (2) processing to recover americium, curium, and plutonium at ORNL for feedstock for production of heavier transcurium elements in the HFIR; or (3) disposition as remote-handled transuranic waste at the Waste Isolation Pilot Plant (WIPP).

Returning the materials to SRS would require retrieval from storage and repackaging the materials at ORNL in aluminum containers suitable for processing in H-Canyon, and transporting the materials to SRS for processing. Processing the materials at ORNL for disposal at the WIPP would include retrieving the materials from storage, dissolving the actinides, grouting the materials, and packaging them for transport and disposal. Recovery of the actinides would involve processing the materials in the facility where they are presently stored to recover and purify the Am/Cm for future use. In the latter case, the Am/Cm product would be processed to an oxide and stored for future irradiation in the ORNL HFIR to produce heavier transuranium elements. In addition, the Pu-244 in the Mk-42 segments would be recovered along with the other plutonium isotopes, processed to an oxide, and stored for future enrichment.

The disposition options for the Mk-18A materials include (1) recovery of Pu-244 and heavy curium for future use, (2) recovery of Pu-244 only for future use combined with processing of the heavy curium for disposal as waste, and (3) processing the targets for disposal as waste without recovery of Pu-244 or heavy curium. The major steps in the disposition pathways include packaging/transport of the targets to treatment/storage/disposal facilities, dissolution of the targets, and processing of the material for recovery of isotopes and/or disposal as waste. In scenarios where the isotopes are recovered for future use, the recovery steps could be performed at SRS or ORNL, and the disposition endpoint is storage as oxides in sealed capsules at ORNL. In scenarios where the isotopes are dispositioned as waste, all processing would occur at SRS.

Recovery Options

Since the Am-243 and Cm-244 in these targets are essentially irreplaceable and are attractive feedstocks for the production of heavy actinides, studies are under way to develop a separations process optimized to recover and purify the Am/Cm for future use in heavy-actinide production. The process is being demonstrated at ORNL for recovery of Am/Cm from Mk-42 materials and is being evaluated for applicability for processing Mk-18 materials.

As discussed in the Introduction section, the Mk-42 segments were initially processed to recover plutonium, americium, and curium products using the process shown in Fig. 3. The time-consuming LiCl anion exchange process was used to recover americium and curium and remove Ln fission product wastes in three separate streams, as shown in the grayed portion of the flow sheet in Fig. 3. When the sponsor requested that ORNL recover only the Pu-242 and hold the Am/Cm for later use, only the unit operations shown in white were performed and the Am/Cm/Ln was stored as metal oxide in SS capsules. Future use of Mk-42 materials for heavy-actinide production does not require separation of the americium and curium but only requires removal of the bulk of the Ln fission products. Currently, simplified processes are available for the recovery and purification of the Am/Cm from both the segments and Am/Cm/Ln metal oxide capsules, as shown in the grayed portion of Fig. 4. The new flow sheet consists of an oxide dissolution step, a plutonium extraction process, a Ln partitioning step, a step to remove complexants, and an oxalate precipitation step to prepare Am/Cm oxide for target fabrication. When Am/Cm/Ln oxides have been stored for long periods of time after the original plutonium separation step, a second plutonium separation step is required to remove the Pu-240 that has grown from the decay of Cm-244. The Reverse TALSPEAK (Trivalent Actinide–Lanthanide Separation by Phosphorus reagent Extraction from Aqueous Komplexes) flow sheet is being used to separate the Ln from the Am/Cm elements [1, 2].

Evaluation Methodology

An alternatives evaluation is being performed to rank the disposition options for each target material using a decision-making methodology that provides a structured framework for comparing both qualitative and quantitative selection criteria. The selection criteria for disposition pathways for the target materials include

- technical feasibility (likelihood of successful implementation),
- implementation costs and schedule, and
- value of the recovered material for future beneficial use.

