A FACILITY FOR THE PRODUCTION OF Pu^{238}*

by

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ABSTRACT

A pilot plant facility for recovering Pu^{238} and unconverted neptunium from irradiated neptunium target elements was operated for 17 months at the Savannah River Laboratory.

The process required that the irradiated target elements be dissolved and the solution be processed through three anion exchange cycles for removal of undesirable fission products and cations, recovery of unconverted neptunium, and concentration of the Pu^{238} solution.

The process equipment was enclosed in three stainless steel boxes that were installed in two general-purpose cells of a ten-cell complex. The basic cells were not modified. The two cells were not separated from the adjacent 8 cells.

Containment of the high-specific-activity alpha emitters was accomplished by ventilation, development of handling techniques, use of procedures, and close Radiation Control coverage.

The facility was dismantled, and the cells were decontaminated and returned to normal use.

With the exception of an accidental overexposure, all personnel exposures were kept below the administrative level of 3 R/yr. None of the personnel assimilated detectable amounts of the material handled.

*The information contained in this article was developed during the course of work under contract AT(07-2)-1 with the U. S. Atomic Energy Commission.

DISCLAIMER

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INTRODUCTION

During the period from May 1959 to October 1960, a pilot plant facility for producing relatively large quantities of Pu^{238} was operated by the Savannah River Laboratory. The facility was installed in two general-purpose shielded cells in the High Level Caves wing of the Laboratories. The facility, the process, and subsequent dismantling of the facility and decontamination of the cells are described to illustrate:

1. General-purpose cells can be used for work with large quantities of high-specific-activity alpha emitters.
2. General-purpose cells can be used for a small scale production unit.
3. The cells can be recovered for their general-purpose use at the conclusion of the special work.

PROCESS DESCRIPTION

The target slugs consisted of a compacted mixture of neptunium dioxide and aluminum powder enclosed in an aluminum can. After irradiation, the slugs were dissolved in 10M HNO_3, with hydrofluoric acid and mercuric nitrate added as catalysts. The dissolver solution was processed through three anion exchange cycles to remove fission products and undesirable cations, to separate the neptunium and plutonium, and to concentrate the neptunium and plutonium solutions. The process is shown schematically in Figure 1.

The first anion exchange cycle removed the major portion of the fission products and non-actinide metals. The second cycle separated the Np and Pu, removed additional fission products,

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and concentrated the Np solution for further processing. The third cycle concentrated the Pu solution and removed additional fission products. The Np solution from the second cycle was removed from the unit into a cask and transferred to a gloved box unit for further processing. The Pu solution normally was transferred directly into a gloved box unit where the volume was measured and the solution was sampled. The solution was then placed in a metal bottle for shipping. If desired, the Pu solution could be removed from the unit into a transfer cask and taken to a gloved box unit for further concentration.

FACILITY DESCRIPTION

Design Consideration

Design, construction, and installation of the facility was done on a high priority basis. The project was completed in a period of 9 months at a cost of approximately $280,000.

The primary design problems were the containment of radioactive contamination and remote replacement of component parts. It was also important that examination of irradiated reactor components, being carried out in adjacent shielded cells, continue with minimum interference from the plutonium production operations.

High Level Caves

The Savannah River Laboratory (SRL) High Level Caves consist of two cell blocks divided into 10 general-purpose cell modules. The caves are housed in a separate wing of the main laboratory building. Figure 2 shows the floor plan and an elevation section of the cave wing.

The Interim Production Facility for the production of Pu^{238} was installed in two of the general-purpose cells. Although material with a high specific alpha activity was to be handled, and common loading and operating areas were shared by all 10 cells, barriers were not erected to separate the Interim Production Facility from the other 8 cells.

The two cells which were used are 6 feet by 12 feet in floor area and 17 feet high. The walls are of high density concrete 3 feet thick with two lead-glass shielding windows in the operating side. A heavy duty electromechanical manipulator and a one-ton bridge crane are mounted on rails that run the length of the cell block. The manipulator and crane can be moved from cell to cell by raising steel barriers in the inter-cell walls. There is an access door at the rear of each cell, and the cell roof covers are removable. A shielded transfer and storage drawer was installed in the inter-cell wall.

