

Capability to Recover Plutonium-238 in H-Canyon/HB-Line - 13248

Kenneth S. Fuller Jr., Robert H. Smith Jr., Charles R. Goergen
Savannah River Nuclear Solutions, LLC
Savannah River Site, Aiken, SC 29802
kenneth.fuller@srs.gov
robert.smith@srs.gov
charles.goergen@srs.gov

ABSTRACT

Plutonium-238 is used in Radioisotope Thermoelectric Generators (RTGs) to generate electrical power and in Radioisotope Heater Units (RHUs) to produce heat for electronics and environmental control for deep space missions. The domestic supply of Pu-238 consists of scrap material from previous mission production or material purchased from Russia. Currently, the United States has no significant production scale operational capability to produce and separate new Pu-238 from irradiated neptunium-237 targets. The Department of Energy - Nuclear Energy is currently evaluating and developing plans to reconstitute the United States capability to produce Pu-238 from irradiated Np-237 targets.

The Savannah River Site had previously produced and/or processed all the Pu-238 utilized in Radioisotope Thermoelectric Generators (RTGs) for deep space missions up to and including the majority of the plutonium for the Cassini Mission. The previous full production cycle capabilities included: Np-237 target fabrication, target irradiation, target dissolution and Np-237 and Pu-238 separation and purification, conversion of Np-237 and Pu-238 to oxide, scrap recovery, and Pu-238 encapsulation.

The capability and equipment still exist and could be revitalized or put back into service to recover and purify Pu-238/Np-237 or broken General Purpose Heat Source (GPHS) pellets utilizing existing process equipment in HB-Line Scrap Recovery, and H-Canyon Frame Waste Recovery processes. The conversion of Np-237 and Pu-238 to oxide can be performed in the existing HB-Line Phase-2 and Phase-3 Processes. Dissolution of irradiated Np-237 target material, and separation and purification of Np-237 and Pu-238 product streams would be possible at production rates of ~ 2 kg/month of Pu-238 if the existing H-Canyon Frames Process spare equipment were re-installed. Previously, the primary H-Canyon Frames equipment was removed to be replaced; however, the replacement project was stopped. The spare equipment is stored and still available for installation.

Out of specification Pu-238 scrap material can be purified and recovered by utilizing the HB-Line Phase-1 Scrap Recovery Line and the Phase-3 Pu-238 Oxide Conversion Line along with H-Canyon Frame Waste Recovery process. In addition, it also covers and describes utilizing the Phase-2 Np-237 Oxide Conversion Line, in conjunction with the H-Canyon Frames Process to restore the H-Canyon capability to process and recover Np-237 and Pu-238 from irradiated Np-237 targets and address potential synergies with other programs like recovery of Pu-244 and heavy isotopes of curium from other target material.

INTRODUCTION

Historically the United States has used plutonium-238 in Radioisotope Thermoelectric Generators (RTGs) to generate electric power and in Radioisotope Heater Units (RHUs) to produce heat for electronics and environmental control for deep space missions. The domestic supply of Pu-238 consists of scrap material from previous mission production or material purchased from Russia. Currently, the United States has no significant production scale operational capability to produce and separate new Pu-238 from irradiated neptunium-237 targets.

In the late 1950s, the Savannah River Site (SRS) began generating and collecting neptunium-237 from the irradiation of uranium-235 in SRS reactors. The neptunium-237 collected was purified, converted to an oxide, and fabricated into reactor targets. The neptunium targets were then irradiated or bombarded with neutrons to produce plutonium-238. The nuclear reactions representing this irradiation cycle process and decay chain is represented in Figure 1. [1, 2]

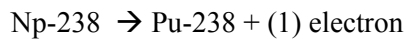
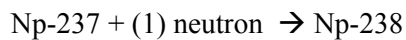


