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**An Evaluation of Neptunium Operations at Savannah River Site**

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## PUBLICATION RECORD

| EFFECTIVE DATE | REVISION NUMBER | DESCRIPTION |
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| 09/19/2016 | 00 | New report for methods and models for reconstructing internal radiation doses to workers at the Savannah River Site who were potentially exposed to $^{237}$Np based on site records. Incorporates formal internal and NIOSH review comments. Training required: As determined by the Objective Manager. Initiated by James M. Mahathy and Billy P. Smith. |

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## ACRONYMS AND ABBREVIATIONS

| | |
|---|---|
| AEC | U.S. Atomic Energy Commission |
| Ci | curie |
| CFR | Code of Federal Regulations |
| cm | centimeter |
| CTW | construction trade worker |
| D&D | decontamination and decommissioning |
| DOE | U.S. Department of Energy |
| dpm | disintegrations per minute |
| ft | feet |
| g | gram |
| HAW | high-activity waste |
| HPRED | Savannah River Site Health Protection Radiation Exposure Database |
| hr | hour |
| in. | inch |
| kg | kilogram |
| L | liter |
| M | molar |
| mCi | millicurie |
| mg | milligram |
| min | minute |
| mm | millimeter |
| mo | month |
| mR | milliroentgen |
| mrem | millirem |
| n | neutron |
| NIM | nuclear incident monitor |
| NIOSH | National Institute for Occupational Safety and Health |
| NMMSS | Nuclear Materials Management and Safeguards System |
| NOCTS | NIOSH-Division of Compensation Analysis and Support Claims Tracking System |
| ORAU | Oak Ridge Associated Universities |
| ORNL | Oak Ridge National Laboratory |
| ppm | parts per million |
| PUFF | Plutonium Fuel Form Fabrication Facility |
| PUREX | plutonium-uranium extraction |
| R | roentgen |
| R&D | research and development |
| RCG | radioactivity concentration guide |

| | |
|---|---|
| SHI | Special Hazard Incident |
| SRDB Ref ID | Site Research Database Reference Identification (number) |
| SRL | Savannah River Laboratory |
| SRS | Savannah River Site |
| WBC | whole-body count |
| wt% | weight percent |
| yr | year |
| γ | gamma particle |
| µCi | microcurie |
| °C | degrees Celsius |

## 1.0 INTRODUCTION

Since reactor start up, $^{237}$ Np has been produced at Savannah River Site (SRS) as a byproduct, but it was not removed from early radioactive waste streams during chemical processing of irradiated fuel elements. Neptunium-237 was produced when depleted uranium (containing $^{238}$ U) or enriched uranium (containing $^{235}$ U) was irradiated in one of the nuclear reactors (Driggers and St. John 1957). Neptunium targets were also irradiated for production of $^{238}$ Pu. In this report, the Oak Ridge Associated Universities (ORAU) Team has provided (1) the history of neptunium recovery, fabrication, and other uses at SRS along with a detailed timeline; (2) information on the radiological design of production processes; (3) information on the radiation monitoring of processes and workers; and (4) methods and models for reconstructing internal radiation doses to workers who were potentially exposed to $^{237}$ Np.

## 2.0 HISTORY OF NEPTUNIUM RECOVERY

By 1957, with the beginning of the Space Race between the United States and the Soviet Union, the U.S. Atomic Energy Commission (AEC) developed a serious interest in using $^{237}$ Np to produce $^{238}$ Pu. Plutonium-238 was thought to be useful as a heat source for thermoelectric generators for powering satellites. SRS and other AEC sites began studying how $^{237}$ Np could be recovered from reactor waste flows (Siddall 1957a; Colven 1957). SRS studied the behavior of neptunium in H-Area (Siddall 1957b; DuPont 1957a). In 1958, the AEC tasked SRS to produce $^{238}$ Pu by irradiating $^{237}$ Np, provoking the following reaction:

$$^{237}\text{Np}\ (n,\gamma)\ ^{238}\text{Np} \rightarrow\ ^{238}\text{Pu} \qquad (2\text{-}1)$$

An evaluation of the feasibility of interim separation of $^{238}$Pu from irradiated $^{237}$ Np resulted in a preliminary process outline, a high spot-cost estimate, and a review of the effects on other programs (DuPont 1958a). To use neptunium as a target for production of $^{238}$ Pu, methods were needed to recover neptunium from waste streams of irradiated depleted uranium and irradiated enriched uranium, and to separate and purify both neptunium and $^{238}$ Pu from irradiated neptunium (Poe, Joyce, and Martens 1963; DuPont 1962, 1963a). Tests of the anion exchange method for recovery of neptunium from high-activity waste (HAW) concentrate were performed at the TNX facility using thorium as a surrogate.