Each cell is equipped with four model 8 master-slave manipulators (MSM's), two at each window, and with a drain sump to catch liquid spills. Liquid can be jetted or pumped from the pumps to the building drain system for radioactive liquids. Service plugs and transfer blocks are provided for piping utility and electrical services into the cells. The cells are fitted with disposable polyvinyl chloride liners to avoid gross contamination of the walls.

Normal cell exhaust is 500 cfm per cell. Air inlet is through fiberglas duststop filters located in slots on the cell roof; there is also a duststop exhaust filter near the floor of each cell. After leaving the cell, the air is exhausted through a second duststop and a CWS absolute filter located in the equipment room. Fan exhaust is released to the environment. Spare filter units and fans are provided.

Figure 3 shows the arrangement of the equipment installed in the High Level Caves for production of Pu^{238}. Process equipment for dissolving the target slugs, decontamination by anion exchange, and separation of the plutonium product and unconverted neptunium were installed in three containment boxes located in Cells 1 and 2. Cell 1 also contained a cask-unloading station where the irradiated slugs were unloaded and transferred to the dissolver in Box 1 or to a shielded drawer for storage.

Instrument panels and control consoles were provided at the control face at convenient locations. A sampling station, a resin removal station, a plutonium bottling station, and a product transfer-cask station were located at the rear face of the cells.

A cold-feed preparation station was located outside the building and was protected from the weather by a canvas tent. The station consisted of an elevated platform supporting tanks for chemical storage and the make-up and addition of process reagents. Pumps and air pressure tanks were provided for transfer of solutions into the process system.

Process Containment Boxes

The primary containment boxes were constructed of stainless steel angle framing and had stainless steel or "Homolite" panels. Each box was 8-1/2 feet high and 4 feet deep. Box 1 was 6 feet wide and Boxes 2 and 3 were 5 feet wide. Each box was equipped with an air lock entry port. The air locks had top-opening lids and a sliding tray. A sliding door separated the air lock from the inside of the box. Figures 4 and 5 are pictures taken of Box 2 during construction.

The containment boxes were fabricated in two sections. The bottom section was completely enclosed with 16-gauge stainless steel continuously welded to the angle frame and separated from the top section by a 3/8-inch reinforced plate. The 3/8-inch plate served as a working-level deck and as a common top to which the process tanks were welded.

The top sections had "Homolite" and stainless steel panels and were equipped with ports for master-slave manipulators (MSM's) and a 50-pound bridge crane. Piping, valves, resin columns, process filters, liquid traps, and other equipment were housed in the top sections. Connections for process piping and electrical and instrument services were made at bulkhead fittings and amphenol plug panels in the upper section.

The MSM's were protected from gross contamination by "Hypalon" and polyvinyl chloride gauntlets. A polyethylene ring at the top of the gauntlets snapped into a groove on the box front panel to provide a seal.

Process Equipment

The principal materials used to fabricate process equipment were 304L stainless steel, polyethylene, "Teflon", and glass to resist nitric acid corrosion. Electric motors, pneumatic valve operators, and the like, which were not fabricated from corrosion resistant materials, were covered with "Hypalon" tape and paint.

Transfer of process solutions was accomplished by vacuum or pressure via dip tubes in each tank. Two tanks in each box could be valved to a vent or to a vacuum or pressure header. One of these tanks was intended primarily as a transfer tank and the other primarily as a resin-column feed tank. The feed adjust, waste, and product tanks were permanently vented. They were equipped with bubbler tubes for liquid level measurements, sample dip tubes, electric stirrers or air sparge heads for mixing, and electric ring heaters and water cooling jackets where required by the process.

All solution transfers between containment boxes and from the boxes to the sample and product loading stations were accomplished by vacuum. Waste solutions were normally vacuum transferred to a waste hold tank located in Cell 1. The tank could be vented and drained by gravity into the radioactive-liquid drain system. Water jets were installed as an alternate system in case the normal system failed.

Stainless steel tubing and "Swagelok" fittings were used for process piping. Two types of valves were used. "Hoke" AOYT-440 bellows seal valves, with "Hypalon" diaphragms and with stainless steel substituted for brass trim in the pneumatic operators, were used for undirectional flow control. Where multidirectional flow was desired, "Republic" plug valves with polyethylene or "Teflon" plugs were used.

