

Development and Performance of Processes and Equipment to Recover Neptunium-237 and Plutonium-238

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Abstract

During the late 1950s, the Atomic Energy Commission (AEC) requested the Savannah River Site (SRS) to begin as soon as possible to produce plutonium-238 for use as a heat source for thermoelectric generators to power satellites. Thus, SRS joined the United States space race with the former Soviet Union, as well as continuing as a key facility in the race to produce materials for thermonuclear weapons.

To accomplish this new task, a massive interdisciplinary effort was required by all of the technical, engineering, maintenance, and administrative staff. New chemical and metallurgical processes were developed and installed in ingenious new equipment in the existing production buildings, with minimum interference with the processes operating to produce weapons materials.

Neptunium-237, the precursor isotope to the production of plutonium-238, was recovered from the existing processes, where it had been produced as a byproduct in the reactor irradiation of uranium. The purified neptunium-237 was fabricated into targets, which were neutron irradiated in SRS reactors to produce plutonium-238. The plutonium-238 was separated from neptunium and fission products and processed to plutonium oxide. The unconverted neptunium-237 was purified, processed to neptunium oxide, and used in additional reactor targets. Plutonium-238 oxide was fabricated into dense fuel forms at SRS and at other sites for use in thermoelectric generators.

Plutonium-238 produced at SRS has powered about two dozen spacecraft launched by NASA, including Transit, Viking, Voyager, Galileo, and Cassini.

Introduction

The Savannah River Site (SRS) began operation in the early 1950s to produce nuclear materials for weapons for the national security effort. The United States was in a weapons race with the former Soviet Union to produce thermonuclear weapons. There was great pressure on the staff of SRS to produce as rapidly as possible the tritium and weapons-grade plutonium (consisting predominantly of the fissionable plutonium-239 isotope) needed for nuclear weapons.

In 1957, the Soviet Union launched its first satellite, Sputnik, and began a race to space with the United States. In 1958, the United States launched its first satellite, Explorer I. In 1960, the Soviets put the first man into space. In

1961, President Kennedy started the National Aeronautics and Space Administration (NASA) program to put a man on the moon. In the late 1950s, the Atomic Energy Commission gave SRS the mission to produce a new reactor product, plutonium-238. This lighter isotope of plutonium, which had been made in only experimental quantities up to that time, was needed as a heat source for thermoelectric generators to power satellites. (Plutonium-238 has one less neutron per atom than its heavier isotope, plutonium-239.) Thus, SRS became an important part of the space race as well as continuing to be a key facility in the weapons race. The first plutonium-238 was made at SRS in 1961.

All of the work during these early years was conducted under strict rules of secrecy to

conceal technical details and production quantities. In fact, in the early program, secrecy was so tight that code names were used to conceal the identities of the isotopes neptunium-237 and plutonium-238.

Plutonium-238 has several characteristics that make it almost unique for fueling a radioisotope thermoelectric generator (RTG). It decays by emitting alpha particles with accompanying evolution of heat that can be readily converted to electricity. Gamma radiation from plutonium-238 is predominantly of low energy, requiring little shielding, which reduces the weight required for space applications. The relatively long half-life (about 88 years) makes it suitable for deep space missions, where sunlight is too weak to generate much electricity from solar cells.

Plutonium-238 fueled RTGs have provided power for about two dozen spacecraft launched by NASA, including:

1961	The first RTG delivered 2.7 watts of electrical power to a Navy Transit navigational satellite.
1969-72	Apollo astronauts left RTGs on the moon to power experiments.
1972	Pioneer spacecraft flew by Jupiter and Saturn.
1975	Viking spacecraft landed an instrumented vehicle powered by an RTG on the surface of Mars.
1977	Voyager 1 and 2 spacecraft flew past Jupiter and Saturn (three 150-watt electrical RTGs on each spacecraft).
1989	Galileo flyby of Jupiter (two RTGs, 285 watts electrical each) was launched. Exploration of Jupiter and its moons is underway.
1997	Cassini probe to explore Saturn (three RTGs, 285 watts electrical each) was launched.
1999	Galileo flew by Jupiter's innermost moon, Io.

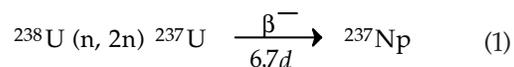
Over the next 10 years, NASA is planning several more missions that are expected to use plutonium-238 for electric power.

Several challenges had to be faced in the development of production technology for plutonium-238.

- Processes and equipment had to be developed that were compatible with the existing plant processes. Time was not available to construct new production buildings. The new processes would have to be incorporated with minimum interference into the facilities that were already operating to separate weapons-grade plutonium from uranium, and these new processes must not slow the production of weapons.
- There was no available supply of the precursor element, neptunium-237. It would have to be recovered and purified first before the production of plutonium-238 could begin.
- The radioactivity of plutonium-238 is about 250 times that of plutonium-239, so radiation damage to process materials, such as organic ion exchange resins, was expected to cause problems. In addition, there were concerns about the adequacy of containment of plutonium-238 in conventional, filtered glove boxes.

