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DEVELOPMENT OF AN ALTERNATE PATHWAY FOR MATERIALS DESTINED FOR DISPOSITION TO WIPP

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ABSTRACT

The Los Alamos National Laboratory currently has an inventory of process residues that may be viable candidates for disposition to the Waste Isolation Pilot Project (WIPP) located at Carlsbad, New Mexico. A recent "Attractiveness Level D" exemption allows for the discard of specified intractable materials regardless of the per cent plutonium. However, the limits with respect to drum loadings must be met. Cementation is a key component of the aqueous nitrate flowsheet and serves as a "bleed-off" stream for impurities separated from the plutonium during processing operations. The main "feed" to the cementation operations are the "bottoms" from the evaporation process. In the majority of cases, the cemented bottoms contain less than the allowed amount per drum for WIPP acceptance. This project would expand the route to WIPP for items that have no defined disposition path, are difficult to process, have been through multiple passes, have no current recovery operations available to recover the plutonium and that are amenable to cementation.

This initial work will provide the foundation for a full scale disposition pathway of the candidate materials. Once the pathway has been expanded and a cementation matrix developed, routine discard activities will be initiated.

INTRODUCTION

With the dwindling of Security Category I nuclear material handling facilities throughout the DOE Complex in the recent past and given the near-saturated condition of nuclear material storage at the Los Alamos National Laboratory's (LANL) Plutonium Facility (PF-4), a study was conducted on the current state of storage at LANL and the potential paths for eliminating the risk associated with SNM storage congestion. The results of that study were documented in the *PF-4 Actinide Disposition Strategy* (PADS)[1].

The Plutonium Facility at LANL was constructed in the late 1970s, at a time when numerous other plutonium production facilities were operational (e.g., Rocky Flats, Hanford, Savannah River, and Lawrence Livermore National Laboratory). Consequently, PF-4, was constructed primarily as a research and development/prototyping facility with a vault sized and configured for that mission. As the other operational facilities have closed and/or de-inventoried their materials, PF-4 has continued to perform major missions, and the SNM storage capability has nearly reached its capacity.

The materials currently stored in the vault have been historically targeted for plutonium recovery. However, as containers are opened and inspected, some contents are determined to be resistant to recovery and may be candidates for disposition to WIPP. Some material continues to be stored in non-robust storage containers. This situation causes significant operational inefficiencies and presents an unacceptable level of risk to both current and future programs.

The current configuration of SNM storage in the PF-4 vault is inefficient and less than ideal. For example, material of little programmatic value is stored in the high security vault, taking up valuable space that might be used to support on-going projects. These legacy containers result in the requirement to wear respirators in the vault while handling containers. (Figure 1) This leads to daily inefficiencies and operational interruptions that unnecessarily increase programmatic cost and risk. Consequently, there is a strong motivation to develop or expand the pathways of these candidate materials to other storage facilities including the Waste Isolation Pilot Plant (WIPP) located at Carlsbad, New Mexico.

Figure 1 Operators in Respirators and Personal Protective Equipment (PPE)

Based on the PADS approximately one third of the containers in the PF-4 vault need to be reviewed for value to LANL programmatic missions and could be considered for offsite disposition. The vault in PF-4 contains materials categorized as most attractive AL "A" through least AL "E". A large fraction of the impure material in PF-4 is composed of process residues and non-combustibles. Many of these items are categorized, according to graded safeguards as AL "D". Graded safeguards is the concept of providing the greatest relative control, accountability, and protection for the types, quantities, and forms of special nuclear materials that can be most effectively used in a nuclear device or easily converted to such materials. The level of control and protection required is based on safeguards categories and ALs of the materials. [2]

SAFEGUARDS ATTRACTIVENESS

MC&A safeguards ALs are a ranking of various types and forms of special nuclear material based on their usefulness in constructing a nuclear weapon or an improvised nuclear device (IND). Generally, the attractiveness of a particular physical form is based on two factors (1) the relative ease of either directly using the material in an IND or converting it to a usable form, and (2) any self-protecting properties of the material, such as high levels of radioactivity, that make the material more difficult to handle or process. Attractiveness levels range from "A" to "E":

Attractiveness Level (AL)	Description
A	Includes nuclear weapons and test devices
B	Includes pure products (metal and directly convertible material)
C	Includes high grade materials that can easily be converted into B materials
D	Includes materials that require greater processing time and complexity to convert to B materials
E	Includes other materials not covered by attractiveness levels A through D, such as highly irradiated materials, low-enriched uranium, and highly dilute materials.