All remotely replaceable process valves, columns, filters, etc. were equipped with "Swagelok" fittings spaced to match vertical tube risers in the process piping. Polyethylene ferrules were used to prevent damage to the tube risers. Equipment pieces of like kind were interchangeable, and jigs were provided for fabricating spare assemblies.

Off-Gas Exhaust (O.G.E.)

The existing off-gas exhaust system was extended to exhaust process equipment and containment boxes. The existing system operated at low-volume and high static pressure, and was equipped with normal and spare absolute filters and exhaust blowers.

The portion of the exhaust header that was extended to serve the plutonium processing facilities was separated from the main header by an additional pair of absolute filters; service runouts from the header were also equipped with absolute filters. All air exhausted to the O.G.E. passed through 3 absolute filters.

Bag techniques were used during filter change operations to minimize spread of contamination.

Sampling Station

Sampling equipment was installed in a "Junior Cave"* located in the cell loading area. The Junior Cave provided shielding equivalent to 3 inches of steel. Transfer of material into and from the Junior Cave was accomplished with an electromechanical elevator and air lock. Operations inside the cave were performed with standard Junior Cave tongs.

After operation of the facility began, it was decided that the operation and maintenance of the sampler could be simplified

*A "Junior Cave" is a semi-portable shield with steel walls 3 inches thick.

by replacing the cave front with a gloved panel front and adding local shielding to the hot lines and sample bottle positions. These changes were accomplished without incident. Samples could then be removed from the cave by a bag-out technique.

Cask Loading Stations and Casks

Two cask-transfer stations were attached to the cell wall in the loading area. These consisted of stainless steel and "Plexiglas" enclosures with sliding door ports.

One resin waste cask, and two neptunium and two plutonium transfer casks were provided. A stainless steel gloved box, with a sliding door to match the sliding door on the loading stations, was mounted to the top of each cask. The cask and gloved box assemblies could be attached to the loading stations with toggle clamps. A neoprene gasket seal was provided between the sliding door faces of the loading stations and the cask gloved boxes.

Piping connections between the loading stations and casks were made with polyethylene tubing and "Swagelok" fittings. Each transfer station was exhausted by an off-gas exhaust system at a rate of approximately 25 cfm. Air inlet was through a small absolute filter mounted on the cask gloved box.

Due to the potential spread of very high levels of radioactive material when product casks were removed from the product transfer station, the equipment was modified to permit a bag seal removal technique. Plates with 8-inch bag rings were attached to the transfer station and cask gloved boxes. Casks were connected to the station via 8-inch plastic sleeves with O-rings at each end to provide seals to the bag rings.

Product Loading Station

A plutonium product loading station was provided in the cell loading area adjacent to the product transfer loading station. The station consisted of a two-section gloved box made of "Micarta"-faced plywood and "Plexiglas". Product solution measuring and sampling was performed in the upper section; the lower section contained a pneumatically operated rack which positioned special stainless steel shipping containers beneath a loading port in the upper section. A bag technique was used to attach or release product containers from the loading station.

Vacuum System

A separate vacuum system was provided for the processing facility. The system consisted of 3 Welch Duo-Seal vacuum pumps, which served the process transfer tanks, the high-level-waste hold tank, and the sample station.

The vacuum pumps were installed on the Cell 1 roof inside stainless steel and "Plexiglas" enclosures connected to the off-gas exhaust system. This prevented release of contamination and permitted removal of the entire pump and enclosure for decontamination and maintenance.

The pump suction lines were equipped with dry-ice liquid traps, valved to the radioactive-liquid drain system. The vacuum pumps discharged to the O.G.E. through a common header equipped with an oil trap.

OPERATIONS

Staff

The staff consisted of 5 engineers, 10 technicians, and 3 Radiation Control Inspectors. Operations were on a 3-shift, 5-day week basis.

Operating Experience

The facility began operation on May 9, 1959, and was shut down on October 25, 1960. All production goals were met on time, and the product quality met the required specifications.

