Disposition Options for Mk-42 Materials - 17072

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ABSTRACT

In the late 1980s, the DOE Defense Programs identified a need to recover special isotopes of americium, curium, and plutonium (Am/Cm/Pu) from Mark-42 (Mk-42) targets that had been irradiated at the SRS. Eighty Mk-42 target segments were transferred from SRS to ORNL for recovery of these special isotopes. Approximately 65% of the target materials has been processed and converted to solids. The remaining Mk-42 materials are either targets stored in their irradiated forms or target material stored from various stages of processing. The Pu, Am-243 and Cm-244 in these targets are valuable materials; for example, the Am/Cm materials are potential feedstocks for production of heavy actinides (Cf-252, Bk-249, etc.). Although the isotopes in the Mk-42 materials have value, there is continual pressure to look at de-inventorying material in storage to only retain the most valuable materials needed to meet mission needs and to allow other materials to be added to the inventory as they become available. Evaluations are underway for potential disposition options for the Mk-42 materials.

INTRODUCTION

Approximately 30 years ago, Mk-42 targets were fabricated from Pu-239 and irradiated at the SRS to produce Pu-242, Am-243, and Cm-244 primarily for defense applications. All but ten of the targets were processed at SRS to recover Pu-242, and the remaining ten where shipped to ORNL to separate and recover Pu-242, Am-243, and Cm-244. At the present time, approximately 65% of the Mk-42 materials transferred to ORNL has been processed. All of the plutonium recovered from the targets has been shipped off-site, and the materials remaining from various stages of processing are in storage at ORNL. This paper summarizes the results of a technical evaluation of disposition options for the various Mk-42 materials in storage at ORNL. The evaluation includes options for recovering Pu, Am-243, and Cm-244 isotopes and evaluates the resulting reduction in the amount of waste that would be sent to disposal facilities.

Why Am-243 and Cm-244 Recovery Is Important

Californium-252 is a radioactive neutron source used in many industrial applications including in oil exploration applications; in process control systems in the cement,

coal, and power production industries; as sources to start nuclear reactors; in nondestructive materials analyses; and in medical research and health care applications such as cancer treatment.

The production of Cf-252 requires both a high-flux reactor and a unique feedstock. The Am-243 and Cm-244 in the Mk-42 materials are attractive feedstocks for irradiation to produce heavy-actinides, as shown in Fig. 1. Since Cf-252 has a short half-life (2.6 years), it decays at a rate of about 25% per year, and new supplies must be manufactured regularly to meet the various user communities' needs. In the process of producing Cf-252, other heavy actinides are produced as by-products. These include Bk-249, Es-254, and Fm-255. In 2009–2010, Bk-249 was recovered from a Cf-252 production campaign and used in a multinational collaborative discovery of element 117 [1].

The actinides contained in the Mk-42 targets were produced by a long-term irradiation campaign in the SRS K Reactor that used ~33 kg of weapons-grade plutonium (WGPu) approximately 30 years ago. Replacement of these actinides would require similar irradiation conditions and amounts of feed material. Since the K reactor has been shut down, irradiation in either the High Flux Isotope Reactor (HFIR) or Advanced Test Reactor (ATR) would be required. Studies by SRS estimate about 40 years of irradiation would be necessary to make Pu-242 from WGPu in these reactors [2]. The main issue confronting irradiation of WGPu to produce Pu-242, Cm, and Am-243 is the fission of Pu-239 and Pu-241. Approximately 64% of the original Pu-239 fissions when producing Pu-240, and 26% of the plutonium fissions as Pu-242 is produced. Overall, about 90% of the original Pu-239 mass is lost to fission as it is irradiated to produce Pu-242. This is shown graphically in Fig. 2. The fission loss requires that a large amount of Pu-239 be irradiated to make Pu-242, and the high fission rate limits target loading due to the fission heat that must be removed to avoid melting of the target. Current reactors (ATR and HFIR) do not have the large capacity of the SRS production reactor, and hence, the irradiation time requirements could be significantly longer than the original SRS production effort.

The Am/Cm/Pu produced in the SRS K Reactor can no longer be produced with the existing US infrastructure. Programs which require heavy actinides for research and industrial applications are relying on the existing supply of Pu and Cm as their feedstock. Disposition evaluations must, therefore, take into account that the materials contained in the Mk-42 targets are unique and may never be made again.