Technical Basis for Plutonium-238 Production

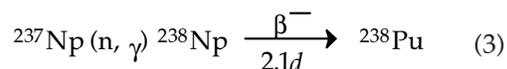
The precursor isotope, neptunium-237, is produced when uranium is irradiated in nuclear reactors. The nuclear physics reactions are the following (Groh 1970)



The reactions illustrate the production of neptunium-237 as a byproduct from the irradiation of either natural uranium, Equation (1), or uranium enriched in the uranium-235 isotope, Equation (2). This byproduct had been produced

at SRS since startup but had been lost to the radioactive waste streams during chemical processing of the irradiated fuel elements. It was necessary to develop modifications to the existing chemical separations processes to isolate the neptunium and to purify it from uranium, plutonium and fission products. In SRS reactors, neptunium production from enriched uranium fuels was the predominant route, particularly after the practice of recycling the irradiated enriched uranium was begun to conserve the intermediate isotope, uranium-236.

Once neptunium-237 is recovered and purified, it is converted to plutonium-238 by returning it to the reactor in target elements, where the reaction shown in Equation (3) takes place:



Short exposures of the neptunium targets in the reactors limit the production of the higher isotopes of plutonium. Typical SRS plutonium-238 product had an isotopic composition of approximately 81% plutonium-238, 15% plutonium-239, 2.9% plutonium-240, and lesser amounts of plutonium-241 and plutonium-242. Unavoidably, some of the neptunium and plutonium are converted to fission products during the irradiation, and these must be removed during the chemical processing.

In the late 1950s, when work was begun at SRS on processes to recover neptunium-237 and plutonium-238, a large body of information already existed in the literature on the chemistry of neptunium and plutonium. But it was necessary quickly to augment that data in some areas and to apply it to practical separations

processes that could be operated in the SRS plants. The byproduct neptunium was recovered from the solvent extraction processes operating in the two canyon buildings by a combination of modifications to the solvent extraction processes to divert the neptunium and ion exchange to recover and purify it. An anion exchange technology was chosen to process the recovered neptunium after it was re-irradiated to form plutonium-238. One of the most important chemical properties of neptunium and plutonium in solution is the ability of these elements to exist in several oxidation states or valences. This is an important tool for the separation of these elements where their behavior is controlled primarily by valence adjustment by oxidizing and reducing chemicals.

The varied oxidation states and ionic species of neptunium and plutonium that can exist under normal conditions are shown in Table 1. Uranium, by contrast, normally exists only in the VI oxidation state in aqueous solutions, as the UO_2^{2+} ion.

The solvent extraction processes are based upon the preferential extraction from nitrate solution of the actinide elements into tri-n-butyl phosphate (TBP) dissolved in dodecane to separate them from contaminating elements such as aluminum and the fission products. The order of extraction into TBP from nitrate solutions is: $\text{U(VI)} > \text{Np(VI)} \text{ Pu(IV)} > \text{Np(IV)} \text{ Pu(VI)} \gg \text{Np(V)}, \text{Pu(III)}$. Thus, U(VI) and Np(IV) can be separated readily from inextractable Pu(III); and U(VI) can be separated from Np(V). Aluminum and most of the fission products are inextractable in TBP. The solvent extraction

Table 1. Oxidation States of Neptunium and Plutonium

Oxidation State	III	IV	V	VI
Neptunium Ions		Np^{4+}	NpO_2^+	NpO_2^{2+}
Plutonium Ions	Pu^{3+}	Pu^{4+}	PuO_2^+	PuO_2^{2+}

process operated in the F-Area canyon building separated weapons-grade plutonium from natural uranium or depleted uranium (uranium from which the uranium-235 isotope had been partially removed). This was called the Purex Process. The H-Area Canyon process, called the HM Process, recovered enriched uranium from irradiated fuel elements.

The anion exchange process is based upon the fact that Np(IV) and Pu(IV) form anionic nitrate complexes of the type $\text{Np}(\text{NO}_3)_6^{2-}$ that are strongly absorbed from concentrated nitrate solutions by strong-base anion exchange resin, such as Dowex 1-X4. The other oxidation states of neptunium and plutonium are very weakly absorbed, as are most fission product species and common metallic cations. Thus Np(IV) and Pu(IV) can be effectively separated from uranium and fission products by anion exchange, and Np(IV) can be separated from Pu(III).

Description of Processes

Recovery of Neptunium-237 from Purex and HM Processes

In the Purex Process, one modification was needed to ensure that essentially all of the neptunium was directed to the high-level waste streams from which it was to be recovered. Nitrite was added to the extraction section of the first solvent extraction contactor in which uranium and plutonium are extracted into TBP to maintain the neptunium in the (V) oxidation state and thus minimize loss to other waste streams. (This contactor was originally a mixer-settler, but later was replaced by centrifugal contactors.) The high-level waste streams were subsequently combined and concentrated by evaporation in preparation to recover neptunium by anion exchange with new modular equipment installed in F Canyon. The concentrated waste was adjusted to 8M nitric acid and treated with ferrous sulfamate and hydrazine nitrate to produce Np(IV), which forms a strong anionic nitrate complex, and was fed to an agitated (stirred) anion exchange column to absorb the neptunium. Most of any plutonium-

239 that was present was also absorbed as it too forms a strong anionic nitrate complex. The agitated resin column, which will be described in a later section, was necessary to accommodate solids that are typically present in concentrated wastes. After the feed step, the column was washed with 8M nitric acid to remove impurities, and then the neptunium and plutonium were eluted with 0.35M nitric acid.