Table 1. MC&A Attractiveness Levels Definitions

Table 1 presents a brief description of nuclear materials Attractiveness Levels "A" through "E" as per DOE M 470.4-6.

In order to dispose of AL "D" material, a series of steps and approvals are required. Figure 2 presents an outline of the steps required in order to obtain approval for the termination of safeguards (TofS) and discard of AL "D" material.

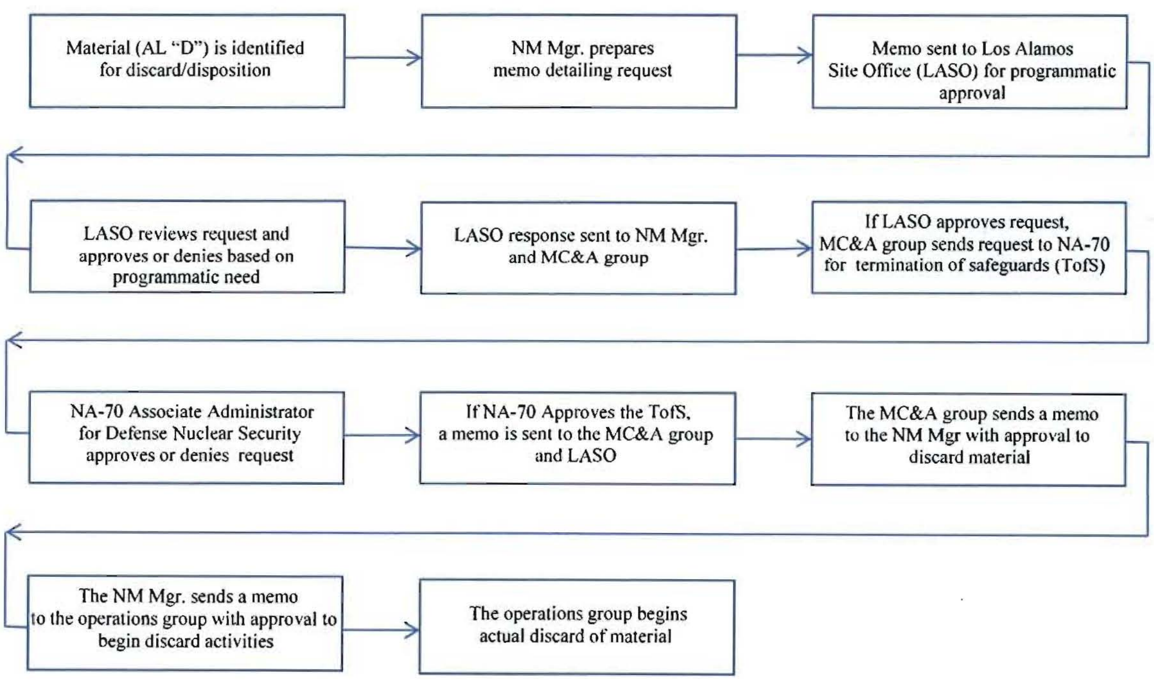

Figure 2. Approval Steps for Termination of Safeguards (TofS) and Discard of AL "D" Material

The most direct and efficient pathway for disposal of this material is through the PF-4 solid waste management operations. These operations are currently functioning at capacity and a backlog of material for discard exists. The cementation option provides an opportunity to address a portion of the backlog of solid waste items in the queue awaiting disposal by developing an alternate pathway for materials that have received approval for termination of safeguards and are destined for discard to WIPP.

