A DISCUSSION AND STATUS OF THE STABILIZATION OF MATERIAL AT THE HANFORD PLUTONIUM FINISHING PLANT

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INTRODUCTION

The Plutonium Finishing Plant or the PFP as it is commonly referred to, was constructed from 1947 to early 1949, with hot operations commencing July 5, 1949. During its production and operation from 1949 to the mid-1980’s a significant portion of the Pu produced by the United States was processed at PFP.

After production ceased at PFP an extensive and diverse inventory of Pu-bearing materials remained. The Department of Energy (DOE) and the Defense Nuclear Facility Safety Board (DNFSB) have established a series of milestones to reduce the risk to the worker, the public, and the environment.

The material remaining at PFP must be stabilized and repackaged before deactivation and dismantling the facility can be completed. These Pu-bearing materials can be categorized into broad families. These families or types of materials include metals and alloys; solutions; polycubes; and residues. The process methodology used to stabilize these materials, the milestones and the current status of the categories is discussed below.

METALS

The metals inventory (typically 16 to 18% Pu-240) at PFP consists of approximately 350 items. Stabilization consists of moving the items from the storage vault to an appropriate glovebox (in Building 234-5Z) where the multiple layers of containment, typically foodpack cans are removed. All loose oxide is removed from the metal item by brushing and the brushed metal item is then placed into an inner DOE-STD-3013 container. The inner can is inerted with helium and the can is sealed welded utilizing the bagless transfer technology developed at Savannah River. The Can is then surveyed for contamination and transferred to the leak test station where the container is leak tested to $1 \times 10^{-7}$ cc/sec.

Because the outer can welder is not yet installed; the completed inner 3013 container is returned to the storage vault until the outer can welding system is available. The above metals process flow shown in figure 1.

Due to the age and degradation of the metal items, an operation assumption was made that 10% of the metal items may auto ignite when exposed to air. To-date approximately 20% of the items have auto ignited. Due to conservative criticality constraints imposed on the glovebox, only 1 metal item has been allowed in the glovebox. As a result, when an item auto ignited, further processing in the glovebox
was halted until oxidation of the item was complete, typically on the order of 8 hours. The criticality posting for the glovebox is being revised to allow two items in the glovebox, thus allowing continuing operations and therefore greater efficiency and throughput.

The brushings from the metal items are collected and thermally stabilized at 950 °C in accordance with the 3013 criteria and then packaged in inner 3013 cans.

The DNFSB milestone to complete repackaging of metals is March 31, 2001. PFP is on schedule to have all of the metal items repacked in inner 3013 cans by that date, with the metal brushings to be in inner 3013 cans, no later than April 30, 2001. Because the outer can welder will not be installed at PFP (in Building 2736-ZB) and operational until April 1, 2001, the metal and brushings will not be in 3013 outer cans until August 1, 2001.

**ALLOYS**

The PFP inventory of Pu-bearing alloys consists of approximately 125 items. Originally, 57 of these items were scheduled to be shipped to SRS for processing. Due to a number of difficulties, the decision was made to process all alloys at PFP. An exhaustive study recently completed has resulted in the categorization of the alloys into smaller subsets. The first subset consisting of approximately 41 items, met the 3013 Pu + U assay criteria and will be brushed and/or thermally stabilized and placed into 3013 cans as was done with metals. The second subset of items, consisting of approximately 60 to 70 items, which do not meet the 3013 criteria due to Pu + U content, will be packaged in Pipe Over Pack container or POCs for shipment to WIPP. A small number of remaining items appear to be more characteristic of other Pu-bearing Residues at PFP and are scheduled to be reclassified as Residues and
placed into the Residues family. The process flow diagram for the different alloys groups is shown in Figure 2.

The DNFSB milestone for alloys is to have all items in their final configuration package by June 30, 2001. PFP’s current schedule is to have all items destined for packaging in either 3013 cans or POCs by that date. For those items going into 3013 cans, the June date would include having all items in the outer 3013 cans. PFP is able to meet that date because the outer can welding system will be installed and operational by April 1, with priority be given to canning alloy items rather than metals. This decision will allow the original DNFSB date for alloys to be met and cause only the metals milestone date to have metals in the outer 3013 cans to be late. The majority of the Pu-bearing alloys that will be packaged for WIPP disposal will also be completed by the June 30, 2001 milestone. The smaller subset of alloys that will be placed into the Residues family will be governed by the Residues milestone.