Figure 1. Nuclear Reactions and Decay of Neptunium-237 to Plutonium-238

The neptunium-237 and plutonium-238 were chemically separated, purified, calcined to oxide, the neptunium recycled into targets, and the plutonium pressed into pellets or spheres and encapsulated into iridium cladding and used in RTGs. From 1961 until 1984, the Savannah River Site operated a complete on-site production cycle for neptunium target fabrication, target irradiation, neptunium target processing, and plutonium-238 encapsulation. The Savannah River Site started and produced or processed nearly all of the plutonium-238 utilized in 44 RTGs and over 200 RHUs for 25 space missions up to and including the Cassini mission in 1997. [3] The historical SRS full Pu-238 production cycle included the following capabilities:

- Neptunium recovery from irradiated recycled uranium fuel (H-Canyon)
- Fabrication of Np-237 targets (235-F Billet Line, 300-M Area fabrication MK-53A/B targets)
- Irradiation of Np-237 targets (SRS Reactors)
- Dissolution of irradiated Np-237 target material, separation and purification of Np-237 and Pu-238 product streams (H-Canyon Frames Process)
- Conversion of Np-237 and Pu-238 to oxide (HB-Line Phase-2 and Phase-3 Processes)
- Recovery of primary Frames and HB-Line losses (H-Canyon Frames Waste Recovery Process)
- Pressing of Pu-238 oxide into GPHS pellets and encapsulation in iridium metal claddings (235-F)
- Recovery and purification of off specification Pu-238/Np-237 or broken GPHS pellets (HB-Line Scrap Recovery, Frame Waste Recovery)

With the shutdown of SRS reactors in the early 1990s, a number of the plutonium-238 processing capabilities were shut down, deactivated, or removed. However, the following capabilities and equipment still exist and could be put back into service or revitalized if needed:

- Recovery and purification of off-specification Pu-238/Np-237 or broken GPHS pellets (HB-Line Scrap Recovery, Frame Waste Recovery)
- Conversion of Np-237 and Pu-238 to oxide (HB-Line Phase-2 and Phase-3 Processes)
- Dissolution of irradiated Np-237 target material, separation and purification of Np-237 and Pu-238 product streams (H-Canyon Frames Process)

- Recovery of primary Frames and HB-Line losses (H-Canyon Frames Waste Recovery Process)
- Neptunium recovery from irradiated uranium fuel (H-Canyon)

DISCUSSION

H-Area assets could be revitalized to support a larger scale Pu-238 scrap purification mission (>4 kg/yr.) to clean up domestic scrap Pu-238 or Pu-238 of foreign origin. This type of campaign would require the use of HB-Line Phase-1 Scrap Recovery process, the H-Canyon Frame Waste Recovery (FWR) process, and the HB-Line Phase-3 process. See the SRS Pu-238 scrap purification process overview diagram in Figure 2. All of these processes still exist today and would require restart with some modifications, piping reconfiguration, repair, and readiness review.

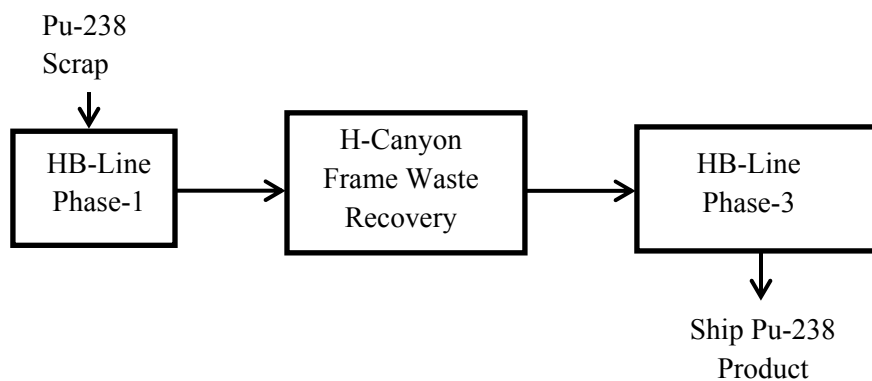


Figure 2. SRS Plutonium-238 Scrap Purification Overview

The HB-Line Scrap Recovery process consists of two dissolvers and associated in-line filters and product hold tanks as shown in the diagram below. Plutonium materials would be received into HB-Line, the shipping containers opened; the material size reduced if necessary, and then dissolved in nitric acid in the Scrap Recovery dissolvers. Once dissolved, the plutonium solution would be transferred to an H-Canyon storage tank through an existing transfer line, accumulated, then transfer to the FWR process as shown in Figure 3.