The solution from the first bed was processed further to separate neptunium and plutonium on a fixed bed of anion exchange resin as follows. Solution acidity was adjusted to 8M, and valence adjustment was repeated using ferrous sulfamate and hydrazine stabilizer, followed by heating to 50°C to oxidize Pu(III) to Pu(IV). The solution was fed to the resin bed, which absorbed the neptunium and plutonium. The bed was washed with 8M nitric acid to further remove impurities, and the plutonium was removed by washing with 5.5M nitric acid containing ferrous sulfamate and hydrazine to reduce plutonium to the (III) state. The recovered plutonium-239 was returned to the Purex second plutonium cycle. The neptunium was eluted with 0.35M nitric acid, processed by another cycle of anion exchange for further purification, followed by a cycle of cation exchange if necessary for thorium removal.

In the HM Process, operating conditions were modified to isolate and purify neptunium entirely by solvent extraction in the existing equipment. The dissolver feed was treated with ferrous sulfamate to maintain the Np(IV) valence state so that neptunium was extracted along with uranium in the 1A mixer-settler and partitioned from uranium in the 1B mixer-settler. Further decontamination of neptunium was accomplished using the Purex flowsheet in second plutonium cycle equipment that was not otherwise being used. The neptunium product solution was concentrated by evaporation in preparation for further processing in HB Line (a lightly shielded, glovebox facility in Building 221-H).

Isolation of neptunium from the HM Process continued for the life of the plutonium-238 program, but was discontinued from the Purex Process after an adequate inventory was accumulated.

The nitric acid solutions of neptunium from the Purex and the HM Processes were transferred to the HB Line for conversion to neptunium dioxide, NpO_2 , which could be fabricated into targets for neutron irradiation to produce plutonium-238. In HB Line, neptunium was further purified and concentrated, if needed, by anion exchange. After anion exchange, the concentration of nitric acid was adjusted within the range of 1-2M, and the valence of the neptunium was adjusted to the (IV) state with ascorbic acid and hydrazine inhibitor at about 50°C. Neptunium(IV) oxalate was precipitated at the same temperature by the addition of 1M oxalic acid solution. After digestion to aid crystal growth, the slurry was cooled to room temperature and filtered and air-dried. Neptunium dioxide was produced from the neptunium oxalate by heating in air to a final temperature of about 550°C (Porter 1964).

Recovery of Neptunium-237 and Plutonium-238 from Irradiated Targets

Processing of irradiated targets to recover plutonium-238 product and residual neptunium-237 was initially performed on a small scale in the High-Level Caves and B-Wing facilities of the Savannah River Laboratory. For the continuing larger scale operations, the very high radioactivity levels associated with the neptunium and the plutonium required installation of new plant equipment in the heavily shielded separations processing areas. Thus, unique modular units called "frames" (to be described later in this paper) were installed by novel techniques in H Canyon to process the irradiated targets. HB Line was also modified to provide facilities to convert the purified plutonium-238 product and the recovered neptunium-237 to the final oxide forms.

A schematic diagram of the neptunium target recovery process is shown in Figure 1. The irradiated NpO_2 -aluminum targets, containing unconverted neptunium-237, product plutonium-238, and fission products, were cooled at least 45 days to allow decay of short-lived fission products and then dissolved in boiling 10M nitric acid containing small amounts of mercuric ions and fluoride ions as catalysts. Dissolution was slow, requiring up to 48 hours for about 85% dissolution. Any undissolved target material was carried over and dissolved with the subsequent batch of targets. The dissolver solution contained small quantities of solids, primarily silica, which was removed by filtration prior to further processing.

The plutonium-238 product and residual neptunium-237 were separated from aluminum, fission products, other impurities, and from each other by anion exchange processing. The resin columns were normally operated as fixed beds, but could be operated as agitated beds when needed for regeneration or replacement of resin. In the first cycle of anion exchange, neptunium and plutonium were both adjusted to the (IV) valence state in 8M nitric acid by treatment with ferrous sulfamate and hydrazine, followed by heating to 50°C. The neptunium and plutonium were absorbed on the resin column to separate them from aluminum, fission products, and other cationic impurities. After washing with 8M nitric acid for decontamination, the neptunium and plutonium were eluted with 0.35M nitric acid. In the second cycle of anion exchange, the solution was adjusted to 8M nitric acid, and neptunium and plutonium were again both adjusted to the (IV) valence state and absorbed on the resin. After washing with 8M nitric acid for further decontamination, the plutonium was separated from the neptunium by washing the column with 5.5M nitric acid containing ferrous sulfamate and hydrazine to reduce the plutonium to the (III) valence state. The neptunium was then eluted with 0.35M nitric acid. The separated neptunium and plutonium were processed by one or more additional cycles of

