#### **Contract No:**

This document was prepared in conjunction with work accomplished under Contract No. 89303321CEM000080 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

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#### Savannah River Site Items of Interest for National Nuclear Material Archive

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#### **Abstract**

The Savannah River Site (SRS) holds legacy materials from its nuclear materials production and processing missions, and also materials received from other locations to support Fissile Materials Disposition and research and development, including recovery/reuse and nuclear forensics. The National Nuclear Material Archive (NNMA), operated by the National Nuclear Security Administration (NNSA), is identifying; sampling; and analyzing typical and rare items to build a library of characterization data on historical process streams. Savannah River National Laboratory (SRNL) renewed a screening of "Items of Interest", including items proposed for disposition (e.g., surplus plutonium and enriched uranium) and historical samples, to assist NNMA in selecting items for inclusion in the Archive and for the activities required to complete analysis in cooperation with NNSA and other National Laboratories.

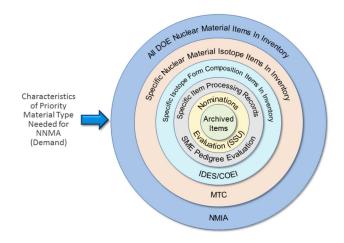
# **Background**

The National Nuclear Material Archive (NNMA) program is operated by the Office of Nuclear Forensics. Its function is to establish the capability to identify, characterize, evaluate, and determine the origins of nuclear materials. The Archive seeks to collect and preserve representative physical samples for analysis, evaluation, reference, and comparison, especially to assess whether nuclear materials that are discovered outside regulatory control are consistent with a DOE origin.

The scope of interest is broad, spanning historical weapons-related production, but also addressing fuel cycle materials and naturally occurring materials. Characteristics evolve over the breadth of management from mining, separation, irradiation, fabrication, recycle, and recovery. The time of production and use is also important: Processes implemented in the 1950s may differ from those that would be used today.

The NNMA developed a detailed matrix of precursor, in-process, and product accountable materials that are currently under DOE/NNSA custody, related to materials utilization, science, and fuel cycle research. The matrix, documented in a series of "Collection Catalogues," established a "demand" for candidate items for transfer to the Archive to fill the slots in the matrix. Once demand is identified, the potential "supply" of items is matched to the matrix. This step evaluates the process knowledge that could show whether an item is typical of a physical or chemical category, the chain of custody, and the provenance of prior characterization and process history.

Based on the Collection Catalogues, DOE/NNSA sites identify "Items of Interest" (IoI) that may be candidates to represent a category in the demand matrix. These Items are then evaluated for whether they should be considered for a nomination for use in the Archive. Nominated items that are selected are then evaluated for the long-term program of sampling, analysis, storage, and often transportation. The figure shows how the process proceeds through the identification phase through archiving.<sup>1</sup>



#### Status

Savannah River Site has more than 70 years of involvement with nuclear materials production and processing for most components of the matrix, from production and recycle supporting weapons programs, from fuel cycle research, and from nuclear science and engineering. It is also the designated consolidation site for plutonium-related materials that are achieving disposition following the end of the Cold War and the deactivation of legacy operations, including those at Rocky Flats and Hanford. Although plutonium-related items may constitute the largest bulk of IoI candidates, the site also holds uranium-related items from reactor development and samples from production and processing campaigns for other isotopes.

SRNL evaluated the entire site inventory of accountable nuclear materials, identified IoI that may meet the needs of the demand matrix, and noted other items that could be added to the initial demand matrix. This IoI list is updated annually and items may be added on request when identified by the NNMA program. This list will be revised again during the development of the Nuclear Materials Inventory Assessment (NMIA) for the end-of-fiscal-year-2021 inventory. The NMIA process is performed at the item level to fulfill requirements of DOE Order 410.2, *Nuclear Materials Management*.<sup>2</sup>

The first items nominated and accepted were slugs of fuel from the Oak Ridge X-10 reactor, perhaps the oldest nuclear production material remaining.<sup>3</sup> In 2010 SRNL agreed to receive and store legacy slugs that were previously managed at Brookhaven National Laboratory but that no longer had a defined use there. The purpose of this transfer was specifically to maintain rare and unique materials for the NNMA, which at the time was coordinated by the DOE Office of Intelli-

gence and Counterintelligence. Non-destructive assay of these slugs is completed and laboratory analysis is planned by NNMA and other programs.<sup>4</sup>

In 2021, SRNL submitted nomination packages for two items from legacy Rocky Flats production operations. These nominations are pending.

## **Identifying Items of Interest**

After the NNMA program develops the matrix of material categories that represent all significant process groups and time periods ("demand"), sites evaluate their current inventories of nuclear materials to identify potential matches ("supply") and document the evaluation in the annual NMIA.

Once a potential candidate is identified as an Item of Interest (IoI), the NNMA selection team can express interest in an IoI. Then the site and other cognizant personnel can determine the degree of knowledge on the item, including knowledge of process history plus characterization provenance, with chemical and physical analyses of the specific item or the process stream that generated it.