The Mk-42 Inventory at ORNL

Mk-42 targets were fabricated from Pu-239 and irradiated at SRS to make Pu-242, Am-243, and Cm-244. The ten targets allocated for ORNL were shipped from SRS to PNNL 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, and the recovered plutonium was shipped to customers.

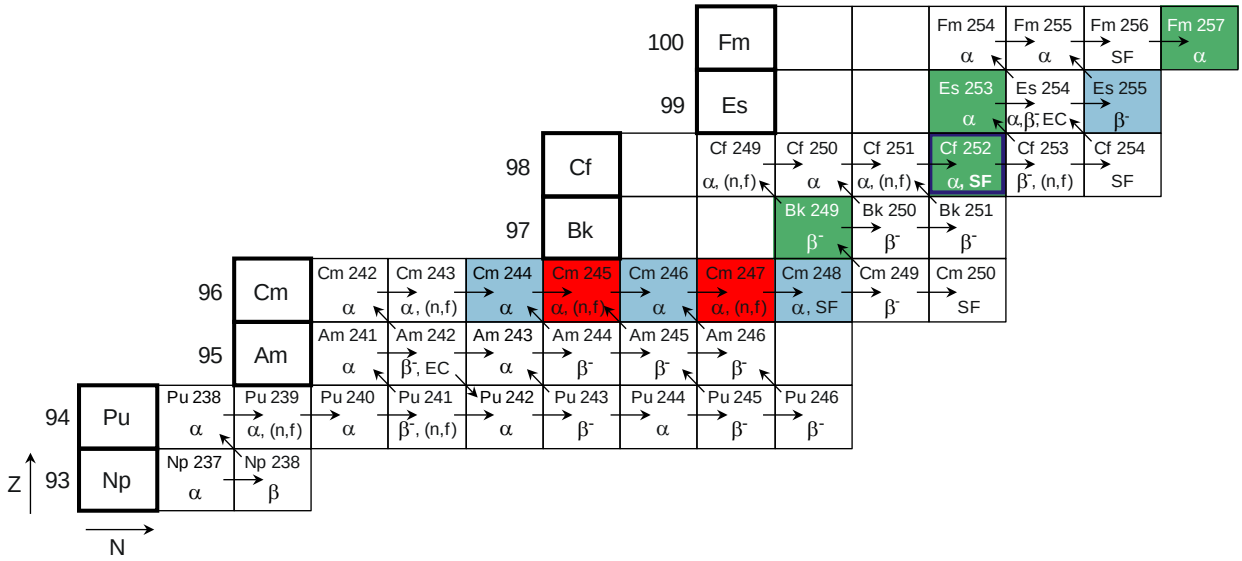


Fig. 1. Production of heavy actinides from Am-243 and Cm-244.