SRS designed a neptunium slug and bean pilot fabrication by October 1958. Of the 13 steps, 11 were to be performed in a glovebox. Fabrication was performed in Buildings 235-F and 773-A in October 1958 in very small-scale demonstrations. The slugs were irradiated at the Materials Testing Reactor and the K Reactor. In November 1958, a 1/1,000-scale agitated-bed anion exchange resin system was set up in the laboratory. The process was satisfactorily demonstrated in the laboratory using one irradiated neptunium oxide slug (DuPont 1958a). The recovery of irradiated neptunium was tested in a high-level cave at Building 773-A (Burney and Karraker 1958; DuPont 1958a). To develop a procedure for preparing neptunium oxalate, experiments were conducted using 100 mg (-0.7 µCi) of "Oak Ridge" neptunium that contained small amounts of plutonium and iron (Pollock and Walz 1958). Testing continued in Building 773-A from January through April 1959. Staff training on the recovery procedure was performed at that time. By July, 70 g of 237 Np had been recovered from dilute solutions using the test set-up in Building 773-A (DuPont 1959a). The workflow for producing $^{238}$ Pu was developed using some of that recovered neptunium.

As discussed in the following sections, SRS recovered neptunium using different waste streams through 1984 and in the H-Canyon starting in the 2000s (DuPont 1980a; Fuller, Smith, and Goergen 2013; Taylor and Phifer 2012). Between these dates, neptunium in reactor waste streams was sent to waste tanks. A diagram of the movement of neptunium material being processed at SRS is shown in Figure 2-1.

