Planning for the Decontamination of the Plutonium Fuel Form (PuFF) Facility Savannah River Site (SRS), South Carolina - 10095

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ABSTRACT

The Department of Energy (DOE) Environmental Management Office of Decontamination and Decommissioning (D&D) and Facility Engineering is actively pursuing risk reduction across the DOE complex by identifying high hazard facilities and prioritizing planning funds to ensure safe and efficient execution of the decontamination and decommission work. The Plutonium Fuel Form (PuFF) facility in Building 235-F at the Savannah River Site (SRS) is estimated to contain up to one kilogram of plutonium oxide (presently 67% Pu-238) as micron and submicron particles, which makes it one of DOEs most hazardous radiological structures to decontaminate, dismantle, package and remove.

Safe and effective decontamination of this facility presents many technical challenges that must be understood by personnel planning the work evolution to mitigate the imminent health hazards associated with inhalation or uptake into the body (via puncture wounds) of plutonium oxide particles. Personnel who plan and perform the work must be cognizant of the properties and behavior of Pu-238 in the PuFF facility. This knowledge is necessary prior to tackling the challenges associated with the selection and testing of decontamination reagents, remote tools, and non-destructive assay instruments.

Decontamination reagents must be effective, easy to apply and remove with remote equipment, and pose no problem for waste acceptance criteria of the selected disposal facility. The type and number of remote tools must meet the needs to map out the debris and equipment within the hot cells, remove contamination with vacuum systems, apply and remove decontamination reagents, and size-reduce and package equipment. Accurate design of such tools requires detailed 3-Dimensional models of the facilities to allow personnel to measure and visualize access ports, radius of reach for tools, pinch points, and material volumes of waste streams. Non-destructive assay methods are required to minimize worker contact and exposure to the waste streams, and the methods must be accurate to ensure waste packages will meet shipping and disposal requirements.

INTRODUCTION

The Plutonium Fuel Form (PuFF) facility, located in Building 235-F at the Savannah River Site (SRS), was designed to manufacture fuel forms for radioisotope thermoelectric generators used in National Aeronautics and Space Administration (NASA) space vehicles. Fuel forms were manufactured in a series of hot cells and glove boxes using plutonium oxide with an initial loading of 84.5% Pu-238 (present oxide weight percents are estimated at 66.7% Pu-238, 12.3%

Pu-239, 2.6% Pu-240, 0.1% Pu-241, 0.1% Pu-242, 0.4% Am-241 and 17.8% U-234) [1]. The Pu-238 is an ideal fuel isotope because of its high decay heat (0.57 watts/g). As part of the manufacturing process, the plutonium oxide feed material was milled to a fine powder with an average diameter of approximately one micron (about the size of a typical bacterium). Due to the small particle size, high specific activity (17.3 Ci/g) and alpha decay energy (5.5MeV) of Pu-238, this material is highly mobile and has been responsible for a number of radiological incidents at SRS and elsewhere in the Department of Energy (DOE) complex. At the SRS, the nature and extent of the contamination in the PuFF facility make it one of DOE's most hazardous radiological facilities. Inhalation of a single 8 μm diameter spherical agglomeration of plutonium oxide particles (density 50% of plutonium oxide) containing 66.7% Pu-238 oxide could result in a worker exceeding the annual allowable dose of 50 rem to the bone surface [2].

In the early 1990s, the PuFF facility was shut down and de-inventoried of free product. However, non-destructive assay (NDA) of the hot cells and glove boxes revealed several hundred grams of Pu-238 in the process equipment. Since the PuFF facility and building 235-F have no current or future mission, DOE is planning a $20,000,000 project to decontaminate, dismantle, package and remove the five most contaminated PuFF hot cells, which will remove 90% of the existing Pu-238. As hundreds of SRS workers are located within 100 meters of building 235-F, safety and As Low As Reasonably Achievable (ALARA) radiological exposures are major drivers for the decontamination and dismantlement of the PuFF facility. Project success will require state-of-the-art technologies, safe execution of work scope, and vigilant monitoring and management.