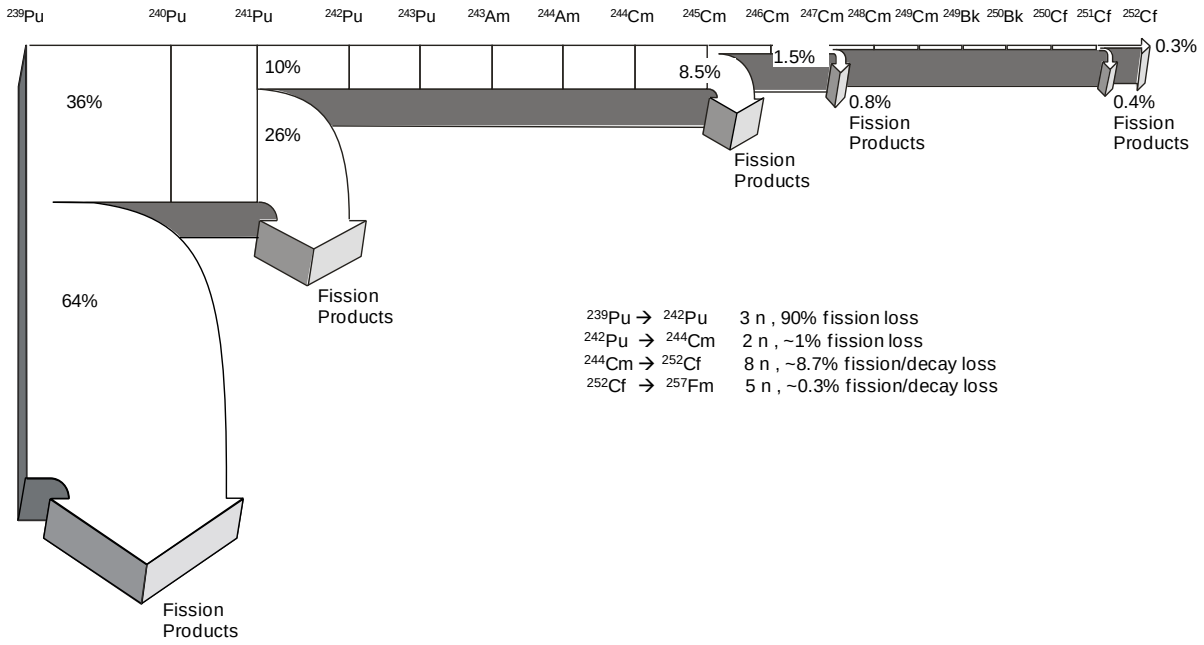


Fig. 2. Fission loss during thermal neutron irradiation of plutonium.

Early in the program, the program sponsor requested that ORNL recover only the Pu-242 and hold the Am/Cm for later use. Based on that request, ORNL recovered and purified the Pu-242 (~2.2 kg has been shipped, and another ~1 kg remains in unprocessed segments) and stored the remaining Am/Cm material that contained lanthanide (Ln) fission products, i.e., rare earth elements of atomic numbers 57–71. This mixed Am/Cm/Ln fission product material was solidified and transferred to small, welded stainless steel capsules and put in storage.

These Am/Cm/Ln materials contain significant quantities (~3 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. A flowsheet was developed and demonstrated from 2011 to 2013 to remove the bulk of the Ln fission products [3]. The flowsheet, shown in Fig. 3, consists of an oxide dissolution step, a PUREX (plutonium uranium redox extraction) process to separate the Pu from the Am/Cm/Ln, a Ln partitioning step to remove the Ln from the Am/Cm-rich stream, a Cleanex (Cleanup Extraction) step to remove complexants from the Am/Cm rich stream, 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 in from the decay of Cm-244. The Reverse TALSPEAK (Trivalent Actinide–Lanthanide Separation by Phosphorus reagent Extraction from Aqueous Komplexes) process is being used to separate the Ln from the Am/Cm elements. After the PUREX separation, the Pu rich stream is purified by anion exchange followed by an oxalate precipitation step to convert it to an oxide form.