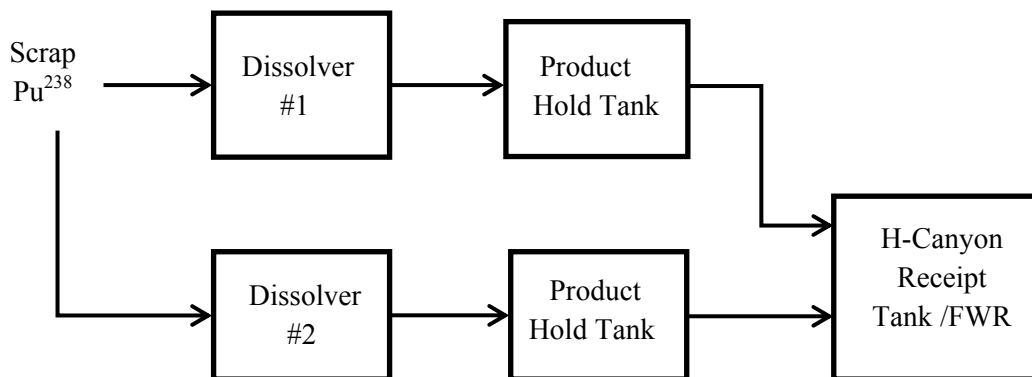


Figure 3. HB-Line Phase-1 Scrap Recovery

The FWR Process consists of a single 85 liter anion exchange resin column with an associated feed tank, waste tank, product tank, and waste evaporator with corresponding support tanks. See Figure 4 for diagram of FWR Process. In FWR, the feed solution is chemically adjusted, then fed to the single resin column and adsorbed onto the resin, decontaminated by washing the resin bed with clean nitric acid, then eluted (washed with weak nitric acid) to a product hold tank.

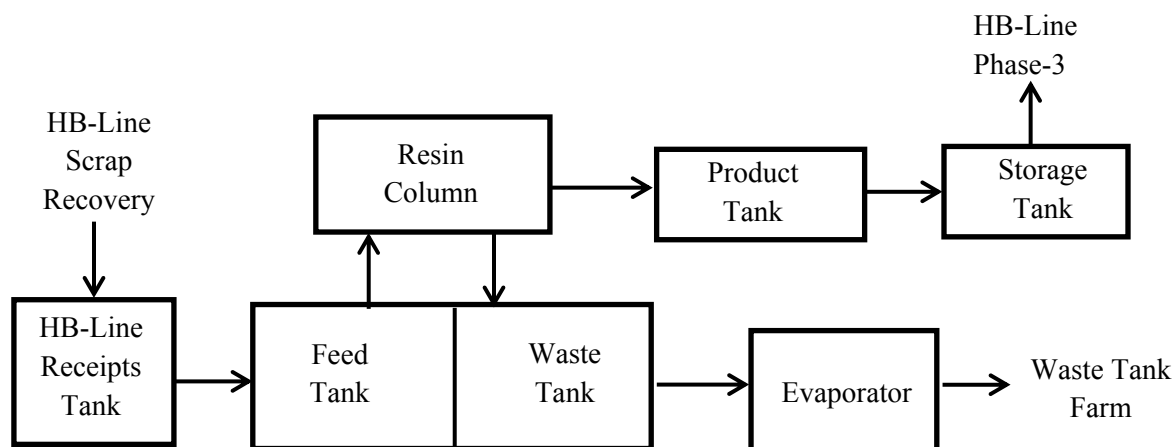


Figure 4. Frame Waste Recovery Process

The purified FWR product solution is transferred to the HB-Line Phase-3 process. In Phase-3, the plutonium solution is chemically adjusted, the plutonium precipitated using oxalic acid, filtered, washed, dried, calcined to an oxide, and packaged for shipment. See Figure 5 for the HB-Line Phase-3 Pu-238 Oxide Conversion process.