CEMENTATION METHODOLOGY

Nitric acid based processing is one of the major process operations used at LANL to prepare material for consolidation and disposition. Nitrate based operations include nitric acid dissolution, leaching, anion exchange, oxalate precipitation, hydroxide precipitation, evaporation and nitric acid recycle. Standard feeds are plutonium-containing materials that do not contain chlorides, such as impure plutonium oxides, non-chloride salts, and sand slag and crucible pieces. Product from the aqueous nitrate process stream is packaged in site-standard storage containers and sent directly to dry operations and subsequently to the vault. The nitrate support operations also include recovery evaluation, cementation, effluent disposition, and WIPP-Waste Acceptance Criteria (WAC) packaging operations.

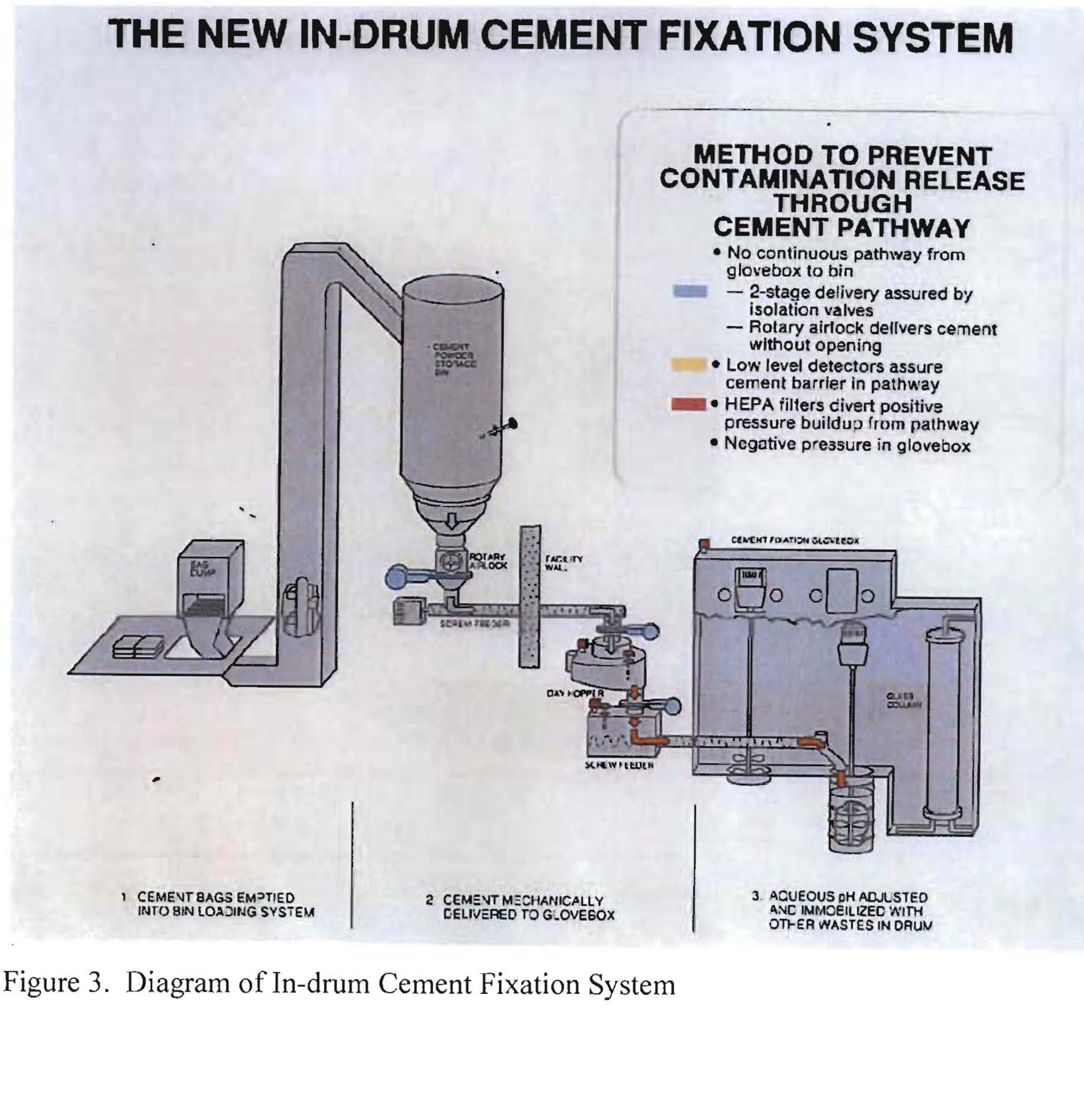

Figure 3. Diagram of In-drum Cement Fixation System

Cementation is a key component of the nitrate flowsheet and serves as a "bleed-off" stream for impurities separated from the plutonium during processing. Figure 3 presents a diagram of the overall cementation infrastructure including the 55 gallon drum that is mated to the floor of the specially designed glovebox. The main "feed" to the cementation operations are the "bottoms" from the evaporation process. Currently, cemented waste drums containing these "bottoms" are prepared at Pu^{239} Fissile Gram Equivalent (FGE) levels that are well below the WIPP-WAC FGE drum limit. An opportunity exists to increase the drum loading of each drum leaving the facility up to the allowable FGE limit by adding intractable residues, not suitable for Pu recovery, to each drum. This would increase the efficiency of drum handling and the disposition of excess residues. This proposal does not increase the number of cement drums but rather increases the drum loading to maximize efficiency. This cementation option is consistent with strategies to accelerate existing pathways for material disposition and opening new pathways for material disposition. The PADS [1] states "...clearly those disposition paths that are currently "open," i.e.,... WIPP, and on-site consolidation, should be vigorously pursued...".

PROCESS DEVELOPMENT METHODOLOGY

