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Plutonium Oxide Process Capability Work Plan 13.006C

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1.0 Introduction

Pacific Northwest National Laboratory (PNNL) has been tasked to develop a Pilot-scale Plutonium-oxide Processing Unit (P3U) providing a flexible capability to produce 200g (Pu basis) samples of plutonium oxide using different chemical processes for use in identifying and validating nuclear forensics signatures associated with plutonium production. Materials produced can also be used as exercise and reference materials. PNNL is well positioned to establish this capability within the Radiochemical Processing Laboratory (RPL) using existing facilities that can be rapidly configured to support this activity. The existing RPL Documented Safety Analysis (DSA) covers the proposed work, and the RPL has a full suite of analytical capabilities for analyzing the product and waste streams under approved NQA1 protocols. PNNL also has additional state-of-the-art analytical facilities for forensic analysis that can support this capability. Similar operations were performed in the RPL facility during the Nuclear Waste Vitrification Project in the 1970s, including ion exchange, conversion of plutonium nitrate to oxide, and packaging and shipping of plutonium oxide samples.

Technical staff at PNNL are renowned leaders in the areas of spent nuclear fuel reprocessing and signature discovery. Staff currently working at PNNL provided process expertise to the Plutonium Finishing Plant (PFP) at the Hanford Site during the Hanford production runs which included conversion of plutonium nitrate to plutonium oxide. Other PNNL staff were involved in the characterization of plutonium, uranium, and mixed plutonium and uranium samples for fabrication of MOX fuel at Hanford. PNNL provided technical leadership to the Pit Disassembly and Conversion Facility for all of the plutonium processing design including flow-sheet development, criticality safety, industrial safety, safeguards and security, analytical laboratory support, process equipment design, and glovebox design was provided by staff at PNNL. Samples from the ARIES process were received and characterized at the RPL. This testing included dissolution studies with nitric acid and hydrofluoric acid and characterization of samples for critical elemental and radionuclide compositions. These experienced staff have been involved in mentoring and training junior scientists at PNNL to carry on this heritage of excellent leading edge research in spent nuclear fuel reprocessing, signature science, and forensic analysis. Management at PNNL is very supportive of these experimental capabilities as signified by recent major laboratory level initiatives on signature science and nuclear archeology.

The RPL is a Hazard Category II Non-Reactor Nuclear Facility. The work proposed by this project is well within the nuclear safety basis of the facility and allows for sufficient material inventories. In addition, RPL now operates 16 hot cells, 25 gloveboxes, 160 fume hoods and 87 radiological laboratories including limited area laboratories.

PNNL has comprehensive analytical capabilities that can be brought to bear on the plutonium oxide production capability, including both inorganic and radiochemical analyses. Characterization analyses will be performed on the feed and process materials in order to obtain trace elemental and morphological information. State of the art elemental and isotopic analytical support is available on site, as are advanced microscopy (Optical, Scanning Electron, and Transmission Electron), spectroscopy (e.g., micro UV-Vis, Raman, and Atomic Fluorescence, as well as solid UV-Vis, and FTIR) and (X-ray) diffraction instruments needed to fully characterize the PuO_2 product, process and feed materials. PNNL also offers flexibility to other client-driven analytical capabilities beyond those listed here.

There are two major programs (PNNL's safeguards and criticality safety) that constrain the amount of plutonium oxide that can be received and handled for this project. PNNL's safeguards program restricts the total inventory of material at PNNL (campus wide), while the criticality program establishes limits within a given control area, (generally defined as a single laboratory space). Current safeguards rules allow for a total inventory at PNNL of 1800g of PuO_2; however, PNNL is pursuing an engineered solution that would raise the total inventory limit. Currently at PNNL, there are three criticality safety specifications (CSS) that can be used when working with fissionable materials. Each CSS provides administrative and engineered controls that define the boundaries and parameters for working with fissionable material. There are three CSS in use at RPL.

CSS-1 provides administrative and engineered controls that allows for the handling of up to 170g of plutonium. This CSS has stricter guidelines with respect to moderating and special reflecting materials.

