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SRNL Development of Recovery Processes for Mark-18A Heavy Actinide Targets

Jeffrey S. Allender, Nicholas J. Bridges, Bradley M. Loftin, and Michael D. Dunsmuir

Savannah River National Laboratory
Aiken, SC, USA

ABSTRACT

Savannah River National Laboratory (SRNL) and Oak Ridge National Laboratory (ORNL) are developing plans for the recovery of rare and unique isotopes contained within heavy-actinide target assemblies, specifically the Mark-18A. Mark-18A assemblies were irradiated in Savannah River Site (SRS) reactors in the 1970s under extremely high neutron-flux conditions and produced, virtually, the world's supply of plutonium-244, an isotope of key importance to high-precision actinide measurement and other scientific and nonproliferation uses; and curium highly enriched in heavy isotopes (e.g., curium-246 and curium-248). In 2015 and 2016, SRNL is pursuing tasks that would reduce program risk and budget requirements, including further characterization of unprocessed targets; engineering studies for the use of the SRNL Shielded Cells Facility (SCF) for recovery; and development of onsite and offsite shipping methods including a replacement for the heavy (70 ton) cask previously used for onsite transfer of irradiated items at SRS. A status update is provided for the characterization, including modeling using the Monte Carlo N-Particle Transport Code (MCNP); direct non-destructive assay measurements; and cask design.

INTRODUCTION

Mark-18A Heavy Actinide Targets were irradiated in the Savannah River Site (SRS) K Reactor to support the production campaign for californium-252 (^{252}Cf).[1] Twenty-one of 86 irradiated assemblies were processed in the 1970s to produce the world's supply of plutonium-244 (^{244}Pu) and U.S. supply of heavy curium, both of which have numerous scientific and forensic applications. The supplies recovered in the 1970s are depleted, but another 65 irradiated assemblies are available for processing to extract valuable transuranium isotopes and potentially valuable fission products.

The ^{244}Pu in these targets was produced in a special high-neutron-flux campaign at SRS, under conditions that cannot be duplicated, and the inventory of isotopes cannot be replaced with any capabilities that exist today. The isotope in the targets was designated a "National Resource"[2] in 2001 and was reconfirmed as a key international asset in the 2011 Department of Energy (DOE) National Strategic Plan for Nuclear Materials.[3]

Beginning in 2012, the Office of Nuclear Materials Integration (ONMI), within DOE's National Nuclear Security Administration, led a study with ORNL and SRNL to evaluate disposition options for the targets;[4] potential recovery schedules and costs; and the formation of an Interagency Working Group to confirm interest and requirements for programmatic uses inside and outside the U.S. Government agencies, including the international community represented by the International Atomic Energy Agency.[5] In 2015, ONMI obtained agreements to pursue a program to prepare the targets and recover the heavy isotopes.

A series of option evaluations concluded that the optimum pathway for the Mark-18A targets is to remove them from storage in the SRS L Basin, to transfer them to the SRNL SCF for chemical processing to remove cladding and stabilize actinide oxides, and then to transfer the recovered plutonium oxide (with the ^{244}Pu) and a curium/americium/fission product oxide (with the heavy curium) to ORNL for future conversion into forms that are ready for programmatic use.

In Fiscal Year 2015, SRNL and ORNL are pursuing R&D to enable the operation in the Shielded Cells and to reduce program risk. Both the evaluations of program requirements and the recovery operations will provide unique opportunities for the advancement of actinide science and the science of long-term and high-flux reactor operations. Key scientific focus areas include:

- Improving estimates of transplutonium isotopes and fission products produced in unique targets. These studies support process design by providing a basis for radiation protection, but also will identify additional isotopes that may be required by the scientific community. In 1970, the primary focus of the production campaign was to supply ^{252}Cf for use inside and outside the DOE, but byproduct isotopes may also be identified in these and other types of targets stored in L Basin.