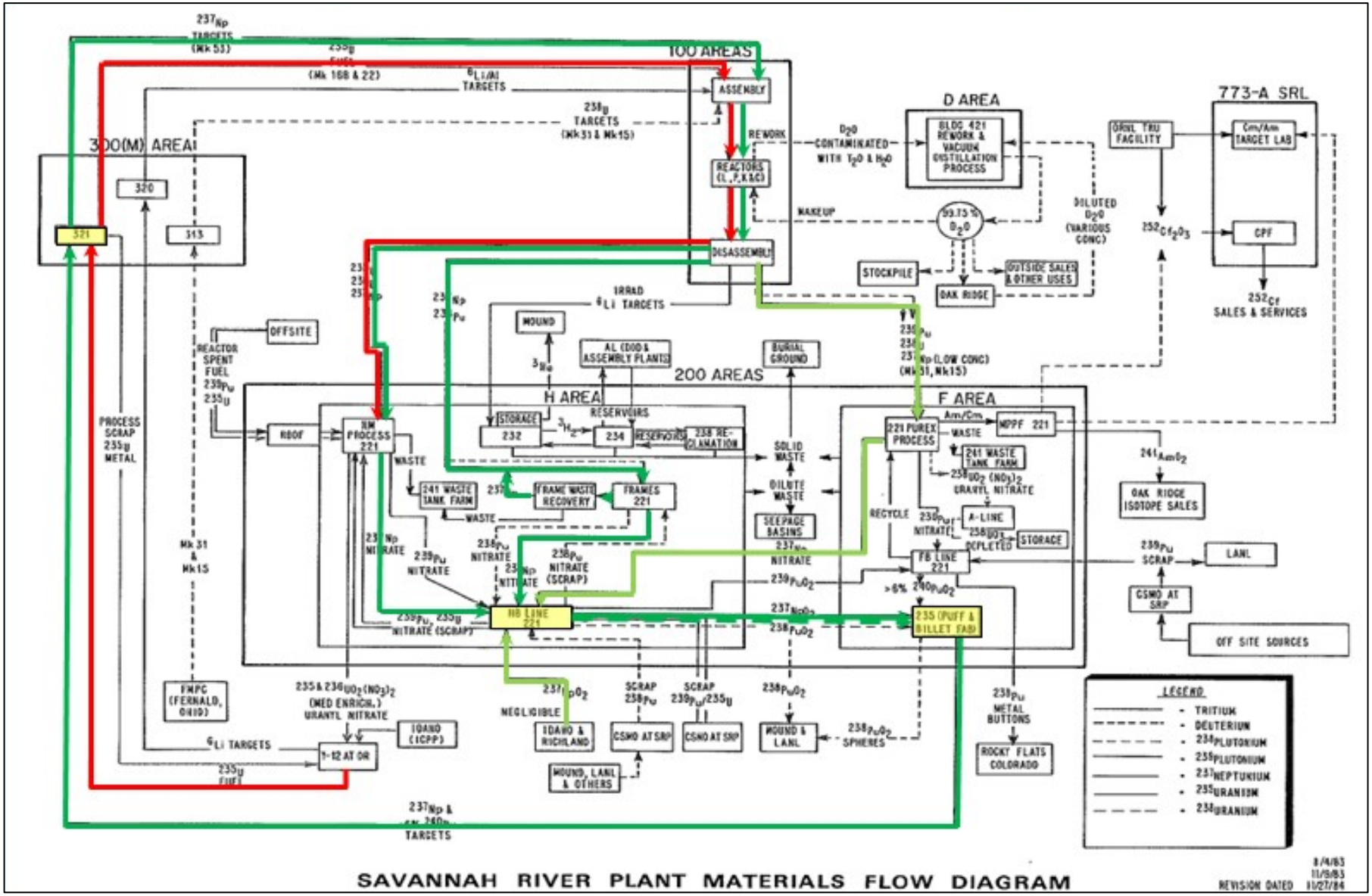



Figure 2-1. Neptunium material flow (DuPont 1984a).

## 2.1 RECOVERY FROM IRRADIATED URANIUM

Experimental equipment for testing the feasibility of neptunium recovery from HAW was designed and ready for use by July 1958. In August 1958, small-scale experimental equipment to establish the feasibility of neptunium recovery from acidic HAW was installed in the H-Canyon. The first test in H-Canyon was performed in September 1958. Testing revealed that, with one modification to the plutonium-uranium (PUREX) process, nearly all neptunium in the irradiated depleted uranium feed appeared in the 1AW high-level waste from which it was recovered by ion exchange. Wastes from PUREX streams and from the neptunium purification steps were combined and concentrated and therefore represent essentially all of the neptunium present in the PUREX plant feed (Poe, Joyce, and Martens 1963). High-level waste streams containing neptunium were concentrated by evaporation in preparation for neptunium recovery via anion exchange using new modular equipment installed in F-Canyon. Recovered $^{239}Pu$ was returned to the second plutonium cycle (Poe, Joyce, and Martens 1963; WSRC 2000).

A diagram of the PUREX process is provided in Figure 2-2. Production recovery of neptunium using the modified PUREX process was performed in the F-Canyon (WSRC 2000). Small modular units called “frames” were used in the canyons for the neptunium separation. Once an adequate neptunium inventory was accumulated by 1965, isolation and recovery of neptunium from the PUREX process was discontinued (WSRC 2000).