SOLUTIONS

The solutions inventory at PFP consists of approximately 4300 liters of Pu-bearing solutions, typically packaged in 10-liter containers. The concentration of the material varies from 1 to 2 gms/l to 200+ gms/l, depending on the source or family of the material. These families include pure nitrate and impure nitrate solutions, caustic solutions, chloride and chloride-contaminated solutions, and organic solutions. These solutions are left over solutions from weapons and fuels production programs, laboratory experiments to better understand the minimum quantity of material to achieve a nuclear critical mass, and process waste solutions that were too rich in plutonium to discard to the Hanford Waste Tanks. The stabilization process method chosen for the majority of these solutions is a
magnesium hydroxide precipitation process \([\text{Mg(OH)}_2]\) similar to that used at RF. Some solution families such as the organics are not suited for this process. Additional laboratory work is required before a path forward is defined for these solutions. Only the magnesium hydroxide process will be discussed here in greater detail. The process begins with the solutions containers being downloaded into tanks where samples are taken to determine the concentration and molarity of the solution. The pH and concentrations are adjusted (typically diluted) and the adjusted (diluted) solution is transferred to another set of tanks where the \(\text{Mg(OH)}_2\) chemicals are introduced and the material in solution is precipitated out. The current process is limited to concentrations of less than 40 gm/l.

When the precipitation process is complete the material is removed from the filters and placed in boats which are then taken to hot plates for drying (removal of excess moisture is important to proper furnace operations. Excess moisture in the material being stabilized in the furnace has resulted in condensate buildup in the furnace offgas line). The boats are then taken to furnaces where the material is thermally stabilized in accordance with the DOE-STD-3013 criteria. The process flow diagram for solutions processing is shown in figure 3.

Numerous operational difficulties have slowed the processing rates that were assumed in the operational templates and therefore the schedule. The original schedule date, used to establish the completion milestone for solutions was to complete construction and hot startup of the of solutions gloveboxes was July, 2000. Due to construction delays, the actual hot startup date was late September, or three months late. Another significant problem was the operational template assumptions pertaining to the number of boats of precipitate that would be produced. Based on the concentration of Pu in the solutions, the number of boats produced by several families of material would have been 1 boat (approximately 1-½ liters of wet cake) per precipitation batch (approximately 36 liters of diluted
solution). When the family of solution was processed the number of boats produced was up to 7 boats per precipitation batch. Because the critical path in the process flow was the number of furnace cycles or number of furnaces available, the number of boats produced drives the schedule completion date. The cause of the number of boats produced by the solution family has been traced to excessive amounts of tramp material, typically iron, chrome and manganese in the solutions.

A sampling plan was formulated and completed in which representative samples of all the solution families where processed to validate or adjust the number of boats which would be produced. A feed shift to another (pure nitrate) family of solutions has been made and the number of boats produced is consistent with the assumptions and operational templates used to establish the production schedule. While these “well-behaved” families of solution are being processed, various alternatives or enhancements to the current process are being evaluated. Some of these alternatives will include solidification of the lower concentration solutions in drums or other containers, transferring low concentration solutions to Tank farms, and modification of the MgOH process the allow only precipitation of the Pu in solution.

The DNFSB milestone for processing solutions at PFP is December 31, 2001. If no alternatives or process improvements are implemented, the current scheduled date to complete solutions will be March 31, 2002 or three months late.

POLYCUBES

PFP currently stores approximately 260 cans of polycubes (polystyrene cubes containing plutonium and in some cases uranium at approximately 25 wt% Pu or U) that require stabilization. Each can of these 260 cans contains multiple polycubes ranging in size from 2” x 2” x 2” to 1” x 1” x 2”. These cubes were used in laboratory experiments in the 1960’s and early 1970’s to better understand the minimum mass required to achieve nuclear criticality. The majority of the polycubes were stabilized (and the plutonium recovered for other uses) in the 1970’s and 1980’s. Stabilization was performed using a pyrolysis process. Processing was halted in the mid-1980s due to continuing problems with the off-gas system.