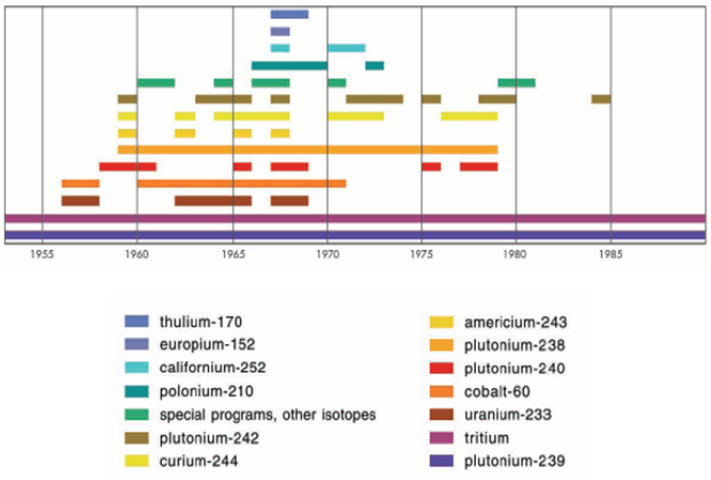
- Calibrating and confirming computer codes for the prediction of isotope production in the unique reactor configurations, which may also be applicable to future activities at the ORNL High Flux Isotope Reactor (HFIR). Current inventory estimates are based on computer codes for isotope depletion and production that have been used for nearly 50 years worldwide, but that are being replaced by more modern calculational methods.

- Non-destructive analysis (NDA) of selected targets in Basin storage to confirm the integrity of targets exposed to very long irradiations, including the impacts on aluminum cladding materials that will have been converted partially to silicon. The NDA will provide a confirmation of the actinide and fission product estimates from the computer simulations.

- Based in part on the improved isotope inventory knowledge, SRNL is modifying the design of an existing 70-ton shipping cask that is used onsite to transfer irradiated fuels to H Canyon. The dominant radiological impact from the Mark-18A targets is from neutrons, not gamma energy from fission products, and light-element shielding suitable for neutron protection will replace the much heavier lead shielding required for gamma protection.

ISOTOPE PRODUCTION

Figure 1. Major Isotope Production Campaigns at SRS



Through the history of materials production at SRS, DOE and its predecessor agencies devoted a portion of the reactor capacity to the production of special isotopes with key properties, both for National Defense applications and for uses outside the U.S. Government.[1] Figure 1 shows the progression of major isotope production campaigns. As early as 1956 significant quantities of ^{242}Pu were produced by continuing to irradiate plutonium beyond the boundaries of "weapons-grade" or "fuel-grade" composition, and ^{60}Co was created for heat-source or irradiation-source applications. In 1959, a portion of the accumulated ^{242}Pu was irradiated in several Curium Campaigns to create ^{244}Cm and ^{243}Am. This time period saw the first focused, quantitative, production of ^{238}Pu for space and terrestrial applications.

Following the initial Curium Campaigns, SRS performed further research involving irradiations of high-isotopic-content plutonium to create heavier isotopes, most notably ^{252}Cf. The site developed the first significant DOE effort to transfer heavy isotopes to industry, health care, and education and developed the first implants containing ^{252}Cf for use in cancer therapy. During this time, SRS authorized a High Flux Demonstration in the site's K Reactor, where fuel tubes were cycled frequently and neutron fluxes reached a record 7 n/cm^2/sec, a level impossible to recreate in any reactor that is operating today. This high-flux operation was also used to produce 150 isotopes of 66 elements for research at nine universities and laboratories. The Mark-18A targets underwent high-flux irradiation for one year and some were retained in K Reactor for up to ten years to allow the further concentration of transplutonium isotopes such as californium and heavy curium.

Figure 2 shows the benefit of irradiating ^{242}Pu and other higher isotopes for the purpose of creating even heavier isotopes.[6] If the starting point for irradiation is weapons-grade or fuel-grade plutonium that is dominated by ^{239}Pu, up to 90% of the starting isotopes are lost to fission products. Only about 1.5% of the starting material becomes a target for further neutron capture, as "heavy curium" (generally made up of ^{246}Cm and ^{248}Cm), berkelium and californium, and eventually a small fraction of even heavier isotopes. Recovered material with a high content of heavy curium is of particular interest as starting material for the production of high-atomic weight isotopes.[4]

Table 1 shows the content of potential inventories that could be used for new heavy isotope production in HFIR. Figure 3 shows the relative efficiency predicted for ^{252}Cf production. Without the availability of the heavy curium from the Mark-18A targets, the best way to provide feed for new production may be to irradiate "light curium" to raise its isotopic content, process to chemically concentrate the heavier isotopes, and fabricate new targets for the ultimate production.