A high degree of flexibility was achieved by designing the facility to permit operations with the master-slave manipulators. It was possible to replace all lines, valves, and columns in the box and to repair equipment when necessary.

All of the valves in the boxes were replaced by the use of the master-slave manipulators. As many as one hundred such replacements were accomplished.

The original installation used irradiated polyethylene nuts on the "Swagelok" fittings. It was found that these nuts were easily split when loosened or tightened with the MSM's. Literally hundreds of "Swagelok" connections were broken. The polyethylene nuts were replaced with stainless steel nuts.

On one occasion, due to a change in the process, it was necessary to do considerable repiping in the first cycle box. It was possible to cut lines, cap stubs, and install new lines remotely.

Operating Techniques

During the 17 months of operations, equipment and procedures were modified, and techniques were developed to improve operations and safety. Whenever possible, a bagging technique was used to minimize the spread of contamination. Transfer casks and product bottles were bagged in place and could be removed without breaking containment. When pipe connections had to be broken or lines cut, bags were installed, and the work was done inside the bag. The MSM's were removed by pulling them into large bags. In some cases large "walk-in" bags were used.

Material was removed from the in-cell boxes through a transfer port into a shielded container or bagged out through a panel in the cell door. The use of the transfer port resulted in the cells becoming heavily contaminated. The ventilation barriers in the general-purpose cell proved to be adequate to contain the contamination within the cell.

On two occasions tanks were overfilled through errors in processing, and process solutions were drawn from the boxes in the cell into tubing in the loading or operating area. To correct the situation, it was necessary to break lines outside the cell, recover process solutions, and flush and replace lines. When the operating area was involved, plastic curtains were installed, and the cleanup was successfully accomplished without affecting the work of the other 8 cells.

DISMANTLING

Background

Production was stopped during October 1960, and the facility was placed in a standby condition. Permission to dismantle the facility was received on May 19, 1961.

Man-Power Requirements

The shift personnel were utilized to dismantle and decontaminate the facility. The shift force consisted of one engineer, 2 foremen, 10 technicians, and 2 Radiation Control Inspectors. The work was done on 2 shifts.

Actual cleanup began on May 19, 1961, and was completed by August 1, 1961. With the exception of the month of July, the cleanup operation was on a part-time basis. The man-power requirements were:

Engineer Time	100 man-hours
Foremen Time	720 man-hours
Technician Time	3,120 man-hours
Radiation Control Time	560 man-hours
Total	4,500 man-hours

General Plan

When the facility was placed on a standby basis, an engineer was assigned the task of planning the dismantling and decontamination of the facility. Due to this planning, the dismantling operation began on the day permission was received to dismantle the facility.

Plastic curtains were hung in the operating and loading area to isolate the Interim Production Facility Area from the other 8 cells. The dismantling operation was completed without interfering with the operation of the other 8 cells.

The floor area within the curtains was covered with heavy, waterproof paper, and this in turn was covered with a lighter weight, easily removed paper covering. All wall areas were covered with plastic or paper.

The consoles on the operating side of the cells, the product bottling station, sampling station, transfer cask staions, resin removal station, and vacuum pumps were removed first. All equipment was bagged in place. Extra bags were used around piping and tubing. All connections were broken inside the bag. Frequently gloves were installed in the bags to make the work easier.

Concurrently with the removal of the external equipment, the tubing, piping, and electrical lines between the boxes and the auxiliary equipment were cut. A hydraulically operated tubing cutter and crimper that could be operated from the cell roof was designed and fabricated. The radiation exposure was reduced by removing the cell roof slab and cutting the lines from a distance. Most of the tubing and wires were cut with this device. Tubing that could not be cut with the hydraulic cutter was cut with bolt cutters. Cell entry was necessary in these cases. An electrical saber saw was mounted on an extension handle and used to cut the larger O.G.E. pipes. The saber saw was mounted on a handle long enough to permit cutting from the roof.

After all connections were broken the cells were entered, and all external surfaces of the boxes were sprayed with a strippable coating to hold the contamination.

Initial cell entries were made from the roof, and personnel were lowered on a large steel plate, which served as a shadow shield from the high sources of radiation at floor level.