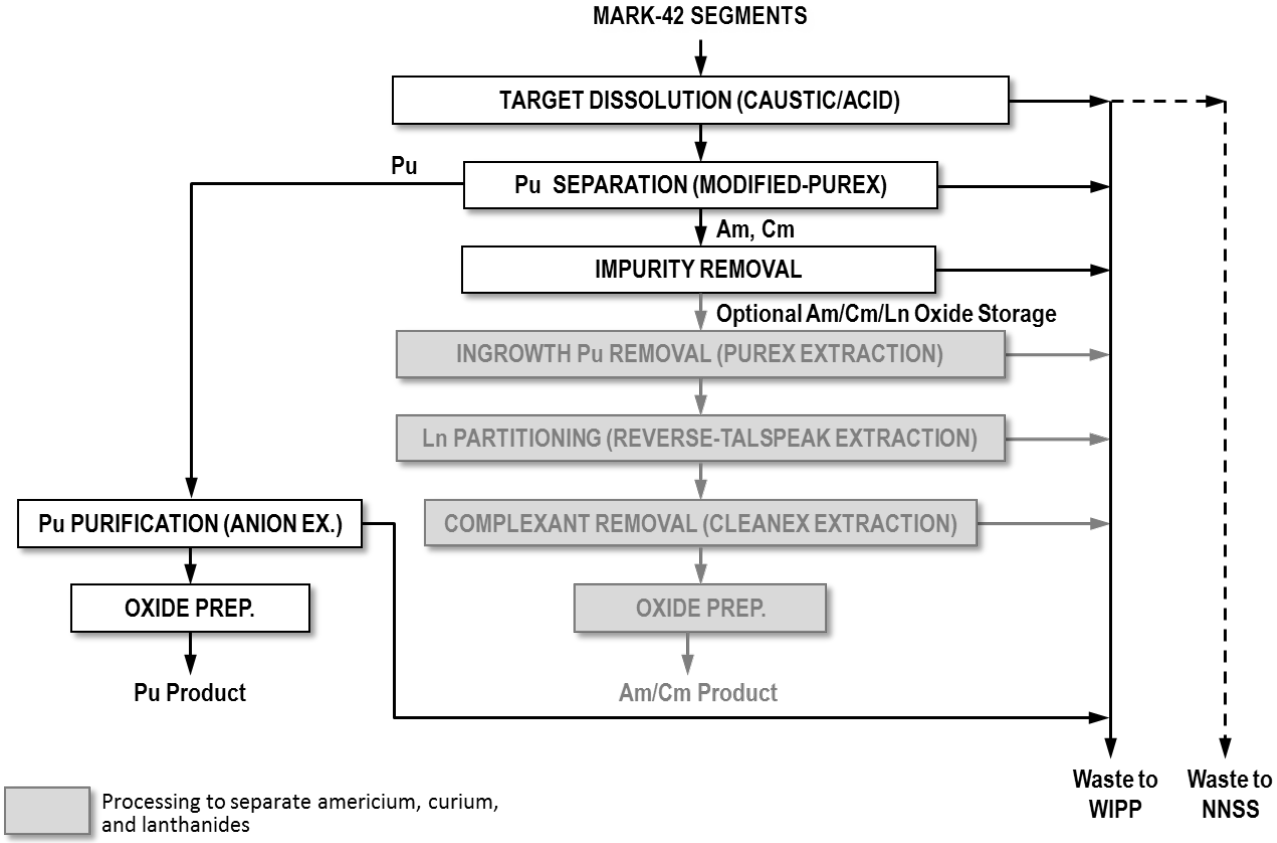
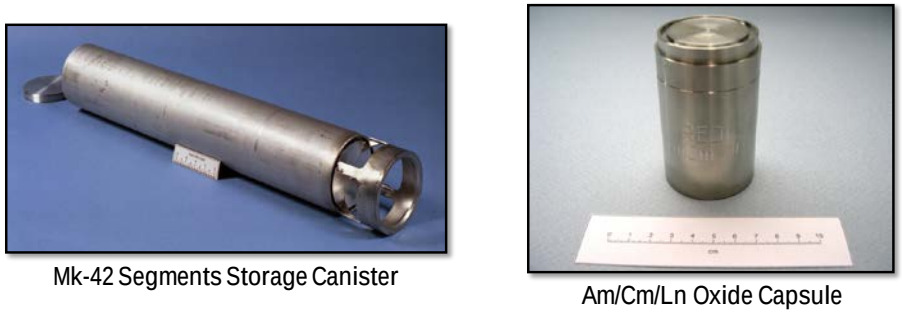


Fig. 3. Mk-42 separations process flow sheet.

Currently three Mk-42 materials are in storage at ORNL: (1) unprocessed Mk-42 segments, (2) capsules containing partially processed Am/Cm/Ln oxides (processing stopped just before the grayed portion of the flowsheet in Fig. 3), and (3) capsules containing fully processed Am/Cm oxide materials available for use as feedstock for heavy actinide production. Examples of the unprocessed target

segments and capsules containing the Am/Cm and Am/Cm/Ln oxides are shown in Fig. 4, and the inventory of materials is summarized in Table I.



Mk-42 Segments Storage Canister

Am/Cm/Ln Oxide Capsule

Fig. 4. Mk-42 target segments and oxide capsules in storage at ORNL.

TABLE I. Estimated inventory of Mk-42 (decayed to October 1, 2022)

Item	Fission Products	Plutonium (grams)	Americium (grams)	Curium (grams)
28 unprocessed target segments	~20 kCi mixed	1200	312	74
74 Am/Cm/Ln oxide capsules	3,290 grams lanthanides only	88	357	87
6 Am/Cm oxide capsules	~45 grams lanthanides only	3	128	34

DESCRIPTION

Although the isotopes in the Mk-42 materials have value, there is continual pressure to look at de-inventorying material in storage to only retain the most valuable materials needed to meet mission needs and to allow other materials to be added to the inventory as they become available. A technical evaluation of disposition options for the various Mk-42 materials in storage at ORNL is presently underway. The evaluation includes options for recovering Pu, Am, and Cm isotopes and estimates the resulting reduction in the amount of waste that would be sent to disposal facilities compared to processing the waste for disposal without recovering the actinides. Potential waste streams include transuranic waste, liquid high-activity waste, and irradiated items with fission products that could potentially be disposed of directly.