Figure 2. Efficiency of Heavy-Isotope Production from ^{242}Pu

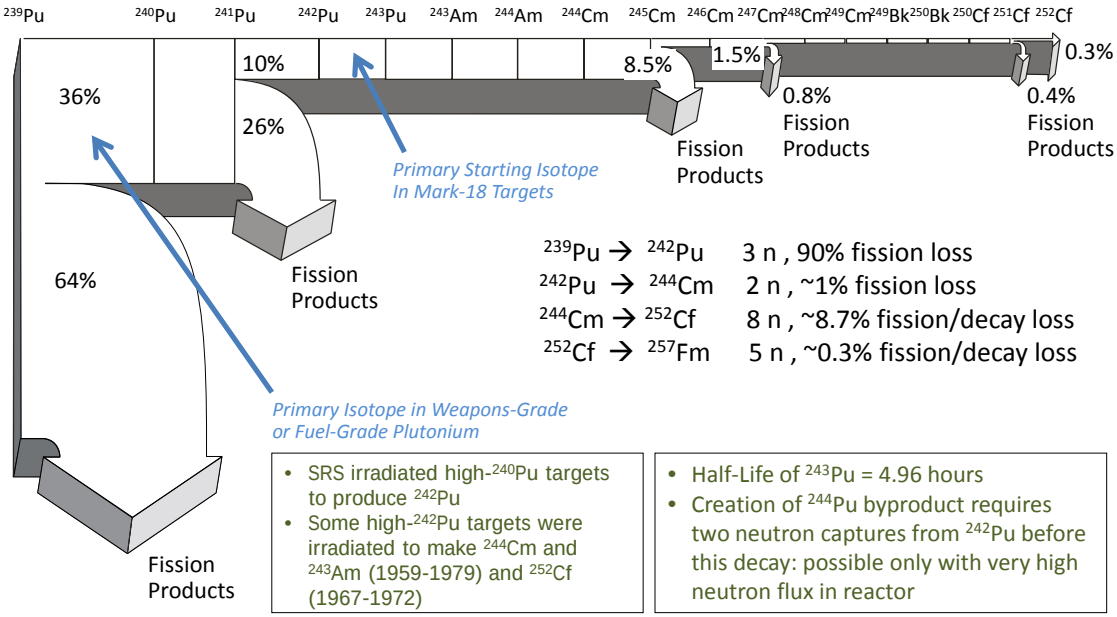
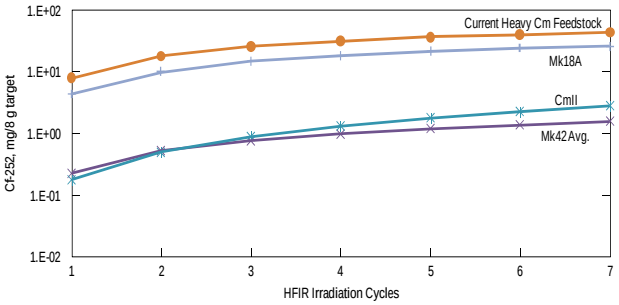


Table 1. Isotopic Abundance in Potential HFIR Feeds

	Mark-18	Curium-II	Mark-42	SRS Cm
^{244}Cm, %	20%	80%	81%	66%
^{245}Cm, %	1%	3%	7%	12%
^{246}Cm, %	72%	16%	11%	20%
^{248}Cm, %	7%	0%	1%	1%
Total Cm, grams	667 g	255 g	260 g	91 g
^{244}Pu, grams	22 g	0 g	0 g	0 g