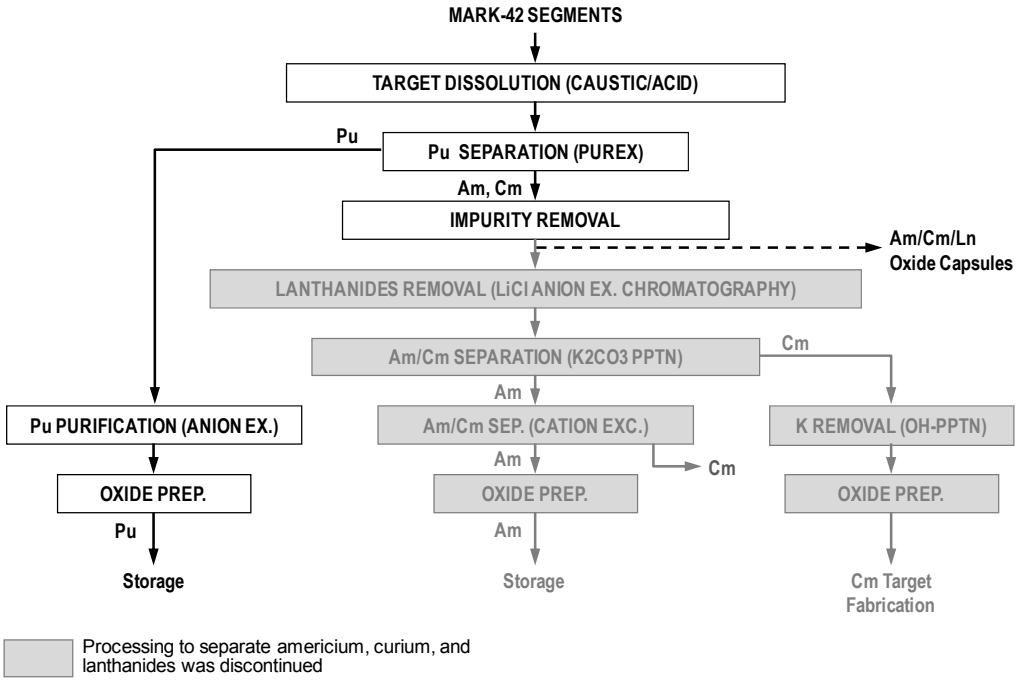

Fig. 3. Historic Mk-42 material separations flow sheet.

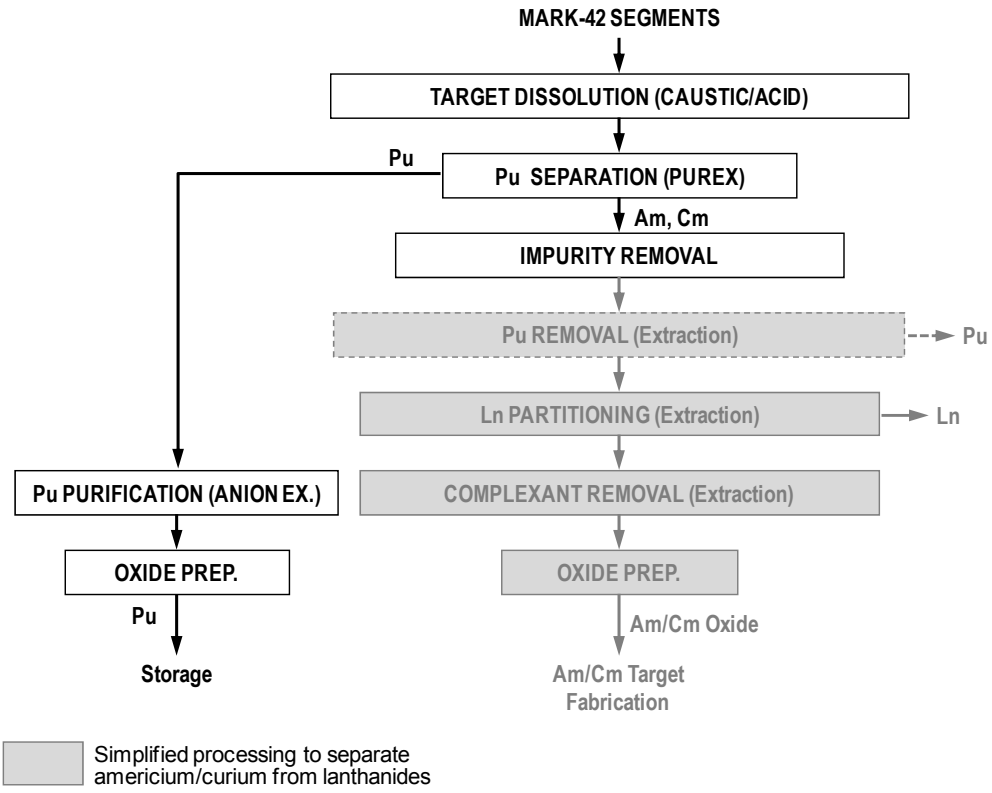

Fig. 4. Simplified Mk-42 separations process flow sheet.