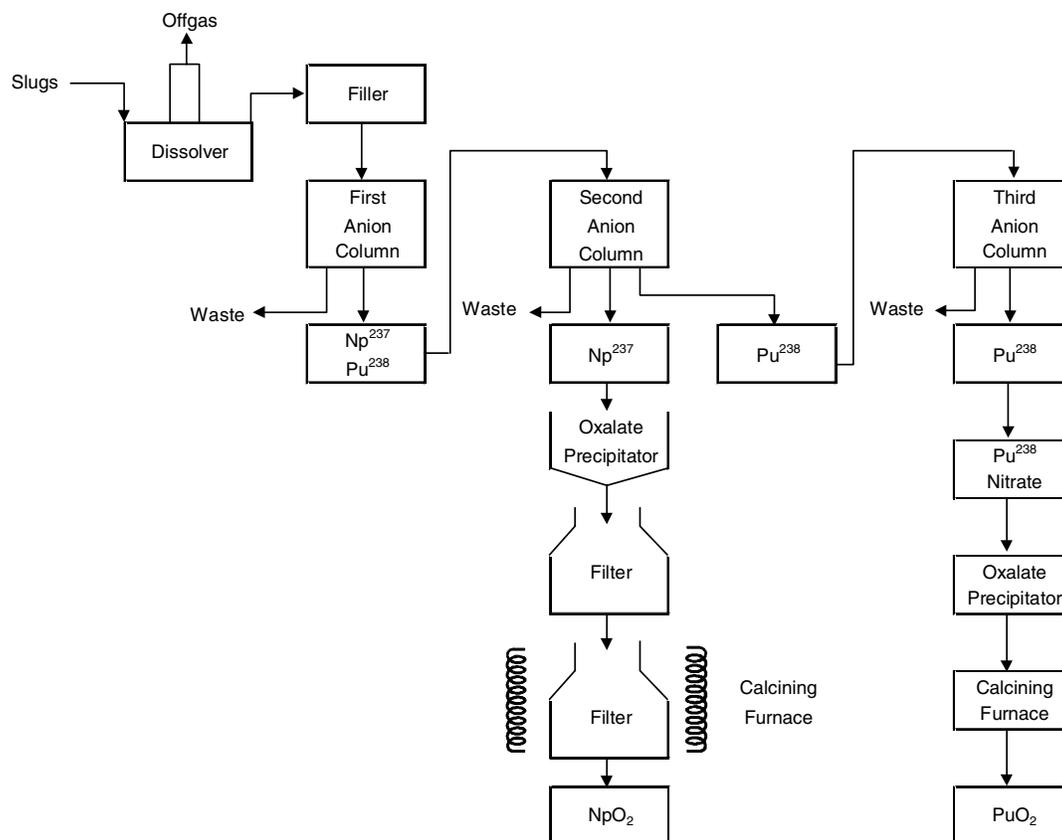


Figure 1. ^{238}Pu recovery from irradiated ^{237}Np at Savannah River

anion exchange as needed to obtain the desired purity. The purified neptunium solution was transferred to HB Line for further processing to produce NpO_2 for recycle. The purified plutonium solution was further processed as described below.

Conversion of Plutonium-238 to Oxide for Use in Heat Sources

In the early phases of the program, plutonium-238 was isolated as a dilute nitric acid solution that was shipped to Mound Laboratory in Miamisburg, Ohio, for additional processing and fabrication into heat sources. Later, to improve shipping safety, the plutonium was processed further in the HB Line to the solid oxide form either for shipment to another site or for onsite fabrication of heat sources.

In the HB Line, the plutonium solution from H Canyon was processed by anion exchange, if required, for decontamination and concentration. Then, the nitric acid concentration was adjusted within the range of 1-2M and plutonium valence was adjusted to (IV) by adding ascorbic acid and hydrazine nitrate and heating to 50°C . Plutonium(IV) oxalate was precipitated at the same temperature by the addition of 1M oxalic acid solution. After digestion to aid crystal growth, the slurry was cooled to room temperature and filtered and air-dried. Plutonium dioxide was produced by heating the plutonium oxalate in air to a final temperature of about 550°C .

An innovative technique was used at times in HB Line to reduce the rather high emission rate of neutrons from $^{238}\text{PuO}_2$. Neutron emission results from spontaneous fission of plutonium-

238 and also from (α,n) reactions with impurities and with the small quantities of ^{17}O and ^{18}O isotopes that occur in natural oxygen, which is mostly ^{16}O . Research at Mound Laboratory and at Savannah River Laboratory showed that the ^{17}O and ^{18}O content of $^{238}\text{PuO}_2$ can be reduced to very low values by isotopic exchange with ^{16}O . This can be readily accomplished by heating the plutonium oxide in the presence of oxygen gas or water vapor that is highly depleted in ^{17}O and ^{18}O . In HB Line, this was implemented by flowing isotopically pure $^{16}\text{O}_2$ over the oxide at a temperature of a few hundred degrees centigrade. Neutron emission rates were reduced to near the value expected from spontaneous fission and impurities, and resulted in lower radiation exposure of personnel.

Some of the $^{238}\text{PuO}_2$ produced at SRP was shipped to other sites for production of heat sources, but a large number of heat sources was also produced onsite in a new facility installed in 1978 in Building 235-F. D. T. Rankin describes the production of heat sources at SRP in another paper in this symposium proceedings.