CSS-4 provides administrative and engineered controls that allows for the handling of up to 45g of plutonium with no limit to the use of moderating materials or special reflectors.

. CSS-5 provides administrative and engineered controls that allows for the handling of a plutonium metal sealed source up to 199g of plutonium. This CSS has stricter guidelines with respect to moderating and special reflecting materials.

We intend to develop a new CSS for the work conducted under this project that will allow handling 200 g Pu (227 g plutonium oxide) as desired by the client. Until that new CSS is developed, the operating envelope of this project will assumed to be that of CSS-1.

1.1 Milestones and Status

Four milestones are associated with FY13:

1. 13.006A Preliminary Safety Evaluation (submitted October 11, 2013)
2. 13.006B Draft Work Plan (submitted December 13, 2013)
3. 13.006C Final Work Plan (due February 28, 2014)
4. 13.006D FY13 Annual Report (due March 31, 2014)

This document fulfills the 13.006C deliverable requirement.

2.0 Work Plan

This work plan was developed by the PNNL team to provide a generalized overview of the P3U capability. The work flow diagram, depicted in Figure 1, identifies the associations and prerequisite pathways required at critical junctions regarding this effort.

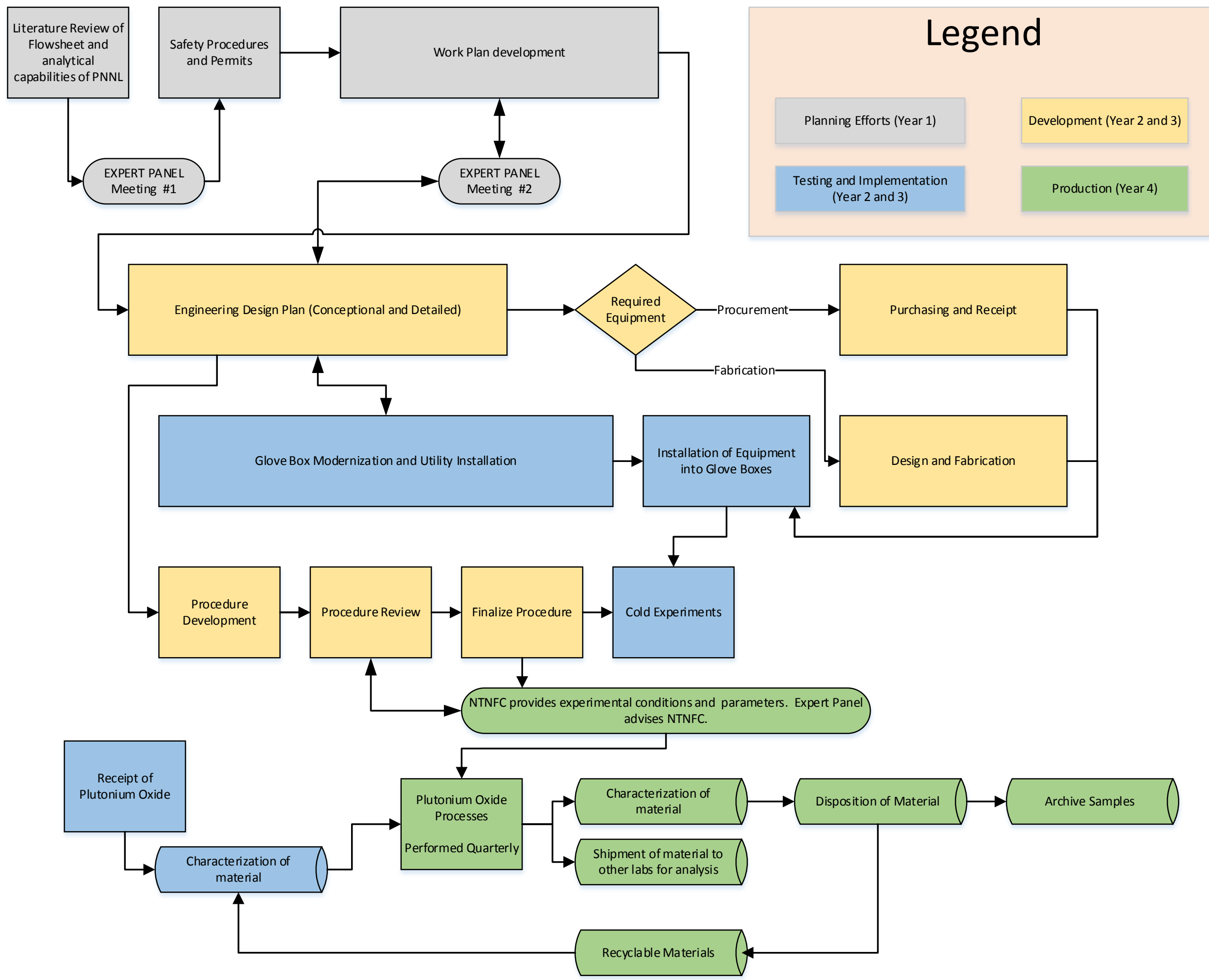

Figure 1

Work flow diagram of Plutonium Oxide Processing Project.

2.1 Literature Review of Flow Sheets and Analytical Capabilities of PNNL

During the first Expert Panel meeting, criteria were established that would identify specific flow sheets for this effort. An important consideration was selecting flow sheets that have a high probability of success and whose methods can be easily obtained in the literature. This requires that the flow sheet be technically feasible and that there are sufficient records and/or published literature describing the historical products so that it is possible to determine if the modern products replicate the older products with regards to specific characteristics. The quantity of plutonium produced in the US by any flow sheet is also important, as many processes were used for R&D purposes or for a short period, but they are not considered important flow sheets with respect to the US stockpile.

Various criteria were presented at the first panel meeting and the Expert Panel decided that the order in which the flow sheets should be accomplished is:

1. Plutonium(III) oxalate precipitation
2. Plutonium(IV) oxalate precipitation
3. Plutonium(IV) peroxide precipitation
4. Plutonium metal oxidation