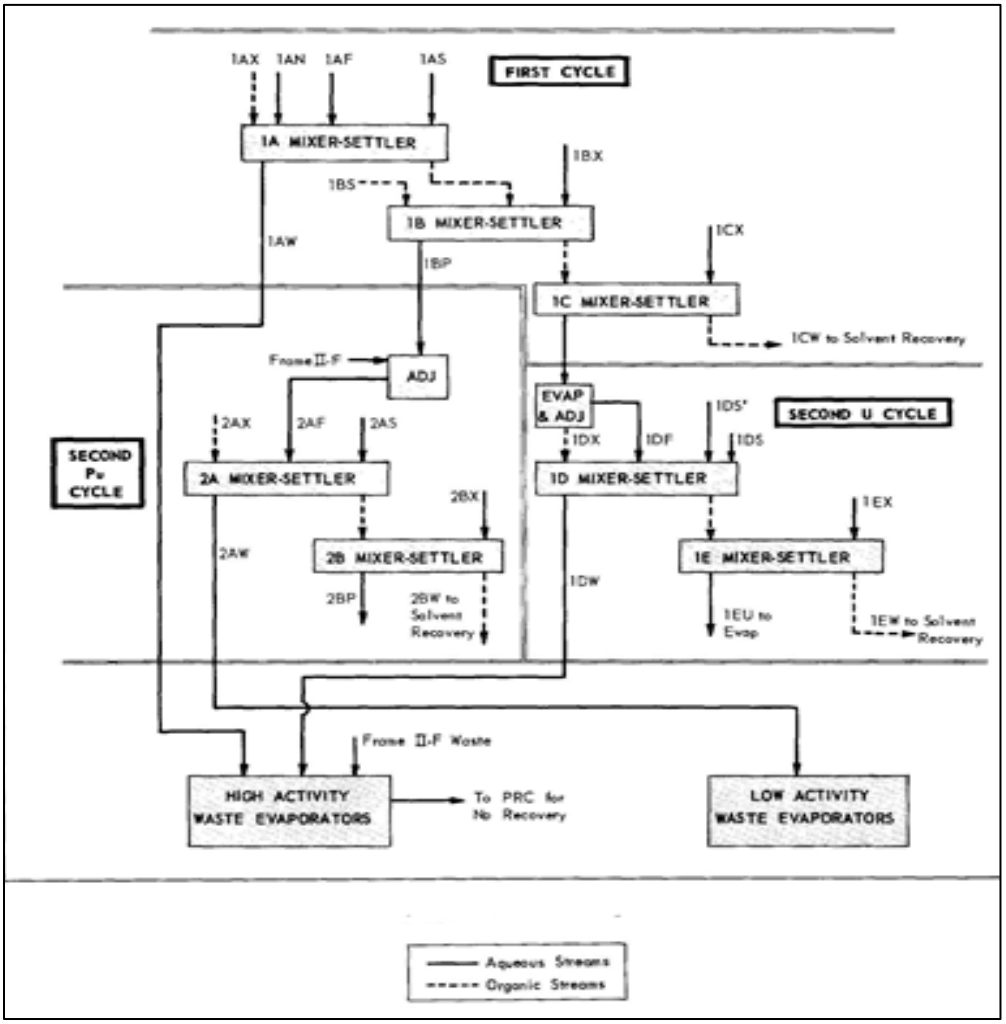



Figure 2-2. Modified PUREX process for recovery of neptunium source (Poe, Joyce, and Martens 1963).

Representative results for recovery in January 1963 for the modified PUREX process are shown in Figure 2-3 (DuPont 1963a). Less than 1% of the neptunium processed in the 221-HB Line came from the PUREX process in F-Canyon (Epperson 1996).

| Column Feed | | | Raffinate | | | Recycle* | | | Eluate of Stage 1 | | | | % Recovery | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Run No.** | Bed Volumes | Rate (lb/min) | Np, g | Pu, g | U, lb | Np, g | Pu, g | U, lb | Np, g | Pu, g | U, lb | Gamma, curies | Np | Pu |
| 2FSC-1 | 24 | 16 | 2.0 | 0.7 | 0.6 | - | - | - | - | - | - | - | - | - |
| -2 | 26 | 22 | 2.5 | 0.8 | 1.3 | - | - | - | - | - | - | - | - | - |
| -3† | 24 | 14 | 2.5 | 1.2 | 0.4 | - | - | - | 62 | 29 | - | - | 89.0 | 91.5 |
| -4 | 16 | 22 | 5.0 | 0.8 | 1.9 | - | - | - | - | - | - | - | - | - |
| 2FSH-1†† | 22 | 26 | 2.3 | 1.9 | 34.4 | 1.9 | 8.0 | 3.5 | 29.4 | 169 | 0.028 | 75 | 80.2 | 98.6 |
| -2 | 17 | 20 | <1.8 | 2.0 | 17.8 | 6.8 | 17.8 | 5.2 | 57.2 | 276 | 0:015 | 63 | >96.6 | 99.3 |
| -3 | 15 | 18 | 3.4 | 6.2 | 22.8 | § | § | § | 67.0 | 286 | 0.013 | 185 | >95.3 | 97.7 |
| -4 | 19 | 21 | 4.8 | 77.5 | 12.5 | 18.1 | 22.8 | 1.9 | 76.2 | 192 | 0.003 | 202 | 93.8 | 71.0 |

* Recycle consists of eluate from stages 2 and 3, most of the decontamination wash of stage 1, and the displacement volume from elution of stage 1. Decontamination washes were omitted on the SCRUP runs (8M acid containing hydrazine and ferrous sulfamate was fed at about 2 lb/min to keep the products fixed on the bed while the next batch of feed was accumulating).