Radiation Effects of Plutonium-238

In the chemical processes, the high radiation level of plutonium-238 was dealt with in several ways. Perhaps the most serious problem was radiation damage to the organic ion exchange resins. This was controlled by choosing resins, such as Permutit SK, that were more resistant to radiation, by limiting the exposure times of the resin to plutonium-238, by agitating the resin beds between cycles to redistribute the resin particles, by maintaining continuous flow of process solution through resin beds loaded with plutonium-238, and by periodic remote replacement of the resin. In solutions, radiolysis products from plutonium-238 alpha particles interfered with the stabilization of the desired oxidation states of plutonium and neptunium. This was controlled by the addition of reducing agents and stabilizers such as hydrazine. A combination of ferrous sulfamate and hydrazine proved to be most effective for maintaining the

lower oxidation states of neptunium and plutonium while minimizing undesirable side effects such as gas formation.

Conventional gloveboxes provided adequate containment of plutonium-238 in the final steps of processing. The boxes were constructed of stainless steel and operated at subatmospheric pressure with several stages of high efficiency filtration for the box offgas. Local shielding for gamma and neutron radiation, including leaded gloves, protected the workers from radiation exposure.

Description of Production Equipment

Unitized Frame Concept

The recovery of neptunium-237 from the Purex and HM processes and the recovery of plutonium-238 from irradiated neptunium targets involve gamma radiation levels that are equivalent to those encountered in processing spent nuclear fuels; therefore, these processes were installed within the shielded space of the Savannah River canyons. The term canyon refers to the heavily shielded buildings (Buildings 221-F and 221-H) that house and supply services to process equipment for recovering plutonium and uranium from spent reactor fuels.

The canyons at Savannah River provide a module 10 feet square by 17 feet high for each vessel having standard services (i.e., inlet and outlet piping, steam, cooling water, electricity, sampling, and instruments). Each building section has four modules, isolated by a low curb to contain liquid spills or leaks, and has a sump and permanently installed transfer jet to move spilled liquids or leakage to a rework system. Transfers between vessels in the canyon are made on a pipe rack in which the pipe is remotely removable. A direct jumper can make transfers between adjacent vessels.

As discussed above, ion exchange was selected as a key process for recovery, separation, and decontamination of neptunium-237 and plutonium-238. The ion exchange vessels and flows of solutions to and from them are much smaller than those used in the normal solvent extraction processes for which the canyons were designed. Ion exchange processes had never before been installed and operated in a facility that was remotely maintained. Since experimental work performed by the Savannah River Laboratory showed that it would be necessary, from time to time, to replace the ion exchange resins in the canyon process vessels, a technique was developed to transfer them to and from the canyon as slurry (Bebbington 1990).

The recovery of neptunium from depleted uranium required installation of four ion exchange columns in F Canyon. The recovery of neptunium-237 and plutonium-238 from irradiated neptunium targets required five ion exchange columns and a dissolver in H Canyon (Poe 1964). Unitized frame design was adopted for these units to minimize canyon space requirements. The frame concept means installing a number of small equipment pieces in a steel frame that is installed and removed by the normal canyon crane. The specialized equipment was designed by adapting the small-scale equipment to remote operation in the canyon. After building a complete prototype unit to test the feasibility of the concept, three units were constructed and used many years in the canyons. Figure 2 is a photograph of one these frames. A fourth unit was installed in F Canyon for initial recovery of neptunium. This unit consisted of a single ion exchange column and its support tankage.

Placing several equipment pieces in a frame in a single canyon module is economical of canyon space but places a heavy burden on the services available in that module. Pipe and electrical connections were made to the frame rather than to individual pieces of equipment. Services were piped to the equipment as part of the permanent frame structure. This efficiently used existing services, but additional services

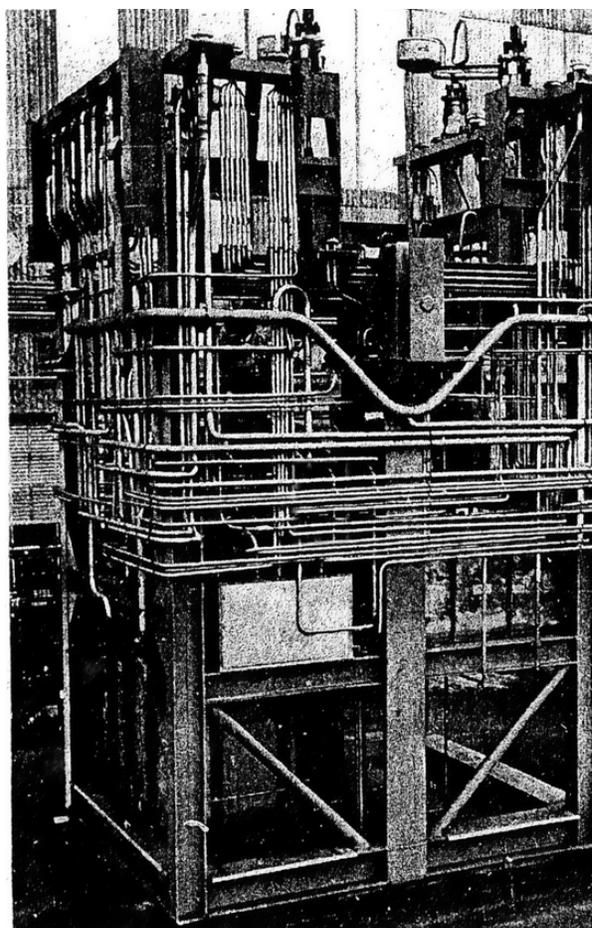


Figure 2. Np and ²³⁸Pu frame (Complete frame without ion exchange columns. Frame as installed remotely with Canyon crane.)

were required, primarily for pneumatic liquid level and specific gravity instrumentation for several vessels in each frame. Bundles of up to 6 stainless steel tubes (1/4-inch o.d.) were drawn through the 3-inch pipes embedded in the canyon shielding walls. Demonstration of this concept had great utility in adapting existing facilities to alternate processes. No significant problems were encountered in installing these lines, and their useful life has been equivalent to the life of embedded piping.