The DOE Environmental Management Office of (Decontamination and Decommissioning) D&D and Facility Engineering provided nearly $1,000,000 in research funds to the Savannah River National Laboratory (SRNL) to investigate and document the physical, chemical and radiological properties of Pu-238, with emphasis on performing safe and effective decontamination of the PuFF facility, and technologies that are suitable to support the decontamination effort. Research status will be summarized for the following topics:

- Properties and Behavior of Pu-238 Relevant to Decontamination of Building 235-F
- Decontamination Materials
- Remote Tool Systems for Equipment Decontamination and Size Reduction
- Non-Destructive Assay Methods for Waste Streams

TECHNOLOGY SUMMARY

Properties and Behavior of Pu-238 Relevant to the Decontamination of 235-F

Plutonium-238 is a man-made nuclide that is created by irradiating Np-237 targets to produce Np-238, which decays by beta emission to produce Pu-238. The Pu-238 isotope has relatively high values for specific activity (17.3 Ci/g), decay heat (0.57 W/g) and energy (5.5 MeV). It decays (half-life of 87.7 yrs) to U-234 by emission of an alpha particle, and the recoil energy can be an important mechanism for mobilizing nanometer (nm) size fragments of plutonium oxide.

The plutonium-oxide feed powder was ball milled to reduce the size of the particles to a target diameter of 0.6 μm (600 nm), which created a size distribution of less than 1μm for most particles at the time of fabrication. As the facility was shut down and de-energized in the 1990s, water adsorption (ambient relative humidity is near 60%), agglomeration by Brownian collisions, and gravitational settling are likely mechanisms for the removal of the initial plutonium particles that were suspended by ball milling. However, the agglomerated particles can become re-suspended temporarily when physically disturbed and recoil during alpha decay will fragment some particles and temporarily suspend particles less than 20 nm. Alpha recoil is also an important mechanism for transporting plutonium nanoparticles through HEPA filters [3].

Based on these observations and additional information provided in [1], the majority of the settled particles must be removed with an effective vacuum tool and/or decontamination reagent prior to dismantling the hot cells and glove boxes, and HEPA filters must be monitored closely to prevent plutonium breakthrough. The following sections will discuss the appropriate decontamination materials, remote tools needed to vacuum particles and to apply and remove decontamination reagent, and plutonium assay instruments for HEPA filters and other waste streams.

Decontamination Materials

Traditional decontamination actions on glove boxes and hot cells include wiping down surfaces with rags soaked in dilute nitric acid or applying proprietary liquids to the surface, scrubbing and wiping the surfaces with rags [4]. In general, several applications are required to achieve release criteria for highly contaminated surfaces, and a substantial volume of waste rags can be generated because rags are used only for a single swipe to eliminate recontamination of the surface. There is interest in the DOE complex to perform such activities with reagents that have greater removal efficiency, while using less labor time and generating less waste volume.

Cellular Bioengineering Inc (CBI) produces a decontamination gel that is spread or sprayed on the surface and allowed to cure before it is removed as a strippable film. This gel is water based and contains 3 to 7% ethanol (per the material safety data sheet), and it was used to decontaminate a plutonium contaminated glove box at Lawrence Livermore National Laboratory [5]. Two applications to the Lexan window and three applications to the steel floor were needed to remove over 99 percent of the measured activity. The gel also formed an impermeable film that blocked over 90 percent of the measured radiation, which provided extra protection to the worker by eliminating re-suspension and reducing extremity exposure. However, labor hours, waste type (e.g., transuranic (TRU) or low level), waste volume and disposition were not reported, and the primary plutonium isotope removed was Pu-239, which has a much lower specific activity (0.063 Ci/g) than Pu-238.

Although the application of the CBI decontamination gel immobilized the plutonium and removed greater than 99% of the measured radioactivity, the classification of the waste as transuranic (>100 nCi/g) would require that the waste stream meet the Waste Isolation Pilot Plant (WIPP) waste acceptance criteria (WAC) prior to disposal at WIPP. Because of the ethanol content of the gel, and the expectation that the use of the gel in the PuFF facility will result in the generation of a transuranic waste stream, volatile organic compounds (VOC) may be generated in the waste containers at levels that exceed the WIPP WAC. Therefore, SRNL is performing tests to measure the VOC concentration in containers filled with uncontaminated gel film. There is also a need to test the product on a small area contaminated with Pu-238, and this must occur prior to its full-scale use in the PuFF facility.