Figure 3. Californium Production Efficiency vs. Feedstocks



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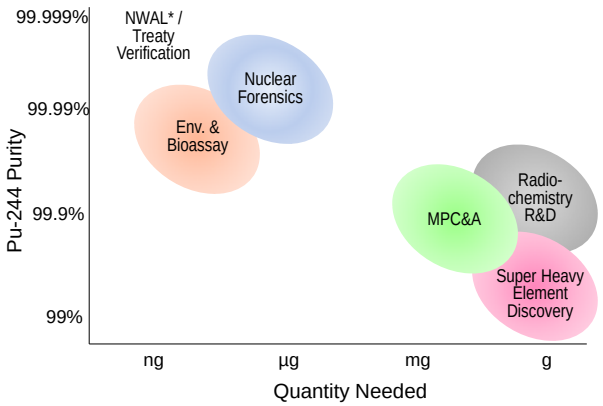
The high-flux operation provided yet another byproduct: ^{244}Pu. This isotope is virtually stable with a radioactive half-life of 81 million years. It is not produced in significant quantities under any other conditions because it requires the capture of two neutrons in nuclei of ^{242}Pu. First ^{243}Pu is produced, but this isotope has a decay half-life of less than 5 hours, and a high neutron flux is necessary to allow the activated ^{243}Pu nucleus to capture a second neutron before it decays to ^{243}Am, which is the major precursor for neutron capture to produce the heavier isotopes (as shown in Figure 2).

Plutonium-244 has become increasingly important because it does not exist within any other inventory of plutonium.[7,8] It is of central importance as a tracer and calibration isotope for high-precision plutonium analysis, especially nuclear forensics.[9] It is approximately 3 times as efficient as ^{242}Pu as a calibration source, and even more important because measurement of ^{242}Pu content can be significant in forensics evaluations.

The world's supply of ^{244}Pu was created in the Mark-18A targets. Current, almost-depleted supplies were recovered from 21 targets that were processed at ORNL in the 1970s. Figure 4 shows the ranges of assays that are used for current applications.[10] Other applications are proposed that would not require isotopic concentration, but those applications are judged to be less critical than the use in high-precision chemistry and analysis. After irradiation, the ^{244}Pu has been diluted because of the decay of ^{244}Cm into ^{240}Pu and now makes up only 5% of the remaining plutonium. For full proposed use of the isotope, a further step to enrich the isotopic concentration will be required.

Figure 4. Known Demand for ^{244}Pu and Isotopic Purities



*NWAL - Network of Analytical Laboratories

FLOWSHEET VALIDATION

Figure 5 shows the configuration of the target within its "J-Tube" carrier. Figure 6 shows the major steps in the processing flow for the Mark-18A retrieval, recovery, and storage. The first chemical operation within the Shielded Cells is to dissolve aluminum, including target cladding and meat and the J-Tube, with caustic and thereby reduce the volume of material to process further. No previous process has confirmed the details of this step using targets that were irradiated for a decade.

Figure 5. Mark-18A Target Inside J-Tube

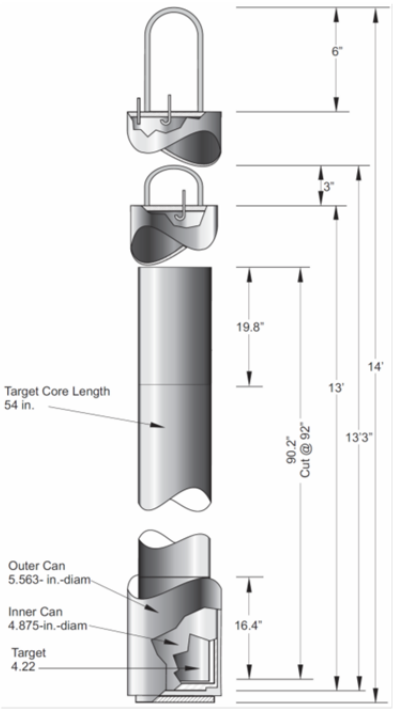
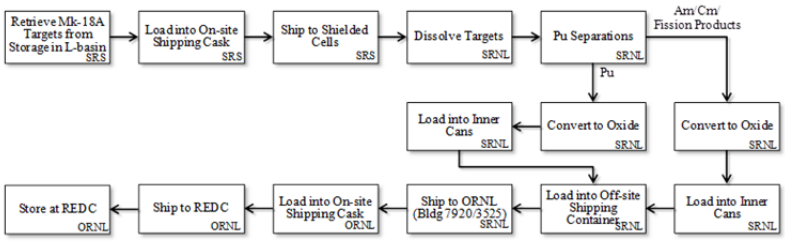


Figure 6. Mark-18A Target Recovery Flow Diagram



Simulation tests and the first target dissolutions will confirm the behavior of the matrix with respect to undissolved metals and the removal of fission products. Most of the remaining solids will be oxides, which will be dissolved and separated by ion exchange before two oxides are produced, one of plutonium and one of americium/curium/fission products. Surrogate testing will demonstrate the efficiency of dissolving and the potential benefits of peroxide fusion pretreatments.