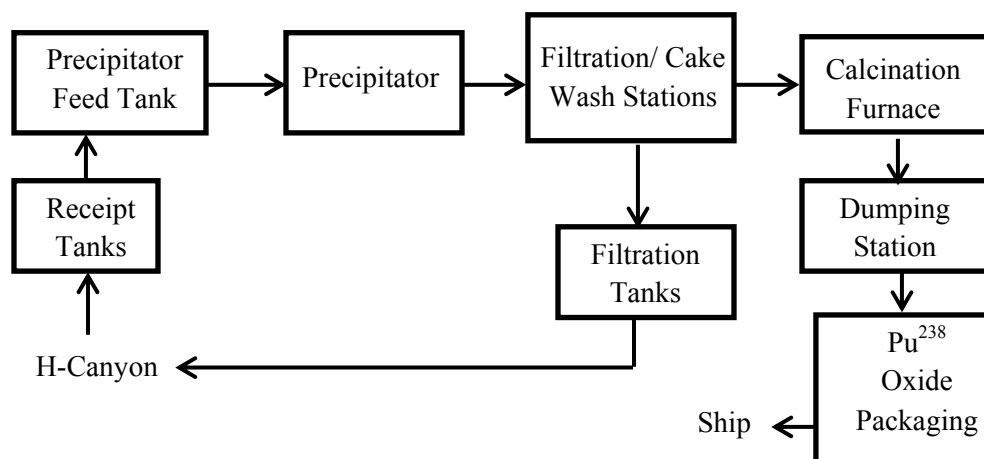


Figure 5. HB-Line Phase-3 Pu-238 Oxide Production Process

H-Area assets could also be revitalized to support a larger scale combination Np²³⁷ target processing and scrap Pu²³⁸ purification mission (>4 kg/yr.) This type of campaign would require the use of H-Canyon Frames Process, HB-Line Scrap Recovery process, the H-Canyon Frame Waste Recovery (FWR) process, and the HB-Line Phase-2 and Phase-3 processes.

The original H-Canyon Frames Process for recovering Pu-238 from irradiated Np-237 targets was installed in 1961 and operated until 1984 recovering and separating Pu-238 and Np-237 for the NASA Space Program. After shutdown and cleanout of equipment in 1984, a project to upgrade the H-Canyon Frames Process equipment was begun and nearly completed, but stopped prior to completion. Most of the H-Canyon Frames Process services, instrumentation, and support equipment were installed during the previous upgrades. The actual canyon Frames process modules were not installed, but are stored as spare equipment and are still available for installation.

To recover Pu-238 from irradiated Np-237 targets, the H-Canyon Frames Process would have to be restarted to separate, purify, and concentrate the Np-237 and Pu-238. These product solutions would then require conversion to oxide form, and packaging for shipment in the HB-Line Phase-2 and Phase-3 facilities. If a high percentage of recovery of these two isotopes is required, then the Frame Waste Recovery Process will be required to collect and recycle any losses from the H-Canyon Frames Process and HB-Line Phase-2 and Phase-3 Processes. If rework of any off-specification product is required, then utilizing HB-Line Phase-1 Scrap Recovery Line is also required. Figure 6 shows the H-Canyon and HB-Line processes needed to recover Pu-238 from irradiated Np-237 targets. All of these processes and facilities exist and would require restart with some modifications, piping reconfiguration, repair, and readiness review.