The PFP plan originally called for processing polycubes using an improved pyrolysis process where the polycubes are heated in an inert atmosphere to drive off the polystyrene followed by thermal stabilization of the resulting product (plutonium oxide and carbon char) in accordance with the DOE-STD-3013 criteria. After thermal stabilization the material will be placed in 3013 containers. Extensive testing by the PFP Process Support Laboratory and PNNL has disclosed an alternative processing method. This alternative is direct oxidization/thermal stabilization in PFP’s existing muffle furnaces. The key elements to allow safe operation (avoiding flammable concentrations of gas in the furnace) are the control of the furnace charge size (approximately 400 grams of polycubes) and furnace temperature ramp rate. This process change eliminated the need to develop and install a pyrolysis unit at PFP. PFP originally planned to begin processing polycubes in the mid 2001 time period. But because the furnaces that would be used to stabilize the polycubes are now required to process the excessive number of boats, the polycube processing start date maybe delayed until completion of solution stabilization. If
alternatives for solutions cannot be implemented or operational efficiencies identified to reduce the polycube process schedule, the polycube completion date may slip by up to 2 months (completion projected for October 31, 2002). The proposed process flow diagram for polycubes is shown in figure 4.

RESIDUES

The PFP inventory of residues consists of: Hanford and RF ash, sand slag and crucibles, impure Pu oxides and MOX containing < 30 wt% Pu+U, miscellaneous process residues containing < 30 wt% Pu+U, and miscellaneous combustibles and compounds. The original methodology for processing residues at PFP was to use a cementation process and packaging in drums for shipment to WIPP (cement acted as the diluent to meet safeguards termination requirements). Two problems were encountered. First, the Pu loading limit TRUPAC using the standard 55-gal drum presented logistical problems and second, the inability to accurately NDA the cemented residue presented safeguards concerns. Based on the success of POC use at RF, the decision has been made to package the residues in POCs for shipment to WIPP. Currently, PFP is packaging Rocky Flats ash in POCs and is scheduled to complete this family of residues by April 30, 2001. The residues packaging staff which is already trained and qualified to the WIPP criteria will then package the Pu alloys that have been identified to be packaged in POCs, before starting the packaging of Hanford ash.

The DNFSB milestone for residues is April 2004. All indications are that PFP will meet or exceed the DNFSB milestone date.
SUMMARY

This paper identifies the various material families, described the stabilization process and a status of the stabilization schedule at PFP. PFP is well on its way in stabilizing it’s extensive and diverse inventory of Pu-bearing materials. Once the material is safely stabilized and packaged the task of full scale deactivation and dismantlement of the PFP complex can begin. To support material stabilization at PFP, a series of operational templates were created. These templates have allowed the facility to develop production curves and schedules and monitor progress against the milestones. In addition, they allow critical parameters including facility staff, facility and equipment capability and availability, and operator doses to be identified and assessed. Because they integrate key parameters, they can be used in the generation of both annual and lifecycle budgets, including various budget scenarios and the resultant effect. “What if” exercises can be run and their impacts on the various parameters assessed. Work-arounds for various process upsets, equipment failures, operators burnout or other unplanned events can be quickly developed and effects understood. Using these templates, the facility has determined that the existing or to be installed plant equipment will support the milestones established. In addition, the staff size is adequate although some skills mix problems may arise at a later date. The major issues that must be addressed and resolved are radiation exposures and plant/equipment availability. Whenever the schedule is accelerated while the amount of material to be processed and the staff size is held constant, the radiation exposures to workers will increase. Many options are being evaluated and implemented including improved shielding, staff rotational assignments, and process optimization to reduce exposure times.

The facility can and will find ways to meet or exceed not only the DNFSB milestones but also the deactivation and dismantlement dates.
REFERENCES

1. Integrated Project Management Plan for the Plutonium Finishing Plant Stabilization and Deactivation Project, HNF-3617 Rev 0, April 1999
2. Update and Status Against the Integrated Project Management Plan, May, 2000