Remote Tool Systems for Equipment Decontamination and Size Reduction

The properties and behavior of Pu-238 create a very hazardous working condition that must be mitigated with proper engineered barriers and administrative controls. Glove boxes and hot cells must remain isolated and sealed when workers perform the decontamination activities, and the use of remote tool systems will allow for risk management at an acceptable level. Lights, cameras, small manipulators, and extended vacuum tools are needed to perform visual surveys, characterization tasks, and decontamination of the equipment. The level of success for decontamination efforts will influence the need for additional remote tools to size reduce and package the debris and legacy equipment inside the hot cells.

Dimensional information for the glove boxes, hot cells and adjacent rooms is needed to plan the work evolution and design the proper tools. Available 2-D drawings of the PuFF facility are difficult to manipulate in a manner that presents a clear visual picture of the facility geometry. SRNL has eliminated this visualization problem by developing a virtual 3-D model of the PuFF facility using Pro/Engineer (version Wildfire 4). The 3-D model contains detailed dimensional information necessary for planning all aspects of decontamination of the PuFF facility. Virtual 3-D images can be exported to the Stratasys FDM rapid prototyping machine to construct accurate scale models that provide workers with a clear picture of the challenges associated with tool design and work execution. The virtual and scale models will play an important role in the success of the project.

Mounting brackets and manipulator arms for lights and cameras are the first mechanical tools that must be constructed for the project, as the PuFF facility was shut down, de-inventoried, and de-energized in the 1990s and the amount of debris and equipment within the hot cells is not well documented. Mounts for the lights and cameras have been designed and fabrication will be initiated in the second half of FY10. Lights and cameras must be introduced into the hot cells via the glove ports in the cells to obtain a visual archive of the materials and condition of the cells. This information is required to plan decontamination activities and prepare work plans.

The first decontamination activity will consist of the removal of loose contamination with vacuum tools. Criteria relevant to these tools includes the capability of the equipment to pass through openings as small as 25 cm, the need for the equipment to be fitted with ultra low penetration air (ULPA) filters, and the remote operation of the system. ULPA filters are needed because there is a size fraction in the PuFF hot cells below 0.3 μm from the ball milling process and the nanoparticles produced by alpha recoil (HEPA filters are only 99.77% efficient for particle sizes greater than 0.3 μm). SRNL has identified the Tiger-Vac MV-1CR HH and the Nilfisk GM80 vacuum systems, with ULPA filters, as the initial models to test [6]. Initial testing is required to determine the limitations and capabilities of these systems prior to investing resources on the design and manufacturing of the remote tools that will be needed to operate the vacuums.

After completing the vacuum decontamination task, remaining surface particles will be locked down and removed with decontamination reagents using workers in personal protective equipment (PPE) and/or remote tools. The actual activity path for this work scope will be dictated by health and safety engineers and described in the final safety basis document. For example, this work evolution may involve remotely spraying the entire inside surfaces with decontamination gel to immobilize any remaining particles, followed by workers in PPE entering the hot cell to remove the cured gel. Alternately, health and safety requirements may state that workers are not permitted in the cells and remote tools will be needed to apply and remove the decontamination reagents.

The goal of the decontamination work will be to lower plutonium contamination to levels below 100 nCi/g. If achieved, this will minimize the size-reduction work and the need for a diverse array of tools because equipment disposed of as low-level waste does not have to meet the restrictive size-reduction requirements for transuranic waste placed in TRUPACs. This is a key lesson-learned from size-reduction operations at Rocky Flats, which were executed with mechanical and plasma-arc cutting tools. SRNL is evaluating the cutting methods and tent enclosures used at Rocky Flats to determine if the methods and technologies can be modified for use at the PuFF facility.