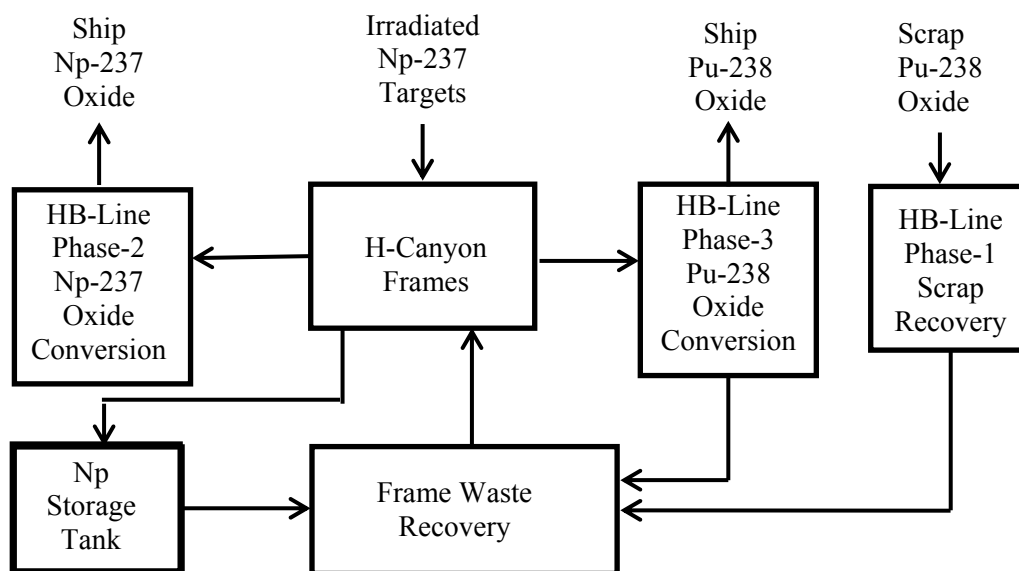


Figure 6. H-Canyon and HB-Line Process Overview to Recover Pu-238 from Irradiated Np-237 Targets

The H-Canyon Frames Process consists of two remotely operated and removable frame units containing integrated tanks, resin columns, instrumentation, airlifts, pumps, piping, and service connections to the

canyon utilities through remotely installed jumpers (by means of an overhead crane). A picture of spare H-Canyon Frames modular equipment is shown in Figure 7.

