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Plutonium Processing Optimization in Support of the MOX Fuel Program

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INTRODUCTION

After Los Alamos National Laboratory (LANL) personnel completed polishing 125 Kg of plutonium as highly purified PuO_2 from surplus nuclear weapons, Duke, COGEMA, Stone, and Webster (DCS) required as the next process stage, the validation and optimization of all phases of the plutonium polishing flow sheet. Personnel will develop the optimized parameters for use in the upcoming 330 kg production mission.

PRODUCTION PHASE

Processing 125 kg of plutonium on a monthly basis presented the following challenges for LANL personnel:
· Dissolution upgrades required testing and validation when completed
· Anion exchange resin, since near the end of life, needed replacement.
· Anion exchange wash volume was excessive
· +4 oxalate precipitation needed validation of key parameters

Each process underwent testing and validation during the optimization phase according to test requirements. Personnel performed rigorous sampling following problem elements throughout the wash phase and the oxalate precipitation process.

Optimization Phase

LANL and DCS agreed to test these processes and validate selected steps.

Table 1 Optimization Phase

Process	Requirement
Dissolution	Vary HF and HNO3 concentrations during runs
Anion Exchange	Validate the new 100% quaternized HPQ Polymer Reillex™ resin. Validate the use of recycled nitric acid as wash, and optimize the volume of wash required.
+4 Oxalate Precipitation	Vary H_2O_2, oxalate flow rates and cooling rate
Calcination	Test inconel 601™ boats

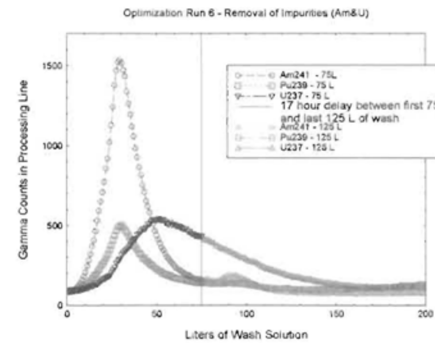

Fig. 1. Gamma Monitor Output for Optimization Run 6

DESCRIPTION OF THE ACTUAL WORK

Using smaller batch sizes (500 g. dissolution runs instead of 1 kg.), personnel performed experiments that showed higher average dissolution efficiencies by increasing both the fluoride concentration and the mechanical mixing during the refluxing and by eliminating the losses associated with poor filtration.

For the anion exchange process, the new 100% quaternized HPQ Polymer Reillex™ resin performed well as expected, without any significant plutonium losses. The output from the gamma monitor, shown in Figure #1, showed when the radioactive impurities were washed through. Working with recycled acid met the strict product purity requirements and demonstrated a reduction in the wash volume.

For the +4 oxalate precipitation process, personnel changed the plutonium valence with H_2O_2 immediately prior to precipitation. Oxalate flow rate addition impacts oxide surface area, while cooling rate of the precipitate does not.

For the calcination process, the inconel 601™ calcination boat met specifications.

RESULTS

Dissolution efficiency increased from 80% to greater than 90%. Ion exchange wash volumes showed a reduction to less than half, using the gamma monitor as a check, while recycled acid use resulted in small increases in wash impurities. In precipitation, timely H_2O_2 addition is a key parameter, as is the addition flow rate of oxalate at temperature.