** "FSC" run numbers refer to SCRUP runs and "FSH" run numbers are runs on SRP feed.

† Runs 2FSC-1, -2, and -3 were eluted as a triple batch.

†† Runs 2FSC-4 and 2FSH-1 were eluted as a double batch.

§ Recycle stream was not analyzed.

Figure 2-3. Neptunium recovered from PUREX waste, January 1963 (DuPont 1963a, p. 140).

SRS used a modified PUREX method (known as the HM Process) to process radioactive waste streams from enriched uranium irradiation. The HM Process was also modified (1) to recover neptunium by isolating most of the neptunium in a new 1BP (1BP Aqueous Product from 1B Bank in First Cycle Solvent Extraction) stream, and (2) to reduce the loss to the 1AW stream by adjusting the 1AF stream with additional ferrous sulfamate. The flowsheet for recovering neptunium from enriched uranium is shown in Figure 2-4. The 1B mixer-settler was placed in service to partition the neptunium into a uranium-free 1BP stream. Production recovery of neptunium from enriched uranium was performed in the H-Canyon. The neptunium product solution was concentrated by evaporation in preparation for further processing in HB Line in Building 221-H (Poe, Joyce, and Martens 1963; WSRC 2000). On limited occasions, SRS shipped neptunium it recovered from the enriched stream to the Hanford site (Tetzlaff 1960a) and the Oak Ridge National Laboratory for further recovery (DuPont 1961a, 1963a; Tetzlaff 1960b).

## 2.2 RECOVERY FROM IRRADIATED NEPTUNIUM TARGETS

Irradiated neptunium oxide targets (originally in a slug design but later changed to a tube) contained unconverted $^{237}$Np, reactor product $^{238}$Pu, and fission products. Due to the fission product activity, irradiated targets were cooled at least 45 days to allow decay of the short-lived fission products. Dissolution of targets required up to 48 hours and then only about 85% dissolution was obtained. The remaining dissolved target material was carried over and dissolved with the subsequent batches of targets. Approximately 8 to 10 kg/mo of neptunium were recovered from dissolved irradiated targets during the duration of the process (WSRC 1996ESH-HPT-96-0117). There was significant plutonium contamination.

Anion exchange was used to separate aluminum, remaining fission products, and other impurities from the $^{238}$Pu product and residual $^{237}$Np. Plutonium and neptunium were absorbed by the resin