The relative importance of each selection criterion is developed by the evaluation team (e.g., how does one weigh the importance of "cost" as a selection criterion relative to "value of the recovered material for future use"?). Each disposition pathway is then given a "score" for each of the individual selection criterion, and weighted ratings are obtained for each criterion by combining the individual score and the weighting factor for that selection criterion. An overall score is assigned to each disposition pathway by "summing" each of the individual weighted ratings. The ratings will be assigned by the team based on experience guided by criteria definitions. In some cases, actual data (i.e., cost estimates for implementing the pathway) will be used to assign ratings, and in other cases qualitative assessments (i.e., value of the recovered material for future use) will be used to assign ratings based on the criterion definitions developed by the team. Example evaluation criteria are shown in Table II.

DISCUSSION OF RESULTS

Demonstration of Improved Am/Cm Recovery Flow Sheet

A series of demonstration campaigns are being conducted at ORNL to demonstrate and refine the overall process to recover Am/Cm shown in Fig. 4, and to evaluate processing parameters to optimize the separation of Ln fission products from the Am/Cm constituents. The first campaign was completed in FY 2011 using one of the Am/Cm/Ln oxide capsules containing ~4 g of americium, ~2 g of curium, and ~60 g of Ln fission products [3]. The demonstration run showed satisfactory results with a Ln decontamination factor (DF) of 34 for the Am/Cm product. The total losses of americium and curium to the waste streams were very low (<0.2% and 0.02%, respectively) for the Ln partitioning step. The Am/Cm losses indicated may be partially due to residual contamination from previous use of the equipment.

A second campaign was performed in early FY 2012 in which nine capsules containing ~40 g of americium, ~20 g of curium, and ~600 g of Ln fission products were processed using the flow sheet from the one capsule test. The concentrations of actinides and Ln in the process streams were increased by a factor of 3 to increase the throughput of the system. The losses to the waste streams for the Ln partitioning step were similar to those for the first campaign: <0.09% and 0.07%, respectively [4]. A Ln decontamination factor (DF) of 22 was achieved for the Am/Cm product. This is adequate for heavy-actinide feedstock.

The third campaign is under way where 20 capsules are being processed using the same stream concentrations demonstrated in the second campaign. The information from the earlier campaigns will be used in the planning for subsequent campaigns.

Estimates of Materials Generated for Storage and Waste Disposal

The amount of materials that would be put in storage and the amount of secondary waste that would be generated for disposal were estimated assuming the materials would be processed at ORNL for the following scenarios:

TABLE II. Examples disposition evaluation criteria[a]

Criteria	Weighting Factor (H, M, L)	Subcriteria	Weighting Factor (H, M, L)
Feasibility of implementation	H	Processing/packaging	H
		Shipping	M
		Acceptance at site	H
		Staging	L
		Regulatory	M
Cost/schedule	M	Cost	M
		Schedule	L
Preserving materials for future use	H	Uniqueness for heavy element production	H
		Uniqueness for R&D and standards	M

[a]H = High, M = Medium, L = Low

- the Mk-42 segments and Am/Cm oxide capsules are processed to metal oxide powders for use as future feedstock materials,
- the Mk-42 segments and Am/Cm oxide capsules are processed for disposal at WIPP, and
- the Mk-18A targets are processed to recover plutonium and Am/Cm as metal oxide powders for use as future feedstock materials.

As shown in Fig. 2, the 65 Mk-18A targets are presently stored in J-cans in the SRS L-basin. They occupy 4.3 m^3 of shielded storage space. For processing at ORNL, they would be removed from the J-cans, packaged for shipment, cut into sizes that can be handled at ORNL, and placed into storage canisters similar to those used to store the Mk-42 segments. It is estimated that 78 storage canisters would be required to hold the 65 targets. These could occupy approximately 0.4 m^3 of shielded storage space. Processing the targets for recovery of feedstock materials using the methods shown in Figs. 3 or 4 would remove the aluminum, silicon, and fission products and process the plutonium-rich materials and the Am/Cm-rich materials into metal oxide powders that could be stored in SS capsules like those used for the Mk-42 Am/Cm oxides. This will result in ~10 capsules containing ~480 g plutonium oxide and 15 capsules of ~780 g Am/Cm oxide. These capsules would require approximately 0.004 m^3 of shielded storage space. As shown in Fig. 5, this will result in a shielded storage requirement reduction of three orders of magnitude.