The cementation development methodology will identify and define the important parameters and activities leading to a cementation plan by collecting relevant WIPP-WAC, selecting a representative listing of vault holdings of excess material for potential cementation, evaluating candidate material according to chloride content (for salts), identifying acceptable matrices such as free flowing powder, ensuring the homogeneity of the material, and selecting items that are amenable to measurement. If the material meets the physical cement fixation acceptance criteria (CFAC), it is mixed with Portland cement in situ in a 55 gallon drum mated to the floor of the cementation glovebox. (Figure 4).

Figure 4. Picture of in-situ cementation of evaporator bottoms

For example, one of the key required characteristics for material to be acceptable to cementation is that it meets the physical cement fixation acceptance criteria (CFAC) of being a free-flowing, single-matrix residue and/or size-reducible to minus 8 mesh. One of the challenges to this approach is to be able to select items amenable to cementation prior to bringing the item into the glovebox line. Item description codes and associated comments for items in many instances are not sufficiently descriptive to ensure that the material will meet the CFAC. Consequently, a representative list of items that have the potential for meeting the CFAC was generated and data collected regarding the item's ultimate disposition.

EXPERIMENTAL INFORMATION

To date, fourteen items have been evaluated through this selection and evaluation process. Of these fourteen items, nine met the CFAC (Table 2). Based on visual examination, it was determined that process residues from the pyrochemical molten salt extraction (MSE) process (item description code R837) did not contain crucible pieces and the salt monoliths were amenable to crushing to meet the particle size requirement in the CFAC. One of the nine items discarded through Cement Fixation was thought to contain high quantities of chloride that could have interfered with the setting reaction of the Portland cement waste form. However, no negative effects were observed. Consequently, this observation indicates that material with higher concentrations of chloride-based salts can be successfully cemented. In addition, eight residue items from the batch dissolver operations were successfully cemented. Another item required blending and splitting because the FGEs exceeded the CFAC for a single cement drum. This item required two cement drums to discard.

Figure 5 shows representative items that met the CFAC and were subsequently discarded via cementation.