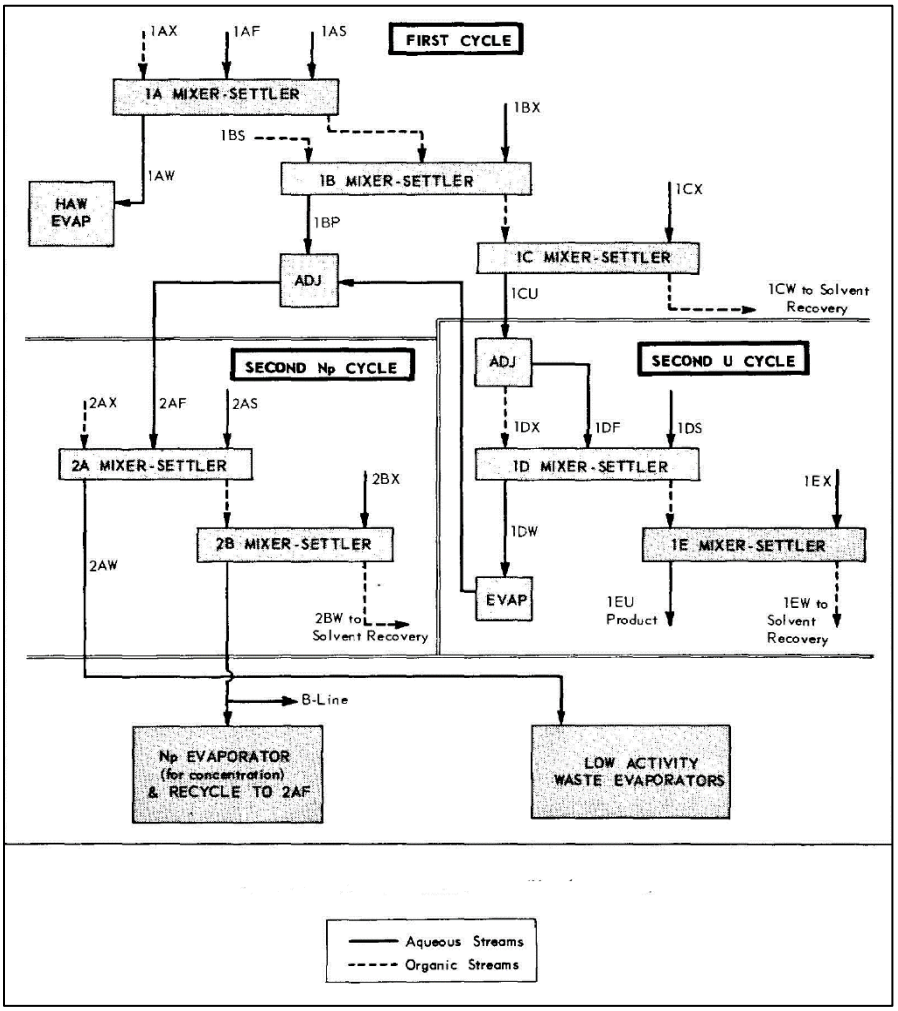



Figure 2-4. Process for neptunium recovery from enriched uranium (Poe, Joyce, and Martens 1963).

column to separate them from aluminum, fission products, and other cationic impurities. Plutonium was separated from the neptunium by washing the column with nitric acid containing ferrous sulfamate and hydrazine to reduce the plutonium to the (III) valence state. The separated neptunium and plutonium were processed by one or more additional cycles of anion exchange, as needed, to obtain the required purity of $^{238}Pu$ and $^{237}$ Np (WSRC 2000). The specification for $^{238}Pu$ was a minimum of 2 g/L (Poe, Joyce, and Martens 1963).

Processing of irradiated neptunium targets was initially performed on a small scale in the high-level caves at Building 773-A. Due to very high radioactivity levels associated with the neptunium and plutonium, novel techniques of equipment installation were employed in the heavily shielded separations processing areas, specifically in H-Canyon where irradiated targets were processed. A frame assembly similar to the ones used in F-Canyon was installed in H-Canyon for neptunium recovery from irradiated targets. The flowchart for the recovery of neptunium and plutonium from irradiated neptunium targets is shown in Figure 2-5. The HB Line was modified to provide facilities for converting purified $^{238}Pu$ product and the recovered $^{237}$ Np to the final oxide forms.

## 2.3 CONVERSION TO NEPTUNIUM OXIDE

Nitric acid solutions of recovered neptunium were transferred to the HB Line, a lightly shielded, glovebox facility in Building 221-H. These solutions were converted to neptunium oxide ($NpO_2$) to be