TARGET CONTENT CHARACTERIZATION

The Mark-18A targets were irradiated in different reactor positions and for varying time periods. The concentrations of actinides were estimated in the 1970s using reactor-modeling codes in use at the time, and were only partially verified by the assay of the 21 targets processed in 1971 at ORNL. Figure 7 shows estimates for a "typical" target using a range of reactor assumptions and two versions of the ORIGIN computer code. Figure 8 shows the scatter of results using two computer codes and two 1971 processed targets.

Figure 7. SRNL and ORNL Actinide Calculations using ORIGEN

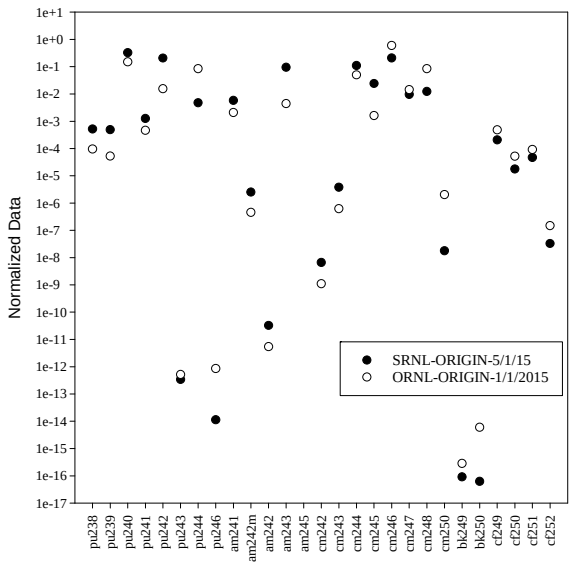
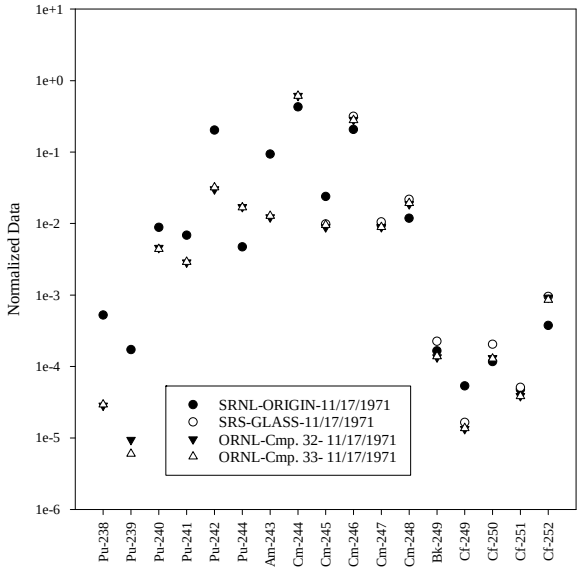


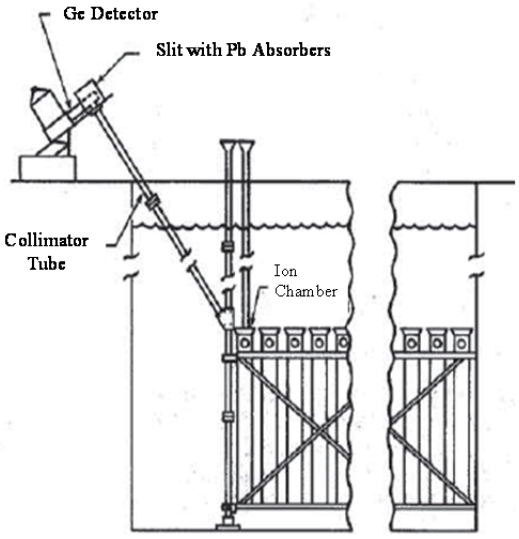
Figure 8. Initial Computer Calculations vs. 1971 Processing



SRNL is re-estimating the actinide contents of each remaining target using the modern Monte Carlo N-Particle (MCNP) transport code.[11] Because the focus of the 1971 estimates was on heavy isotopes, the new calculations will provide significant insights into the fission-product inventories, supporting both shielding studies and the evaluation of further byproduct materials.