Ion Exchange Columns

The ion exchange columns that were built, tested, and used in these operations were either

12- or 24-inch diameter and normally contained 25 or 100 liters of resin. They normally were used as conventional settled bed ion exchange columns but had the capability of use as agitated ion exchange bed columns. One unit was used as an agitated bed column. All operations, including resin replacement, were performed remotely without mechanical valves. Resin was charged and discharged as free-flowing slurry in a solution having a specific gravity close to that of the resin. Two weirs determine flow paths; air pressure is applied to either or both

to direct flow properly for feed, wash, elution, or resin removal. Figure 3 shows an isometric diagram of one such ion exchange column.

Dowex 1 and Permutit SK anion exchange resins were used. Dowex 1 resin provides higher decontamination from fission products, but Permutit SK resin is more stable under alpha radiation. Dowex 1-X4 (4% nominal cross linkage) was used in 40 to 60 mesh range particle size. Selected use of smaller particle resins, some with cross linkages as small as 2%,

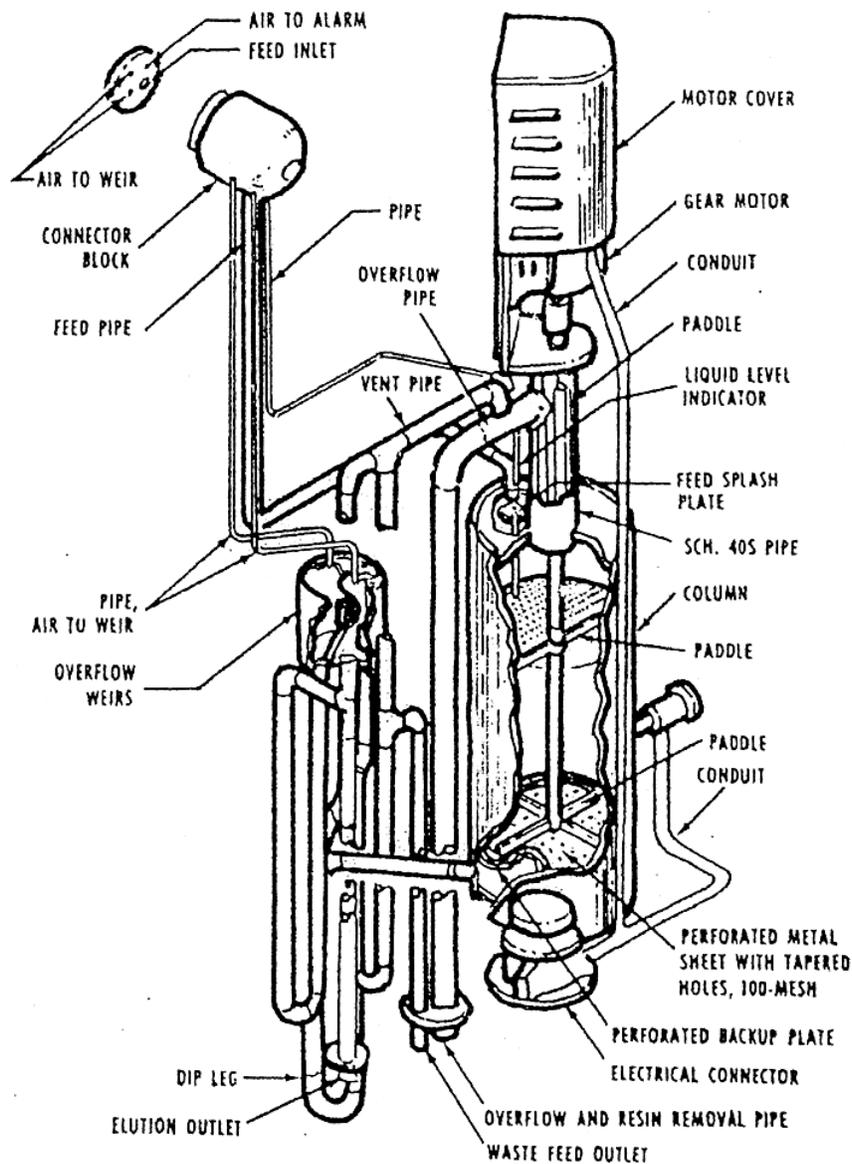


Figure 3. Ion exchange column (for installation on frame shown in Figure 2)

was used where fast kinetic absorption was required, such as in the ion exchange column used for separation of plutonium-238 from neptunium-237. Solution flow through the resin was determined by the resin particle size and gas generated from alpha radiolysis.