Disposition Options for Mk-42 Segments

The options being evaluated for disposal of the unprocessed Mk-42 target segments include

- processing the irradiated materials for disposal at the WIPP by retrieving the target segments from storage, dissolving them, solidifying as a homogeneous waste form, and packaging them for disposal. Two options for waste disposal

are being considered: (1) homogeneous mixing of all waste for disposal at WIPP, and (2) segregating the caustic dissolver waste (which primarily contains cesium and is likely to be the only non-transuranic waste stream generated) for disposal at the NNSS and solidifying the remaining material for disposal at WIPP; and
- recovering the Pu and/or Am/Cm using the flowsheet in Fig. 3 and solidifying the remaining material and packaging it for disposal at WIPP.

The options for disposal of the partially processed Am/Cm/Ln oxide capsules include
- retrieving the materials for storage, processing the materials as required to meet WAC, and packaging them for disposal at WIPP, and
- processing the material to recover the Am/Cm using the grayed portion of the flowsheet in Fig. 3 and solidifying and packaging the lanthanide fission products for disposal at WIPP.

The option for disposal of the completely processed Am/Cm oxide capsules includes
- retrieving the materials from storage, processing the materials as required to meet WAC, and packaging them for disposal at WIPP.

Recovery Options

Recovery of the actinides would involve processing the materials to recover and purify the Pu and/or the Am/Cm for future use. 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 in the Mk-42 segments would be recovered, processed to an oxide, and stored for future use.

The flowsheet in Fig. 3 would be used to process the unprocessed Mk-42 segments to recover Pu and Am/Cm. Historical processing data are being used to determine the waste generated from segment dissolution and recovery of the Pu material.

The grayed portion of Fig. 3 would be used to recover the Am/Cm material from the Am/Cm/Ln capsules. Data from a series of demonstration campaigns conducted to refine the grayed portion of the flowsheet are being used to estimate waste generated from recovering Am/Cm from segments and the Am/Cm/Ln capsules [4]. The first demonstration campaign was completed in FY 2011 using one of the Am/Cm/Ln oxide capsules containing ~4 g of Am, ~2 g of Cm, and ~60 g of Ln fission products. The demonstration run showed satisfactory results with a Ln decontamination factor (DF) of 34 for the Am/Cm product. A second campaign was performed in early FY 2012 in which nine capsules containing ~34 g of Am, ~14 g of Cm, and ~325 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. A Ln DF of 22 was achieved for the Am/Cm product. A third campaign was completed in FY 2013 in which 20 capsules (full scale) containing ~59 g of Am, ~26 g of Cm, and ~600 g of Ln fission products were processed using the same stream concentrations demonstrated in the second campaign. A Ln DF of 21 was achieved

for the Am/Cm product. The recovered material is adequate for heavy-actinide feedstock.

DISCUSSION

For this analysis, it was assumed that WIPP-bound waste forms would be RH-TRU and packaged in 208 L (55 gal) drums with three drums at a time being transported to WIPP in a 72-B Type B shipping container. The waste streams for each of the disposal options described above were evaluated against the U.S. Department of Transportation shipping criteria and WIPP WAC for RH-TRU waste [5, 6]. The limiting WAC was used to estimate the minimum number of shipments that would be required to dispose of the Mk-42 waste. The dose rates on the surface of the 208 L (55 gal) drums were limited to 100 R/h to assure that no more than 5 percent by volume of the RH-TRU received at WIPP may have a surface dose rate in excess of that value, a WIPP requirement. The decay heat limit was not assumed to be a limiting factor for disposal based on historical experience with similar waste streams. This will need to be confirmed as more detailed evaluations are performed in the future. The resulting number of RH-TRU shipments required to dispose of the Mk-42 waste streams is given in Table II as a function of recovery of Pu and Am/Cm.