PNNL has identified suitable lab space to conduct the work and has begun procedural, safety, safeguards and criticality reviews that need to be in place to perform the work.

2.2 Safety Procedures and Permits

<u>Note: The PNNL team previously submitted a safety report authored by Dr. Joel Tingey. The intention of this work plan is not to reproduce the safety report in its entirety; however, certain radiological hazards will be highlighted and discussed.</u>

Pedigreed plutonium oxide will be prepared by dissolving plutonium oxide, concentrating and purifying the plutonium nitrate, precipitating the plutonium, and calcining the plutonium to make plutonium oxide. This effort will be completed in gloveboxes in the RPL.

Several hazards have been identified as part of the plutonium oxide production process. The hazards discussed are criticality, radiation dose, and radioactive material contamination. Each of these hazards has been evaluated and mitigating strategies have been implemented.

Criticality Safety

A criticality event is credible with the amount of fissionable material in the facility; therefore, criticality safety controls will be implemented using the double contingency principle to prevent a criticality accident.

The nuclear criticality safety program at PNNL evaluates the handling of all fissionable material and ensures that staff handling fissionable material or supervising handlers of fissionable material are qualified. The nuclear criticality safety program also ensures that all processes for fissionable material meet the double contingency principle. A criticality safety alarm system is maintained to minimize the impact of a criticality event in the RPL.

A criticality safety specification will be drafted by a certified criticality safety engineer that will provide criticality safety limits and controls for the activities associated with this project. Each scientist and technician handling fissionable material will be trained to this criticality safety specification. This specification will be reviewed by the senior criticality safety engineer at PNNL, line management, and the criticality safety program leadership.

Radiation Dose

The radiological protection program at PNNL protects personnel from sources of ionizing radiation in accordance with 10 CFR 835. This program establishes controls for the acquisition and receipt of radioactive material, identifies hazards associated with work performed with radiological materials, controls work in gloveboxes, identifies controls for packaging and storage of radioactive material, and monitors radiological material and containers.