## Bulk Plutonium from Production

SRS inventories include tonnes of surplus plutonium that were consolidated from discontinued operations at other DOE sites and planned for disposition, particularly plutonium materials. Most of such material was stabilized and packaged to the DOE Standard for long-term plutonium storage, DOE-STD-3013.<sup>5</sup> Key early focus areas include:

- Metals and oxides in product and intermediate forms from Rocky Flats weapons-related fabrication, recycle, and recovery. If not available elsewhere, stabilized examples of many categories are available to represent Rocky Flats operations in the 1980s.
- Metals and oxides in product and intermediate forms from Hanford fuel-cycle research, which can have a very large range of isotopic composition.

One complication for the NNMA program is that plutonium items may be stored in welded DOE-STD-3013 containers that hold several kilograms of material, when the Archive may require only several hundred grams. Future storage, sampling, and analysis may require coordination with another site that will receive the material for other use or for the NNMA specifically.

Key categories that are assigned IoI status include:

- Two specific items representative of Rocky Flats legacy production, nominated in 2021.
- Plutonium buttons from reprocessing of reactor targets to support weapons production up through 1989, which were fully characterized against Rocky Flats feed requirements but were not transferred there prior to their suspension of operations.
- Material from selected, retired weapons components that were transferred from Rocky Flats to SRS for packaging and storage prior to disposition as surplus.

- Oxide produced by the processing of weapons-grade metal as potential startup feed for the Mixed Oxide Fuel Fabrication Facility. This oxide of recent production is highly purified and analyzed.
- Fuel-grade ingots from Hanford. These ingots were sent to SRS with the intent of aqueous isotopic blending with "supergrade" reactor production to meet the Rocky Flats feed specifications.

## Bulk Plutonium from Fuel Cycle

Significant inventories of intermediate and product material remain from research at the Hanford Fast Flux Test Facility (FFTF) and other developmental reactors. Some of the items, including Mixed-Oxide fuels, were fabricated by commercial vendors and even overseas partners. Key categories that are assigned IoI status include:

- "Button Line Oxide" from Hanford, previously analyzed and confirmed to meet feed requirements for fabrication into FFTF fuel. It has considerable provenance for chemical and physical properties.
- FFTF Oxide Feed from British Nuclear Fuels Ltd. (BNFL). Although this material is of "foreign" origin, it is part of the U.S. inventory and historical production process.
- "Barter B" and "Barter C" material from the United Kingdom, acquired in the 1960s and 1970s and designated by the U.S. for use only in fuel cycle research. These items are also well characterized and of high purity.

Hanford also retained finished fuel assemblies that were fabricated by commercial vendors. These finished assemblies are not readily retrievable from large storage configuration and were not designated as IoI.

The British materials are identified for the NNMA but may, in the future, be managed separately under international cooperative programs.

## Samples and Standards from Reactor Production

Samples remain from previous production operations and engineering development. Often these samples are small, but they may be accompanied by previous laboratory analyses.

Key categories that are assigned IoI status include:

- Mini-tube samples from developmental Mark-14 fuel. The Mark-14 with cermet core was the basis for proposed new production fuel with reduced uranium enrichment and aluminum content.
- Scrap from the Mark-15 fuel process. This developmental fuel would have combined fuel and plutonium-production attributes into single slugs of low-enriched uranium.
- Scrap pieces from Heavy Water Components Test Reactor. This experimental reactor, which operated 1962-1964, was a testing prototype for a proposed civilian reactor design using normal uranium fuel.

# Other Samples, Standards, and Analytes

Similarly, samples from historical process development, including materials from offsite that were transferred to SRS for recovery, have been retained. Samples may be small, but may have historical and analytical provenance that could supplement analyses that would be pursued for Archived items. No additional items are identified at this stage of the NNMA evolution.

# **Legacy Irradiated Materials**

The NNMA does not seek irradiated materials. However, some items irradiated more than 50 years ago may have cooled to where they can be handled in the same way as unirradiated items. (A primary example is the X-10 reactor slugs, produced in the late 1940s/early 1950s.) Also, SRS stores special reactor targets from high-neutron-flux reactor campaigns and special production campaigns in the 1960s and 1970s. DOE programs may have interest in recovering isotopes from these targets: If processing for isotope recovery is pursued, samples from the recovery operations may be requested. No items are identified at this stage but the potential may exist if recovery is pursued.

#### Path Forward

SRNL will continue to support the NNMA program by maintaining the tracking of IoI, focusing first on any "bulk process categories" that may not have good representation. Smaller, unique inventories will be evaluated to determine whether there is forensic value in their archiving and characterization.

#### References

- 1. National Nuclear Material Archive, internal DOE/NNSA documents.
- 2. U. S. Department of Energy, "Nuclear Materials Management", Order 410.2, August 2009.
- 3. J. S. Allender, N. J. Bridges, L. T. Sexton, and D. M. Beals, "Screening of Items for National Nuclear Material Archive," proceedings of the 59<sup>th</sup> Institute of Nuclear Materials Management Annual Meeting, July 2018.
- 4. T. J. Aucott, "Non-Destructive Characterization of Irradiated X-10 Slugs," SRNL-STI-2019-00303, August 15, 2019.
- 5. U.S. Department of Energy, "Stabilization, Packaging, and Storage of Plutonium-Bearing Materials, DOE-STD-3013-2018, November 2018.
- 6. S. Robinson, J. Ezold, J. Neal, and B. Patton, "Planning for Receipt of Mark-18A Recovered Nuclear Material at Oak Ridge National Laboratory," proceedings of the Institute of Nuclear Materials Management/ESARDA Meeting, August 2021.