

EXECUTIVE SUMMARY

Review of Plutonium Oxide Receipts Into Hanford Tank Farms

Based upon the results of the M-3 mixing studies for the Waste Treatment and Immobilization Plant (WTP) pretreatment facility, a concern emerged over the settling of greater than 10 micron plutonium-bearing particles in WTP process vessels. Specifically, members of the WTP technical staff believed that the Plutonium Finishing Plant (PFP) may have transferred large, dense plutonium oxide particles to the tank farms which could then be sent to WTP for processing. Therefore, WTP management commissioned a three-person team (WTP team) of former Hanford employees with extensive PFP and tank waste experience to investigate if large, dense plutonium-bearing particles could be present in the tank farms.

In February 2011, the draft report prepared by this team was given to Washington River Protection Solutions (WRPS) management for their review. The draft report indicated that there was a possibility that large, dense plutonium-bearing particles could be present in the waste tanks. WRPS concluded that the presence of such particles could exceed the analyses of the criticality safety evaluation report (CSER) for the waste tanks. In response to these findings, and because the original team did not have broad access to process documentation and no access to classified documentation, WRPS management commissioned a second team (WRPS team) to investigate the issue and determine the extent of the problem using all available sources of information.

The scope of the WRPS team's review was to:

- Determine all source facilities that contributed plutonium oxide (PuO_2) or plutonium oxalate to the tank farm
- Determine all the tanks that received PuO_2 or oxalate
- Determine how much plutonium (Pu) was sent to each tank and identify its chemical and physical form
- Determine the particle size and density of the plutonium disposed to the tank farms, specifically looking for particles greater than 10 microns.

As part of the review, questions about the disposal of plutonium fluoride compounds (e.g., PuF_4) to the Hanford waste tanks were investigated. Accordingly, the WRPS team reviewed the chemistry of Pu oxalate and Pu fluoride compounds to determine if large dense particles would persist in the alkaline tank waste environment. The investigation determined that oxalate and other crystalline forms, when not calcined and after neutralization, were converted to light, fine particles of low-solubility plutonium hydrous oxide, $\text{PuO}_2 \cdot x \text{H}_2\text{O}$, and would not be an issue. In addition, the precipitated plutonium would be bonded to other particles or poisons, making a co-precipitated agglomerate. In this manner, they would be no different than the other plutonium hydrous oxide discarded as reprocessing operations losses in the tank farm. Plutonium fluoride compounds likewise were found to form finely divided low-solubility plutonium hydrous oxide.

When reviewing all the processes that sent Pu to the tank farms, three facilities; PFP, Plutonium Uranium Extraction (PUREX) plant, and Reduction-Oxidation (REDOX) were found that sent large dense particulate plutonium-bearing material. The materials were primarily in the form of calcined plutonium dioxide, PuO_2 . This team estimated that approximately 100kg of Pu was sent to the tank farms from plutonium oxide processing in different facilities. Of this quantity, approximately 30 kg was disposed as large (>10 micron) and dense calcined PuO_2 and plutonium metal fines, with the balance discarded in the form of plutonium nitrate, plutonium hydroxide, or plutonium oxalate and compounds that would readily decompose in alkaline waste solution to form fine plutonium hydrous oxide.

The PFP facility contributed a majority of the calcined PuO_2 with approximately 23 kg plutonium sent that was greater than 10 microns. Of that 23 kg, the WRPS team estimates that PFP discarded as much as 2.5 kg of plutonium as fine metal particles from incomplete metal burning subsequent to oxide dissolution. The PUREX and REDOX facilities sent approximately 7 kg of similar calcined plutonium oxide material.

Eight tanks received an appreciable quantity (>750 g) of oxide or metal from the three facilities. They are TX-105, -109 and -118, 244-TX, SY-102, C-102, AN-101, and S-108. In addition there were eight more tanks that received minimal amounts (<400 g) of oxide or metal and they are A-105, BX-101, S-107, -111, SX-114, B-101, TX-101, and C-104.

The size of the disposed Pu metal and dioxide particles were determined based upon the method used to process the plutonium and historical sources. The PuO_2 derived from oxalate processing has a particle size of 1 to 40 microns and a particle density of 8–11 g/cc. The PuO_2 derived from metal burning has a particle size from 40 to 100 microns and a density of 8-11 g/cc. The Pu metal fines from rework of material and scrap recovery (largely as unreacted metal) have an estimated particle size of 40 to 100 microns and a density of approximately 19 g/cc.

In determining the 30 kgs of material sent to the tank farm, several margins of safety were used and are discussed later in this report. If the margins of safety were reduced, the amount of material sent to the tank farm could be lowered to approximately 18 kgs. However, this investigation has no basis to remove the margins at this time.

The plutonium loss inventory estimates made by this team are consistent with the 65-130 kg of Pu (midpoint 98 kg) sent to the tank farms estimated by the WTP team. The present investigation has refined the estimates, provided information on the plutonium chemical and physical properties, particularly the density and particle size, and delineated the quantities and waste tanks that received the plutonium.

The results of this team were reviewed by an independent review team of 4 experts in nuclear operations management and oversight, scientific investigation, nuclear and criticality safety, radiological characterization, and plutonium chemistry and processing. They spent approximately one month reviewing the report, receiving presentations and

interviewing team personnel. The team concluded that the investigation and data analysis activities were complete, thorough and comprehensive with the conclusions being reasonable, conservative, and useful in full recognition of the uncertainties inherent with the historical waste data quality.

In summary, the WPRS team estimates that approximately 30 kgs of plutonium present in the tank farms was delivered as >10-micron particulate Pu oxide and Pu metal. This inventory is located in 16 tanks; eight with minimal quantities, and eight with appreciable quantities that could challenge the CSER. Particle sizes range from 10–100 microns and densities range from 8–11 g/cc with approximately 2.5 kg present as 19 g/cc metal.