Radiation Work Permits (RWP) will be generated by the Radiation Protection Organization at PNNL to limit dose to individual workers and the project team to As Low As Reasonably Achievable (ALARA). An ALARA review including experts in the process and Radiation Protection Engineers will be completed to ensure that the work is performed safely prior to operation with radioactive material. All work on the project with radioactive material will be performed according to project specific RWPs. Discussions have been held with the Radiation Protection Engineers for the RPL, and they do not anticipate any issues with this project work.

Radioactive materials inventories in the RPL are tracked and evaluated and a surveillance program evaluates facility radioactive material holdup inventories and changes to those inventories to verify dose consequences from potential accidents remain bounded by the accident analysis in the RPL DSA.

The majority of the penetrating dose from materials handled in the project will come from the in-grown ^{241}Am in the feed material. Americium will be removed during the ion exchange process prior to conversion of the plutonium nitrate to plutonium oxide. The accumulated americium will be shielded and ultimately disposed. Plutonium isotopes radioactively decay emitting primarily α particles; therefore, minimal shielding would be required after the ion exchange purification.

Radiological Contamination

Plutonium oxide is considered a highly-dispersible radioactive material; therefore, containment must be implemented during handling of this material to avoid contamination of personnel and facilities. All process work of large batches of plutonium-bearing material will be performed in gloveboxes to minimize the risk for contamination incidents. The ventilation system for the gloveboxes in Room 604 of the RPL will be updated to meet current requirements. These gloveboxes once updated will provide the controls necessary to mitigate contamination hazards. The permanent structural confinement boundaries (floors, walls, roof, and windows) of the gloveboxes are safety-significant components and alterations to these boundaries are reviewed to verify that functional confinement capability is maintained. Glovebox confinement performance is periodically monitored under the radiation protection program.

The Radiation Work Permit (RWP) described in the previous section will include requirements that minimize the potential for contamination of individual workers and facilities.

2.3 Work Plan Development

This document will serve as a general plan for the P3U system for the plutonium oxide processing project. The client and the Expert Panel will review this document and provide PNNL with comments and recommended changes. The evaluation of this document will likely be addressed during the second Expert Panel meeting.

2.4 Engineering Design Plan

Design of the Pu oxide production capability will be done in two stages—conceptual and detailed. The conceptual design will describe the basic process steps required, such as dissolution of the feed PuO_2, Pu purification by ion exchange, precipitation, and calcination. The basic functional requirements for the individual unit operations will be defined in the conceptual design phase, and a general description of the required equipment will be given. The specific details of the equipment to be procured and installed will be developed in the detailed design stage of the project. The scientists and engineers engaged in the project will work closely with the PNNL Facilities and Operations (F&O) organization to develop and implement the detailed design.

2.4.1 Purchasing and Receipt of Needed COTS Equipment

Commercial off-the-shelf (COTS) equipment will be procured through established PNNL purchasing procedures as described in *Acquire Products or Services* workflow.

2.4.2 Design and Fabrication of Non-COTS Equipment

Custom equipment that cannot be obtained as COTS will be designed and fabricated in-house at PNNL. Design and fabrication services will be secured as needed from the F&O organization using the PNNL service request tool. The cognizant scientists or engineers will provide detailed specifications and criteria to the crafts personnel to verify that the items produced meet the needs of the project. Any non-COTS equipment installed in the gloveboxes shall go through formal commissioning as described in RPL-ADMIN-004, *Installation and Commissioning of R&D Instrumentation/Equipment and Systems*.

2.5 Glovebox Modernization and Utility Upgrade

The P3U system will occupy three gloveboxes in the RPL lab facility. Modernization and utility upgrades, to be performed by PNNL F&O organization, must be completed by March 2015 so that the processing system can be installed and shakedown experiments can be conducted prior to performing the 200g Pu experiments scheduled to start in April 2016.