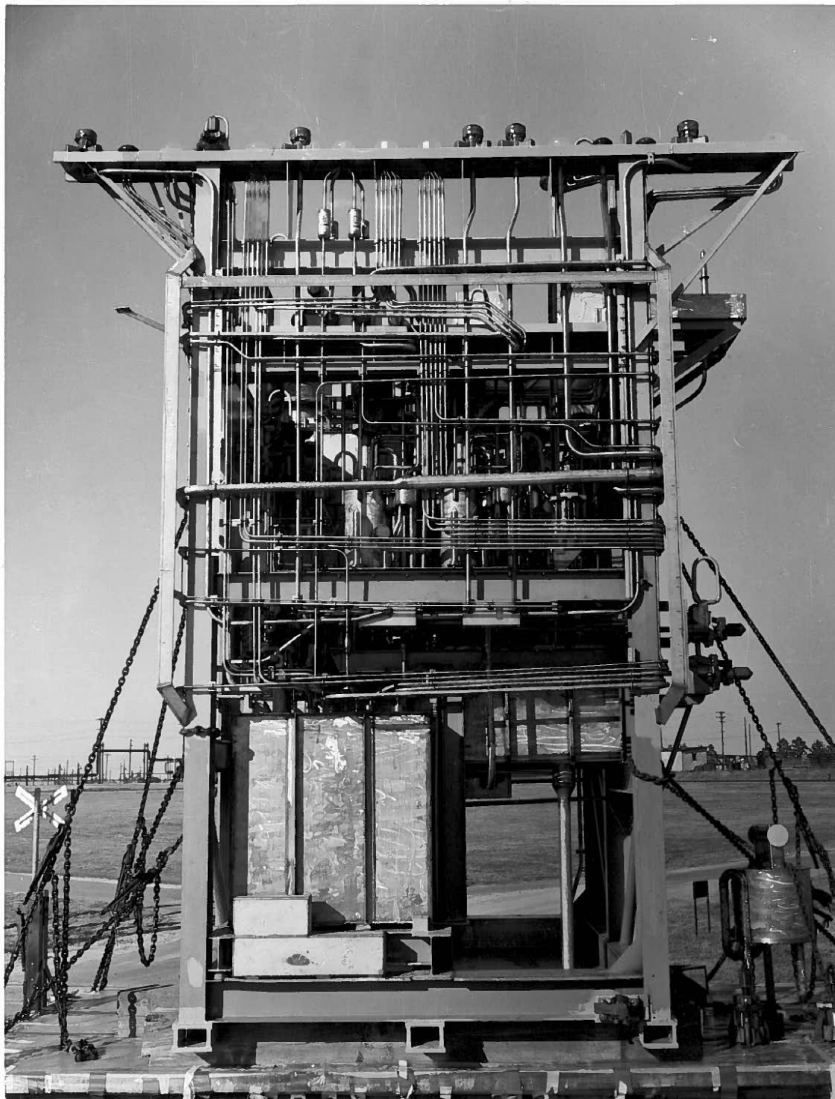


Figure 7. Spare H-Canyon Frames Modular Equipment

A separate, external dissolver for dissolution of Np-237 targets in nitric acid is collocated and piped to the H-Canyon Frames Process receipt tank. A separate, external waste collection tank is also collocated next to the Frames modules and dissolver to collect the H-Canyon Frame Process waste solutions and actinides losses for recycle or disposition in Frame Waste Recovery. See the diagram in Figure 8 of the H-Canyon Frames Process.

Within the Frames Process shown in Figure 8, Np-237 target material dissolved in nitric acid is received, chemically adjusted and feed to resin column #1 where the Np and Pu actinides are adsorbed on the anion resin.[5] The column is then washed to remove residual fission products and other contaminants going to

a waste collection tank, and the Np and Pu are co-eluted to a second resin column adjustment tank. Once chemically adjusted, the feed solution is fed to resin column #2 where the Np and Pu are adsorbed on its anion resin. The resin column is then washed with a partition solution that selectively elutes the Pu from the resin to the resin column #3 adjustment tank. The remaining Np on the resin column #2 is washed to remove additional contaminants and then transferred to HB-Line Phase-2 for conversion to oxide or transferred to a canyon tank for storage. The feed to resin column #3 is adjusted and fed to the column where the Pu is adsorbed onto anion resin, washed, eluted, and transferred to HB-Line Phase-3 for conversion to oxide. All wash solutions are collected in a waste collection tank and subsequently transferred to FWR for additional recovery and recycle of Np-237 and Pu-238.