TABLE II. Estimated RH-TRU shipments to WIPP

Material	Recovery Option		
	No Recovery	Pu Recovery	Am/Cm Recovery
28 unprocessed target segments	72	72	72
28 unprocessed target segments if caustic dissolution stream is segregated[1]	6	6	6
74 Am/Cm/Ln oxide capsules	4	NA	2
6 Am/Cm oxide capsules	1	NA	NA

[1]Disposal at NNSS as RH non-TRU waste.

For the unprocessed target segments, the dose rate on the surface of the drum is the limiting WAC for the waste generated by mixing homogeneously the waste from each processing unit operation in Fig. 3. The dose rate is controlled by the fission products in the waste. Therefore, the number of shipments to WIPP will not be reduced by recovery of Pu and Am/Cm material from the target segments.

The largest contribution to dose rate on the surface of the drum comes from cesium, which is primarily contained in the caustic dissolution waste stream. If this stream were to be segregated and treated separately, it could potentially be a RH non-TRU waste stream which could be disposed of at NNSS. The RH-TRU waste stream resulting from solidifying the remaining waste streams shown in Fig. 3 drops the amount of waste requiring disposal at WIPP by more than 90%. If the target

segments were processed without recovering any of the actinides, six shipments to WIPP would be required. The surface dose on the drum is still limiting, and the number of shipments to WIPP will be six whether or not actinides are recovered from the target segments.

The Pu-239 equivalent activity limit (PE-Ci) for a solidified/vitrified waste transported in a 72-B waste canister is ≤1,800 Ci, and for any other waste form it is ≤240 Ci. If the oxide material in the capsules is assumed to be disposed of in a powder form, the PE-Ci limit will be the controlling factor and would result in 14 shipments for the Am/Cm/Ln capsules and six shipments for the Am/Cm capsules to be disposed of at WIPP. If the materials are solidified, the hypothetical accident condition (HAC) for the 72-B becomes the controlling factor, which will result in four and one shipments, respectively. If the Am/Cm is recovered from the Am/Cm/Ln capsules, the resulting waste for disposal at WIPP is cut in half; the HAC is the limiting factor.

Evaluation results to date indicate that recovery of the Pu, Am, and Cm from the unprocessed target segments will not impact the amount of waste requiring disposal. Segregating the cesium-rich caustic dissolution waste stream from the TRU-rich waste streams would reduce the amount of waste going to WIPP by over 90%, but it would result in a second RH waste requiring disposal at NNSS. Additional evaluations are needed to determine which disposal route would be best.

It may also be worth evaluating whether the unprocessed target segments could be disposed of directly without having to dissolve them and solidify them into a homogeneous waste form. Since the dose rates determine the minimum amount of this material that can be shipped to WIPP in a package, direct disposal would require the same amount of shipments to WIPP. It could, however, substantially reduce the capital and operating costs to ready the material for disposal.

Disposal of the Mk-42 oxides that are stored in capsules would require a minimum of five WIPP shipments. Processing the Am/Cm capsules to recover the Am/Cm from the material would cut the waste disposal for that stream to three shipments and produce a recovered material that could be used for Cf-252 feedstock without additional processing.

CONCLUSIONS

The Mk-42 materials stored at ORNL contain valuable Am-243 and Cm-244, which are attractive feedstocks for production of heavy actinides (Cf-252, Bk-249, etc.) and Pu. For all practical purposes, they are irreplaceable because the reactor capability required to replace the existing inventory of these materials no longer exists. Although the isotopes in the Mk-42 materials have value, there is continual pressure to look at de-inventorying material in storage to only retain the most valuable materials needed to meet mission needs and to allow other materials to be added to the inventory as they become available. A technical evaluation of disposition options, including processing to recover select isotopes from the various Mk-42 materials in storage at ORNL is presently underway.

The order of preference for retaining the Mk-42 materials is (based on the isotopic content and the amount of processing required to make them useful as feedstocks for customer programs): Am/Cm capsules, Am/Cm/Ln capsules, and unprocessed target segments. Based on the complexity of the processing required to prepare the materials for disposal and the volume of waste that would be produced, from a waste management prospective only, the preference for disposal would be in this same order. Additional evaluations will be required to determine the optimum path forward should some or all of the Mk-42 materials be dispositioned in the future.

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