Figure 5. Material Acceptable for Cementation

Cement Drum	Net Wt. (Kg)	Pu (g)	Am (g)	Watt	PE-Ci	NOTES
1	1.81	32	0.32	0.12	4	
2	0.33	118	0.21	0.3	10	
3	0.097	78.5	0.04	0.19	6.3	
4	1.011	91.6	1.7	0.5	16.6	Size reduced for cement
5	0.125	105.9	0.02	0.25	8.35	
6	0.125	105.9	0.02	0.25	8.35	
7	0.062	49.9	0.01	0.12	3.93	
8	0.096	57.7	0.001	0.13	4.54	
9	0.1024	84.6	0.105	0.21	7	
10	0.1108	24.4	0.01	0.06	1.95	

Table 2. Items Discarded In Cement Drums

Five of the fourteen items evaluated through this selection and evaluation process did not meet the CFAC. Three of these items were retrieved from the vault, introduced into the glovebox line, inspected and found to contain mixed matrices (i.e. MgO crucible and salt rocks). It was determined that attempting to segregate each matrix, size reduce and blend to meet the CFAC was not a viable option. Consequently, these items were sent to Solid Waste Management for disposition. The ability to identify vault items that contain multiple matrices before that item is introduced into the glovebox would be very beneficial and significantly reduce handling and exposures. For example, if an item is determined to be non-homogeneous and the SNM content is <150g Pu, it could routed directly to the Solid Waste Operations for discard. Based on data collected so far, we have determined that the item ID may yield a clue. For example, one vault item that met the CFAC and was cementable had an item ID "XLBS-...". Several items that did not meet the CFAC and were not cementable had item IDs of "XBL-...", "XSLT-...". Additional data will be collected in order to better correlate item IDs versus matrix and ability to meet the CFAC. Another finding was that residues from the electrorefining process (item description code R657) contained either mixed matrices (crucible and salt monoliths) or unbreakable large salt monoliths. Consequently, electrorefining salts and residues with item description code R657 will be excluded from further consideration.

Figure 6. Material Unacceptable for Cementation

The items that were evaluated and found to be unacceptable for cementation based on the CFAC, were sent to Solid Waste Management for disposition as opposed to expending time and effort in attempting to segregate each matrix, size reduce and blend to meet the CFAC (Table 3). Figure 6 shows representative items that did not meet the CFAC.

Solid Waste Drum	Net Wt. (Kg)	Pu (g)	Am (g)	Watt	PE-Ci	NOTES
1	2.06	106.6	1.73	0.254	8.53	Crucible & rock
	1.9	83.1	1.44	0.36	11.5	Uncementable
		77.6	1.34	0.34	10.7	
2	3.512	91.6	0.83	0.31	10.1	Crucible & rock
						Uncementable
3	1.79	104.1	0.52	0.3	9.96	Crucible & rock
	1.69	98.52	0.54	0.29	9.59	Uncementable
4	3.0	162				Crucible & rock
						Uncementable
5	4.1	207				Unbreakable rock
						Uncementable

Table 3. Items Discarded In Solid Waste Management Drums

Other residue types remain to be evaluated. For example, zinc-based residues have not yet been cemented so their effect on the setting reaction is still unknown.

CONCLUSION

The cementation process has proved to be an acceptable alternate pathway to WIPP for appropriate materials. Based on data collected so far, several categories of residues have been determined to be acceptable for cementation. In addition, several item description codes have been eliminated from consideration due to the inability to meet the CFAC without considerable effort and expense. Work will continue to identify other acceptable item descriptions and associated data. Based on the information collected as a result of these efforts, a feed list of acceptable items will be developed and will result in a more effective and efficient discard pathway for material currently awaiting disposition in PF-4.

ACKNOWLEDGEMTS

Appreciation is expressed to NA-122 and NA-58 for their support of these activities.

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2. *Nuclear Material Control and Accountability*, DOE M 470.4-6, issued on August 26, 2005

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