Performance of Production Processes

The losses of either neptunium-237 or plutonium-238 in the H-Canyon frame operation used for irradiated neptunium targets averaged 2.7% for the three neptunium processing columns and 4.9% for the four plutonium processing columns. This material was discarded to High-Level Waste for the first two years of operation, and then the frame waste was diverted to a new frame waste column and a further 95 % of this was recovered. High fission product decontamination was accomplished; the limiting fission products were the radionuclides zirconium and niobium. This process gave a decontamination factor (curies in the feed divided by curies in the product) of 100,000 for neptunium and 1,800,000 for plutonium. The second ion column was used to separate the plutonium from the neptunium; this was one of the most crucial activities in the target processing because cross-contamination of one product left in the other product would be lost to waste. Using the Dowex 1-X2 resins, removal of 97 to 99% of the plutonium from the neptunium could be achieved. (This ~1 to ~3% of the plutonium lost to the neptunium is included in the 4.9% waste loss described above.)

HB-Line Facilities for Conversion of Neptunium and Plutonium to Oxides

Decontaminated neptunium-237 and plutonium-238 products from the H-Canyon Frames were converted to NpO_2 and PuO_2 in HB Line, after an initial period of liquid shipments of plutonium-238 nitrate to Mound Laboratory. Initially these operations were performed in facilities designed for weapons plutonium finishing. These facilities had been made obso-

lete when H-Canyon operations were changed from Purex (weapons-grade plutonium) to processing highly enriched uranium in the late 1950s. Neptunium-237 and plutonium-238 finishing operations were performed in HB Line gloveboxes. The plutonium-238 operations were performed in a new small glovebox line, and the neptunium was finished in the old B-Line cabinets. After several years of operation in this manner, it was decided to construct a new HB Line on top of Building 221-H designed for these operations. The new line had three subparts: one for neptunium, one for plutonium-238, and the third for recovery of neptunium and plutonium from scrap. This new facility was located over Sections 2 through 6 of Building 221-H. It consists of the new fifth and sixth level of that building.

The new neptunium oxide line was constructed over Sections 4 and 5 of the 221-H Canyon. The oxide line consists of two glovebox lines constructed with adjacent operating and maintenance rooms to minimize spread of contamination. The line decontaminated and converted neptunium nitrate to neptunium oxide. Processes employed include ion exchange, precipitation, calcination to oxide, and packaging of the neptunium for shipment to Building 235-F for fabrication into targets.

The plutonium-238 oxide process was constructed over Section 6 of the canyon. The process is an improved version of the original plutonium-238 oxide line. The plutonium nitrate solution is converted to plutonium oxide powder by the oxalate precipitation followed by calcination. The cabinets associated with this line were equipped with significant external neutron and gamma shielding to protect the operating staff.

The scrap recovery module consists of two parallel lines of gloveboxes located over Sections 2 and 3 of the canyon. They provide the capability to introduce different types of scrap materials, sorting, dissolution, filtration, and transferring solutions to the canyon for recovery of the actinides and the associated support

capabilities. The cabinet lines are equipped with water jackets and lead shielding over the gloves and lead glass on the windows. This equipment is provided to shield the operating staff from the gamma radiation.

Fabrication of Neptunium Targets in 235-F Building

Neptunium oxide from HB Line finishing operations was blended in Building 235-F with aluminum powder and fabricated into targets for irradiation in SRP reactors. Target fabrication was initiated in 1961 and consisted of a compacted blend of oxide and aluminum clad in an aluminum can. "Green" compacts, 3 inches long and 0.86 inches in diameter, were formed by pressing the blended powder in a tool-steel die at 19.8 tons per square inch (tsi) at ambient temperatures. These compacts were 90 to 92% of theoretical density. A double-acting press was used since it transmits equal force to both ends of the compact to give more uniform compaction. Complete densification during green compact fabrication was undesirable

since some travel of the compact surface relative to the can wall during hot pressing is needed to provide a fresh metallic surface for bonding. Two compacts were loaded into an impact-extruded aluminum can, which was then closed by an aluminum cap. The assembly was loaded into an Inconel-X die, which was placed in a vacuum furnace consisting of a floating Inconel-X back-up die inside a stainless steel sheath wrapped with resistance heaters. This configuration is shown in Figure 4. The slug was heated to 600 to 620°C under a vacuum of <1,000 microns of mercury and then pressed at about 19 tsi to form a metallic bond between the compacts and the can wall and about 99% of theoretical density. All of these operations were performed in glove boxes with contained atmosphere to protect the workers. These target slugs were irradiated between 1961 and 1965 to produce plutonium-238.

In later development, as more neptunium became available for irradiation, the design of the targets was changed from target slugs to tubular targets, which improved heat transfer