2.6 Procedure Development, Review, and Finalization

A procedure will be developed and will encompass all potential methods associated with the processing of plutonium oxide including dissolution, purification, precipitation and calcination of the final product. The flexibility of the P3U system will require the procedure be written as generally as possible allowing for the ability to modify specific parameters of the system (i.e., temperature, acid concentration, resin type, etc.). Once the overarching procedure has been developed and approved, then specific experimental parameters will documented in the form of a test instruction for each experiment. The Expert Panel will provide feedback ensuring that all needed parameters and system requirements are met.

2.7 Installation of P3U System into Gloveboxes

Upon completion of the glovebox modernization and utility installation efforts, all of the procured and fabricated systems will be installed. Installation of COTS equipment in the gloveboxes will be reviewed and approved by Shielded Facilities Operation (SFO) personnel. All equipment installed in the gloveboxes shall go through formal commissioning as described in RPL-ADMIN-004, Installation and Commissioning of

R&D Instrumentation/Equipment and Systems. Shakedown experiments using surrogates will be conducted to verify that all aspects of the P3U system are completely operational prior to the plutonium processing experiments scheduled to begin in April, 2016.

2.8 Receipt and Characterization of Plutonium Source Material

PNNL can receive 1 kg aggregate of plutonium bearing source material packed in individual sample containers not exceeding 100g of plutonium. PNNL staff will work with the provider of the material to ensure that it is packaged in such a way that PNNL can receive, handle, distribute and store the plutonium compounds. As described above, current safeguards and criticality rules limit PNNL to a total inventory of 1800g of PuO_2 and an individual batch size of material to 170g. The latter limit should be increased when a new CSS is established for this project.

2.9 Plutonium Processes and Characterization

The Expert Panel will provide recommendations to the NTNFC Program Manager who will provide specific experimental parameters to the PNNL. PNNL staff will incorporate the modified parameters into the test instruction providing the specific experimental parameters and will conduct the processing experiments according to this test instruction and approved procedure in the P3U system. Prior to conducting the first experiment a project specific CSS will be written that provides administrative and engineered controls that allow the production of 227g of plutonium oxide (200g Pu). Each quarterly process will produce approximately 227g of plutonium oxide and samples of this product and process intermediates will be analyzed at certain national laboratories (TBD) for material characteristics which may be used to determine nuclear forensics signatures associated with the chemical process. A plan will be developed for disposition of the material and sample quantities required for the desired analysis, archival and recycling efforts.

3.0 PNNL Material Limits

LIMIT	Current	Proposed increase to be established by Year 3
Safeguards	1800g of plutonium oxide	PNNL is proposing implementing vaults that can exempt certain inventories from its current overall inventory The total inventory limit is TBD.
Criticality safety	CSS-1: 170g of plutonium CSS-5: 199g of plutonium in a sealed source configuration	Project specific CSS: 250 g of plutonium
Safety basis	7kg of plutonium	No need to change

4.0 Integrated Logistics Support Plan

4.1 ILSP

The Integrated Logistics Support Plan (ILSP) denotes specific resources that mitigate risk to the project. These resources include aspects such as training, facility support, and a logistics plan detailing the disposition of equipment and materials at the end of the effort.

4.1.1 Training

PNNL project staff who work in the glove box are required to complete the following courses prior to handling fissionable materials in a glove box.

- Course 821/822 Radiological Worker II Training
- Course 813 Initial/Escorted Worker Glovebox Training
- Course 814 General Glovebox Operations
- Course 649, Nuclear Criticality Safety Theory Self-Study
- Course 648, Criticality Safety program Training for Fissionable Material Handlers - Classroom
- Course 2301, RPL Criticality Safety Administrative Controls with Examination
- Course 1775, Fissionable Material Handler Qualification Card
- Course 972, Required Reading for Procedure RPL-SA-001, Radioactive Material Tracking Instruction
- Course 1334, Required Reading for Procedure RPL-ADMIN-002, Radioactive Material Tracking
- Course 1421, Required Reading for Procedure RPL-OP-001, Routine Research Operations
- Course 1946, Radiological Material Tracking (RMT) Briefing

4.1.2 Facility Support

Facility staff in RPL routinely conducts maintenance within the facility. Activities that service the various systems include, but are not limited to, fire suppression, ventilation, criticality control, and security. RPL F&O staff will also maintain the glovebox where the work is to be conducted.