Large wooden disposal boxes were fabricated and placed behind the cells. Plastic aprons connected the cell roof to the boxes. The process boxes were removed from the cells, placed in the disposal boxes, and shipped to the burial ground.

The area within the plastic curtains was considered to have airborne contamination, and filter type respiratory protection was required for entry. Two plastic suits with fresh air supplied to the inner suit were required for cell entry. The outer suit protected the inner suit from gross contamination or puncture. The outer suit was sprayed with a strippable coat to fix the contamination, and was removed at the cell exit point.

After all the equipment was removed from the cells, a double airlock was constructed at the floor-level cell entry doors. Initially, the double suit technique was used for cell entries. As the work progressed and contamination levels were reduced, the clothing requirements were relaxed; two pairs of disposable coveralls, a filter type full face mask, "bootees", and a cloth hood were required.

Time Schedule

Preparation of the area was completed in one week. It took 3 weeks to remove the auxiliary equipment, to cut all in-cell lines, and to prepare the process boxes for removal. This part of the operation was done on a part-time basis. All three boxes were removed from the cells in one 8-hour shift. No difficulty was encountered and the spread of contamination was negligible.

The remaining five weeks were required to decontaminate the cells and the area outside the cell.

RADIATION CONTROL EXPERIENCE

Philosophy

The Radiation Control philosophy during this program was that work be completed with no biological assimilation of radioactive material and a minimal amount of external exposure. The radioactivity concentration guide for air was used as 2×10^{-12} μc Pu^{238}/cc.[1] The operational use of this figure was to require respiratory protection if it was anticipated that the air activity would exceed this value during the the exposure time rather than averaging over a 40-hour work week.

Two types of air samplers were used to determine the concentration levels in air -- the impactor grab sampler and stationary filter disc. The impactor samples were taken during specific work where a high contamination potential existed, while the filter discs were run continuously. In conjunction with the air sample program, routine smear surveys were also made of the entire area, as well as spot surveys as a part of each specific job survey. External exposures were to be kept below a Plant limit of 3 R/yr, with an administrative control of 50 mr/shift. One technician received 3345 mr due to an accidental overexposure.

Bio-assay samples were required when personnel were exposed to air concentrations above 100 x RCG with only an assault mask for respiratory protection or when gross personal contamination was detected. All surveys were by the Radiation Control Group with no self-monitoring being done by the Operating Group.

Early experience with the facility indicated that assault masks would be the minimum requirement during any break of containment or potential breaks of containment, such as removing bagged samples, transporting supplies into the cell, removing casks, removing product containers, and decontamination of product containers. During cell entries and some process incidents, air activity as high as 3000 x RCG (6×10^{-9} μc Pu^{238}/cc) was recorded.

(1) Report on Committee II on Permissible Dose for Internal Radiation (1959).

Dismantling of the Facility

Due to the known gross contamination inside of the containment boxes, the contamination was fixed by spray painting without knowledge of the actual contamination levels. The outer surfaces of the box were spray painted. Activity was airborne during the spraying operation. After the external surfaces were sprayed, the boxes were removed. The area had been well papered and curtained from the remainder of the cell complex and removal was accomplished without difficulty. The boxes were removed from the cell with a minimum of vibration and placed in a plastic lined, plywood box. Impactor samples indicated 50 x RCG during the removal of one box.

The decontamination inside the cells proceeded without major difficulty. Since stainless steel tubes with sharp points were sticking up from the floor, personnel entering the cell acted with deliberate caution.

Residual Effects on the Operation of the Caves Facility

As late as a year and a half after dismantling the facility, alpha air activity due to Pu^{238} has been observed in the loading area. The occurrence is sporadic, and analysis has confirmed that individual particles are responsible for the activity. The sources of this activity include:

- Cell 1 and 2 door mechanisms.
- Concrete trenches.
- Crane rails and roof structural supports.

A benefit derived from the operation of this facility was the training in contamination control techniques. Prior to the program, filter changes with filters contaminated to approximately 20 Rad/hr at 4 inches caused air activity and loose contamination problems. After the experience gained with high specific activity alpha work, filters with contamination levels in excess of 200 Rad/hr at 4 inches have been changed with no air activity or loose contamination.