Non-Destructive Assay (NDA) Methods for Waste Streams

Accurate NDA methods are needed to quantify the plutonium isotopic composition of the waste types removed from the PuFF. The density of the waste forms can vary from steel to decontamination gel (polymer hydrogel) and the geometry may consist of odd shapes packaged in one or five-gallon pails, 55-gallon drums or standard boxes. Analytical results for the waste packages must have sufficient quality control to certify the waste stream to the WAC of WIPP and the SRS solid waste management facility (SWMF). Highly conservative estimates of plutonium levels in waste containers are undesirable because they can significantly increase project cost due to less than optimum container loading, particularly for TRUPACs destined for WIPP, or improperly identify the waste as low level when it is transuranic (>100 nCi/g). Therefore, state-of-the art neutron and γ-ray counting systems are needed to obtain accurate quantitative plutonium assays of the waste streams for shipment and waste certification requirements.

SRNL reviewed the limitations of current NDA methodologies for waste forms with a plutonium isotopic distribution of approximately 80% Pu-238 (U-234 in-growth is about 25 atom %), 15% Pu-239, and 4% Pu-240, and identified the optimal γ-ray and neutron counting systems for the characterization of the plutonium in the PuFF waste streams. The required specifications have been established [7, 8] and a contract will be placed with a vendor to provide suitable instruments and to perform the measurements. Prior to award of the contract, the successful vendor must demonstrate that the instruments can meet the performance and operational criteria for measuring gamma photons from low-density waste in one and five-gallon pails (e.g., the instrument shall have the capability to assay uniform density, uniform geometry items with total Pu-238 content between 0.5 and 6 g with equal to or better than 10% uncertainty) and neutron fluxes from high-density waste in a 55-gallon drum and a standard waste box (183cm x 94cm x

137cm) (e.g., in either closed cylinder or expanded counting configurations, the instrument shall have the capability to assay variable density, non-uniform, unspecified volume items with total Pu-238 content between 0.1 and 0.5 g with equal to or better than 100% uncertainty with acquisition times of less than 60 minutes).

The heat generation of Pu-238 particles creates the potential for semi-quantitative assay of the isotope using high resolution thermal sensors. This method could be applied in the cells to identify locations of Pu-238 accumulation to aid in the decontamination effort. Additionally, there is the potential for this technology to be used in re-packaging individual waste items when a shipping container is assayed and shown to contain too much Pu-238 inventory. Individual waste items could be scanned by the thermal sensors to map out the Pu-238 distribution and the items could be removed or size-reduced to meet the limit. However, an initial assessment of the potential for using infra-red technology to assay cell and glove box surfaces for Pu-238 contamination concluded that this was not a promising application because the Pu-238 particles in the PuFF facility are too small to create a significant thermal signal.

CONCLUSIONS

The PuFF facility is one of DOEs most hazardous radiological facilities, and decontamination must be carefully planned and safely executed to manage the risk at acceptable levels. Early planning is in progress and three technical focus areas have been identified: decontamination materials, remote tool systems for equipment decontamination and size reduction, and non-destructive assay methods for waste streams. Technical challenges must be tested and proved before work commences in FY12.

A preliminary review of decontamination materials used across the DOE complex indicates that the CBI decontamination gel shows the most promise for ease of use and waste minimization. Testing of uncontaminated cured gel strips is underway to determine if VOC head space concentrations in sealed containers will meet the WIPP WAC. Additional small-scale tests will be required to determine the performance of the gel in removing Pu-238 contamination from glove box materials.

The first deployment of remote tools will consist of lights and cameras to acquire a photo archive of the materials and debris within the glove boxes and hot cells. Mounts for lights and cameras have been designed and fabrication will begin in the second half of FY10. Manipulator arms must be designed and fabricated for the lights and cameras after these components are tested and final models are selected, and similar tools will be needed for the activities associated with the vacuum systems and decontamination reagents.

Non-destructive assay systems must deliver accurate measurements with acceptable error to allow certification of the waste packages to the WAC of WIPP and SRS SWMF. Specifications have been prepared for the optimal γ-ray and neutron counting systems needed to characterize the plutonium in the PuFF waste streams. Prior to procurement of vendor services to provide instruments and measurements, the vendor must successfully demonstrate that the instruments are capable of performing the measurements to the criteria established in the specifications.

Additional planning is needed to select and generate suitable waste streams for the performance testing.

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