If the 28 Mk-42 segments were processed using the recovery methods shown in Fig. 4, it is estimated that ~1400 g of plutonium oxide would be produced that could be stored in 28 SS capsules, and ~400 g of Am/Cm oxide powder would be produced that could be stored in approximately nine capsules. This would result in a shielded storage space of ~0.004 m^3 for a reduction factor of 25 for storage space requirements, as shown in Fig. 6.

Fig. 5. Processing Mk-18A targets to oxide powders reduces shielded storage requirements by three orders of magnitude.

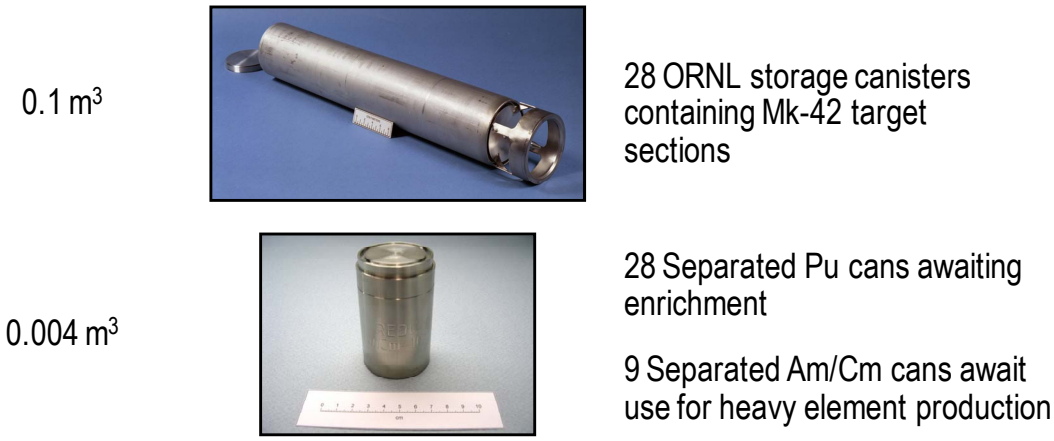

Fig. 6. Processing Mk-42 targets to oxide powders reduce shielding storage requirements by a factor of 25.

The amount of secondary waste that would be generated for disposal at WIPP has been estimated for each of these cases. Processing the target materials for recovery of plutonium and Am/Cm using the methods shown in Figs. 3 and 4 would result in ~90 and ~140 55-gal drums of remote-handled transuranic material for shipment to WIPP for the Mk-42 and Mk-18A target materials, respectively. A 97% recovery of the plutonium and Am/Cm isotopes were assumed for planning purposes.

Processing the Mk-42 materials at ORNL for disposal at the WIPP would include retrieving the materials from storage, dissolving the actinides, grouting the materials, and packaging them for transport and disposal. It is estimated that this would result in ~110 55-gal drums of remote-

handled transuranic waste for disposal at WIPP, or 20 more drums than if the material is processed for recovery of Pu/Am/Cm.

CONCLUSIONS

Options for processing and storage of Mk-18A and Mk-42 products are being pursued as part of feedstock evaluation studies. A methodology for evaluating the materials has been developed and disposition options, including recovery of valuable plutonium, americium, and curium isotopes and disposition as waste, have been identified. The volume of materials that would be put in safe storage if the desired actinides are recovered has been estimated, and the amount of waste that would be generated for disposal has also been estimated for each disposition option. This information will be used to develop recommendations for disposition of the materials in 2013.

ORNL has demonstrated flow sheets to enhance processing efficiencies and minimize waste stream losses associated with recovery of americium and curium from Mk-42 target materials stored at ORNL. A series of campaigns is being performed to demonstrate a simplified process to recover Am/Cm and to optimize the removal of Ln fission products from the Am/Cm constituents. Two demonstration runs have been completed using one and nine of the Am/Cm/Ln oxide capsules, respectively. The amount of material processed in subsequent campaigns will be increased and may include the segments in inventory at ORNL.

ACKNOWLEDGMENTS

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