4.1.3 Disposition of Material and Equipment

Waste materials will be removed from the gloveboxes and disposed of, as needed, throughout the course of the project as to maintain good housekeeping and a safe operating environment. Waste disposal will follow existing PNNL policies and procedures. PNNL has documented procedures and protocols that are to be followed when a project vacates a laboratory. This procedure, in summation, will ensure that the laboratory space is free of any radioactive material, equipment and chemicals that were solely owned by the project, and PNNL requires a walkthrough upon the completion of this process. Upon vacating the laboratory space, all equipment will be disposed of as transuranic waste (TRU) and current TRU disposal rates (FY 2014) are approximately \$500/kg or \$15K/m^3. The project assumes all costs associated with this close out procedure.

5.0 References

RPL Safety Basis Documents

Pacific Northwest National Laboratory; *325 Building, Radiochemical Processing Laboratory, Documented Safety Analysis*; PNNL-DSA-325, Revision 5; December 2012.

Pacific Northwest National Laboratory; *325 Building, Radiochemical Processing Laboratory, Technical Safety Requirements*; PNNL-TSR-325, Revision 8; December 2012.

U.S. Department of Energy**;** *Safety Evaluation Report for the Hanford Building 325, Radiochemical Processing Laboratory, Amendment 5*; October 2012.

PNNL Manuals

PNNL-MA-250. *Nuclear Safety and Facility Authorization Nuclear Criticality Safety Program Manual*. Pacific Northwest National Laboratory, Richland, Washington. March 2012. Available at https://facilities.pnl.gov/weblinks/ma/pnnl-ma-250.pdf.

BEP-325/RPL. *Building Emergency Procedure for Radiochemical Processing Laboratory (RPL)*. Pacific Northwest National Laboratory, Richland, Washington. January, 2012. Available at http://facilities.pnl.gov/weblinks/bep/325/bep-325.pdf.

"Radiological Protection Program Description – Implementation Plan for 10 CFR 835." Pacific Northwest National Laboratory, Richland, Washington. November 2011. Available at https://hdi.pnl.gov/hdi/product/program/pd06d010.pdf.

"Worker Safety and Health Program Description - 10 CFR 851." Pacific Northwest National Laboratory, Richland, Washington. November 2011. Available at https://hdi.pnl.gov/hdi/product/program/pd49d010.aspx.

Regulations

10 CFR 830. U.S. Code of Federal Regulations, Title 10, *Energy*, Part 830, "Nuclear Safety Management."

10 CFR 835. U.S. Code of Federal Regulations, Title 10, *Energy*, Part 835, "Occupational Radiation Protection,"

10 CFR 851. U.S. Code of Federal Regulations, Title 10, *Energy*, Part 851, Worker Safety and Protection Program,"
29 CFR 1910. U.S. Code of Federal Regulations, Title 29, *Labor*, Part 1910,

"Occupational Safety and Health Standards," Section 1200, "Hazard Communication."

29 CFR 1910. U.S. Code of Federal Regulations, Title 29, *Labor*, Part 1910, "Occupational Safety and Health Standards," Section 1450, "Occupational Exposure to Hazardous Chemicals in Laboratories."

29 CFR 1910. U.S. Code of Federal Regulations, Title 29, *Labor*, Part 1910, "Occupational Safety and Health Standards," Subpart Z, "Toxic and Hazardous Substances."

U.S. Department of Energy Orders, Manuals, Standards, and Guides

DOE O 420.1B. *Facility Safety*. U.S. Department of Energy, Washington, D.C. December 22, 2005.

DOE-STD-1027-92. *Hazard Categorization and Accident Analysis Techniques for Compliance with DOE Order 5480.23, Nuclear Safety Analysis Reports,* Change Notice 1. U.S. Department of Energy, Washington, D.C. 1997.

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