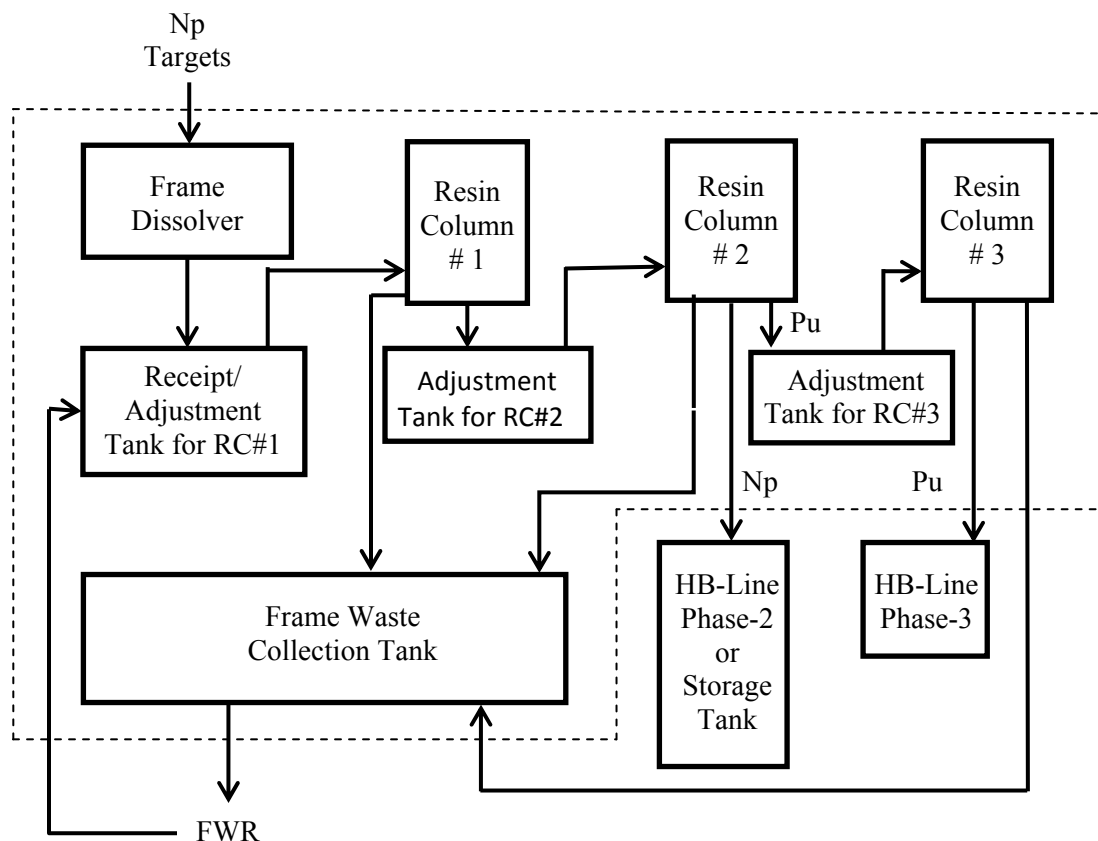


Figure 8. H-Canyon Frames Process

The H-Canyon and HB-Line production capability based on historical operations production rates could support mission needs up to approximately 3 kg Pu-238/month if sufficient Pu-238 scrap is available to blend with irradiated Np-237 target material.

Some of the positive attributes or synergies provided by SRS asset revitalization of Pu-238 capabilities include:

- Utilizes existing H-Canyon and HB-Line Pu-238 processing experience
- Flexibility to recover high percentage of Pu-238 and Np-237 by recycling losses
- Ability to increase Pu-238 production capacity with increased staffing strategies to ~12kg –30kg/yr utilizing irradiated Np-237 targets and/or scrap Pu-238

- Ability to operate and store Pu-238 and Np-237 in liquid form in H-Canyon storage tanks awaiting HB-Line oxide conversion campaigns
- Existing H-Canyon Frames dissolver and resin columns in FWR or primary Frames could be utilized for recovery of Pu-244, Am-243, and heavy curium from MK-18A targets.
- Could increase value of US stockpile of 70% Pu-238 by blending with ultrapure 95%+ Pu-238 produced from irradiation of Am-241 targets.

Some of the potential facility conflicts or impacts with utilizing H-Canyon and HB-Line for Pu-238 processing requiring process prioritization include the following:

- Use of Phase -2 for production of early MOX feed may require storing Np-237 as liquid in H-Canyon until Phase -2 is available
- Potential use of H-Canyon, hot canyon cells where Frames would be installed (section 5H) may be used for R&D projects
- Possible liquid waste impacts in out-years after 2028 per the current Liquid Waste System Plan Tank Farm closure schedule [6]
- DOE-EM long-term H-Canyon/HB-Line base-load funding support to make incremental costing to produce Pu-238 cost effective

CONCLUSIONS

Revitalization of existing SRS assets and Pu-238 capabilities could be utilized to support various mission needs while supporting the DOE-Environmental Management cleanup missions. The ability to use irreplaceable assets to serve missions of national security and furthering development activities or missions for the Department of Nuclear Energy are worth pursuing.

REFERENCES

1. Groh, H. J., W. L. Poe, J. A. Porter, "Development and Performance of Processes and Equipment to Recover Neptunium-237 and Plutonium-238", WSRC-MS-2000-00061, Savannah River Site, Aiken, SC, 2000.
2. Poe, W. L., A. W. Joyce, R. I. Martens, "Np-237 and Pu-238 Separation at the Savannah River Plant", USAEC Report DPSU 63-30-44, Savannah River Plant, Aiken, SC, 1963.
3. Buden, David, "Space Nuclear Propulsion and Power Book 1: Space Nuclear Radioisotope Systems", Polaris Books, 2011.
4. Rinehart, G. H., "Design Characteristics and Fabrication of Radioisotope Heat Sources for Space Missions", Progress in Nuclear Energy, vol.39, No. 3-4, pp. 305-319, 2001.
5. Fuller, K. S., T. M. Thompson, "Recovery and Purification of Plutonium-238 in the H-Area Frames - Training Manual", DOSPU 82-11-20, 1982.
6. Chew, D. P., B. A. Hamm, "Liquid Waste System Plan", SRR-LWP-200900001, revision 17, 2012.