Several of the targets will be analyzed *in situ* in underwater storage at L Basin to confirm the calculations and demonstrate the continued integrity of the J-Tubes and cladding. Figure 9 shows the configuration that will be used for the NDA.

Figure 9. Non-Destructive Analysis in L Basin



SHIPPING CASK MODIFICATION

Figure 10. Current 70-Ton Cask in L Basin

One key task is to optimize the configuration of the cask that will be used to transport the targets from L Basin to the SCF. The cask that is commonly used for irradiated materials transfers at SRS (Figure 10) is a large, heavy (70-ton; 62.5 metric tons) cask with considerable lead shielding to protect workers from the gamma radiation typical of fission products. This cask is not compatible with remote-handling cranes and current floor weight limits at SRNL.

Therefore, SRNL is modifying the cask design to replace most of the lead gamma shielding with polyethylene neutron shielding, more appropriate for the radiation fields of these targets. The resulting cask will use existing fixtures for transfer of assemblies in J-Tubes from underwater storage, with a weight less than 25 tons (22.3 metric tons).

Figure 11 shows an open view of the new cask. Figure 12 depicts the modified cask and its configuration after it is transferred to the SRNL Shielded Cells.

Figure 11. Conceptual Design – Modified Cask in L Basin

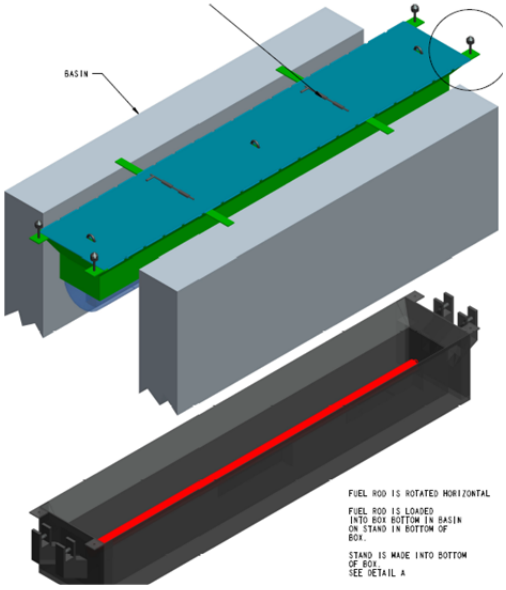
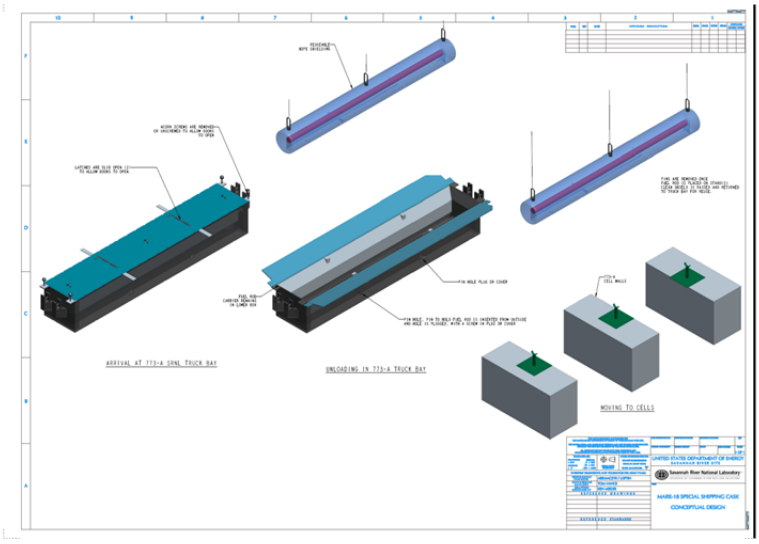


Figure 12. Conceptual Design – Modified Cask in SRNL Shielded Cells



PATH FORWARD

SRNL would receive and process a single target per month, with the potential to manage up to two of the least-irradiated targets, with a nominal output of oxide from 9 targets per year. Design development, component testing, and facility evaluations are projected to be complete by Fiscal Year (FY) 2021 at projected funding levels, at which time the first product transfers are expected. The campaign could extend through FY2028 if all 65 of the remaining targets are processed.

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