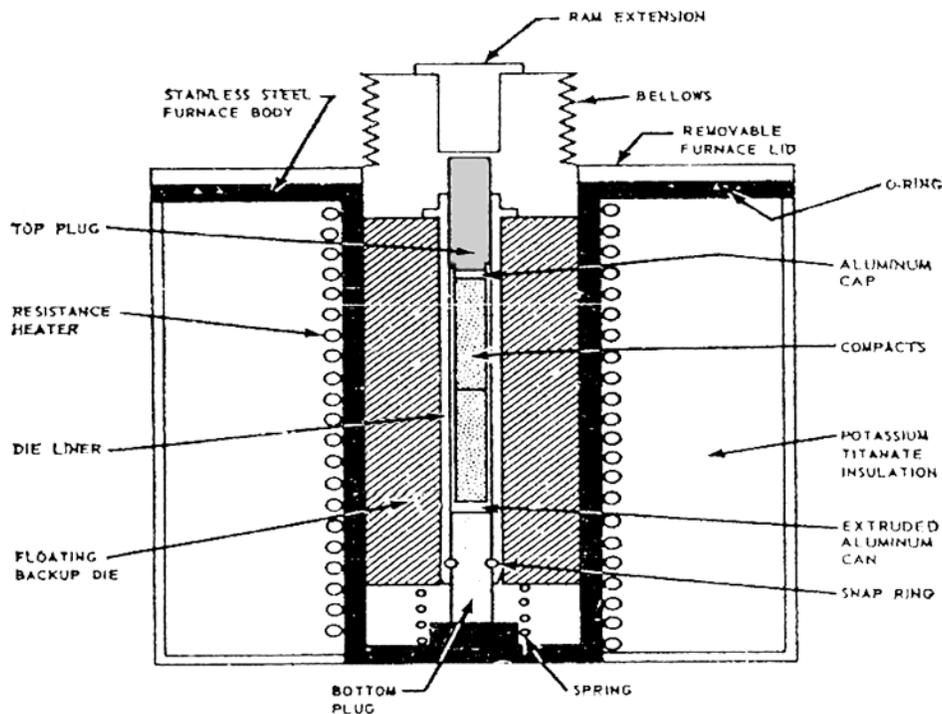


Figure 4. Hot-press-bond furnace for fabrication of Np target slugs

and increased plutonium-238 production and purity by allowing operation at higher neutron fluxes. Between 1966 and 1978, the design of these tubular elements changed three times. Billets containing the neptunium target were fabricated in Building 235-F, and the tubes were extruded in Building 321-M. Billets consisted of cylindrical inner and outer sleeves welded to the bottom fitting. Compacts were placed in the space between the two sleeves, and a top fitting with a breather tube was welded in place. This created a contamination shield, which allowed the billet to be removed from containment cabinets in Building 235-F and transported to Building 321-M for extrusion. The initial tubular targets were constructed with cylindrical compacts used in the target slugs, which on extrusion yielded tubes with ribbed target cores. Later compacts were redesigned into trapezoidal compacts to provide tubular targets with uniform neptunium core thickness. The different target design used differing amounts of neptunium so that the final tubular targets varied from 120 and 190 grams of neptunium per foot of active tube length, and the tube diameter varied between 3 and 3.7 inches. The initial cladding thickness was 0.065 inch and later was decreased to 0.040 inch as fabrication and irradiation experience showed these targets were safe for irradiation. Fabrication of neptunium target was continued until the SRS reactors were shut down in the early 1990s.

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Biographies

Harold J. Groh

Harold J. Groh has 47 years experience in nuclear programs. Employed at Savannah River Site from 1952 through April 1990 in professional and management positions, he has broad familiarity with DOE nuclear weapons complex and SRS capabilities, with particular technical expertise in nuclear fuel processing, waste management, and plutonium technology. He worked ten years as an independent consultant in numerous DOE programs including surplus weapons plutonium disposition, spent fuel management, chemical safety reviews, and high-level waste management. Mr. Groh has a Ph.D. in physical chemistry from the University of Rochester, and a B.S. in chemistry from St. Louis University.

W. Lee Poe, Jr.

W. Lee Poe, Jr. has more than 48 years of experience in providing technical and management support for large-scale nuclear projects including chemical processing, waste management, environmental protection. Employed by Du Pont Company and WSRC from 1951 through 1989, his experience includes operation of chemical reprocessing of irradiated spent nuclear fuels (F and H Canyon Buildings), purification and finishing of ²³⁹Pu, ²³⁸Pu, ²³⁷Np, and U (depleted, natural, and enriched) (in FB

and HB Lines and A Lines in both areas), target fabrication, heavy water production, and site and DOE complex planning. Process safety and operational assessments, environmental protection, safety and health protection, and a thorough understanding of the processes used in all areas of the SRS are an integral part of this experience. Specific responsibilities for ^{237}Np and ^{238}Pu processing were technical support within the various plant units for design, testing, start-up, and operation of all of the processes described in this paper. His experience also includes ten years consulting with DOE and private contractors associated with DOE nuclear programs in a wide range of areas. During this time, he has participated in public forums on waste management activities; risk management alternatives, comparisons, and ranking; environmental remediation; etc.

John A. Porter

John A. Porter has 42 years experience in research, development, and production in the defense nuclear industry. Employed by DuPont Company and Westinghouse Savannah River Company from May 1957 through July 1989 in professional and management positions at the Savannah River Site, his experience includes operation of chemical processing plants for

irradiated fuels and targets (F and H Areas separations plants, ^{239}Pu metal production line, ^{238}Pu oxide production line, plutonium storage facility, plutonium recovery facilities, tritium facilities); operation of waste management facilities and analytical control laboratories supporting processing plants; manager of industrial hygiene and radiation protection functions; coordinator of environmental protection programs; and conduct/management of research and development activities relating to the above. With ten years independent consulting in DOE nuclear programs, his specialty areas include plutonium (and other actinides) processing, waste management, conduct of operations, personnel safety, and environmental protection. Mr. Porter has a Ph.D. in physical chemistry from Vanderbilt University and a BS in chemistry from Clemson University.

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