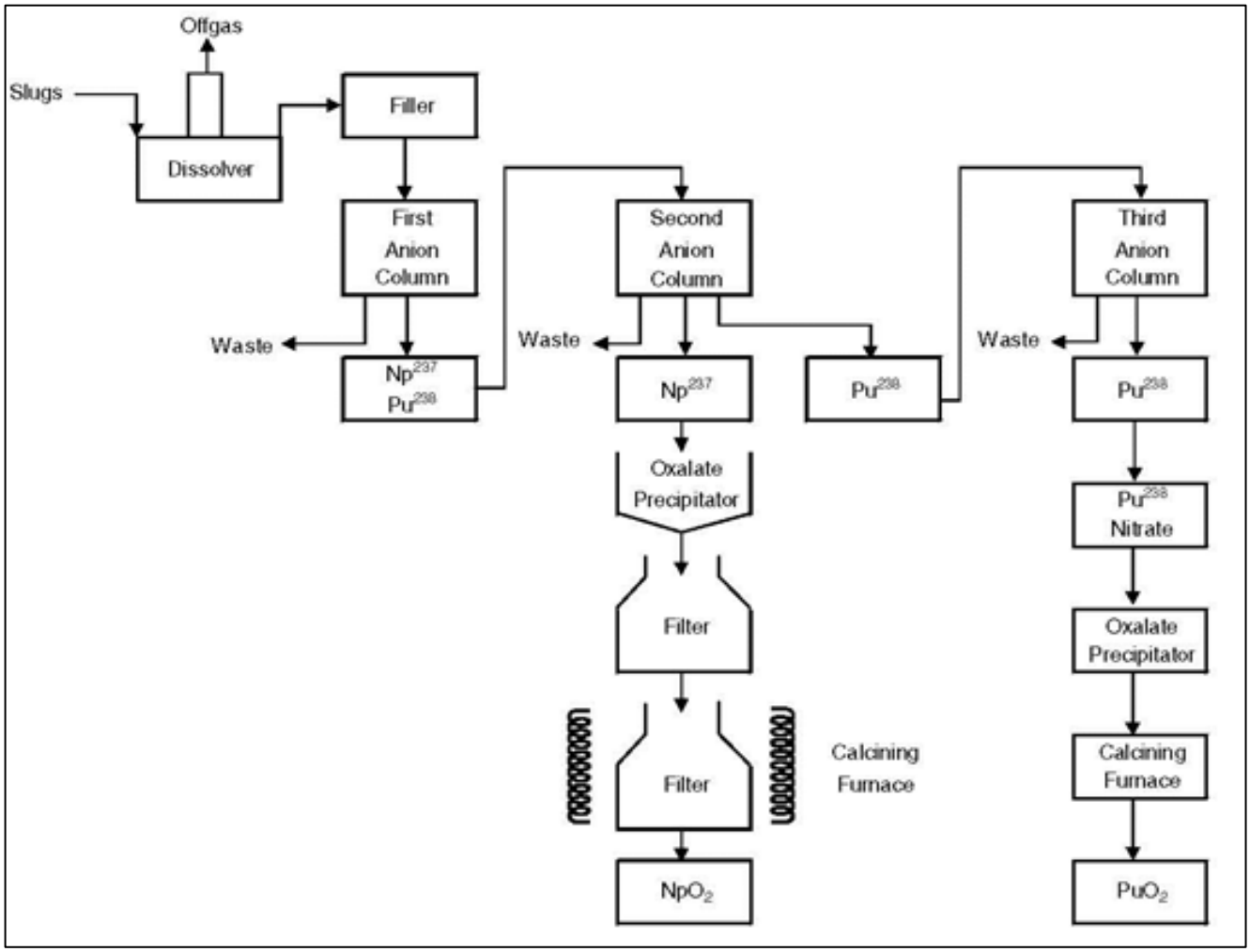



Figure 2-5. Process for recovery of neptunium from irradiated targets (Groh, Poe, and Porter 2000, p. 181).

fabricated into targets for neutron irradiation to produce $^{238}$ Pu. Neptunium received directly from Hanford in roughly 1-kg quantities was also processed in the HB Line in 1967 through 1972 (Kelley 1969). Neptunium was further purified and concentrated in the HB Line, if needed, by anion exchange (WSRC 2000).

The scrap recovery module consisted of two parallel lines of gloveboxes located over Section 2 and Section 3 of the canyon. They provided the capability for introducing different types of scrap materials and for the sorting, dissolution, filtration, and transfer of solutions to the canyon for actinide recovery and associated support capabilities. These lines were equipped with water jackets and lead shielding over the gloves along with leaded glass on the windows to shield the operating staff from gamma radiation (WSRC 2000).

In the HB Line, neptunium was finished in the old B-Line cabinets. A new HB Line facility was constructed in the early 1980s on top of Building 221-H. The new line had three subparts: one for neptunium, the second for $^{238}$Pu, and the third for neptunium and plutonium recovery from scrap. This new facility was located over Sections 2 through 6 of Building 221-H on the fifth and sixth levels. A new neptunium oxide line was constructed over Sections 4 and 5 of the 221-H-Canyon. The oxide line consisted of two glovebox lines constructed with adjacent operating and maintenance rooms to minimize spread of contamination. Neptunium nitrate was decontaminated and converted to $NpO_2$

using the same technique described in the preceding paragraph (Fuller, Smith, and Goergen 2013). However, the new HB Line was not used to process $^{237}$Np until 2004 (Koenig 2013).

## 3.0 FABRICATION OF NEPTUNIUM TARGETS

After finishing in HB Line, neptunium oxide was transferred to Building 235-F to be blended with aluminum powder and fabricated into irradiation targets for SRS reactors. Neptunium oxide shipments to Building 235-F would typically be in batches of about 1 kg each with total shipment weights ranging between 2 and 25 kg (DuPont 1977a, 1977b, 1977c, 1977d). Batches of neptunium oxide from HB Line were contaminated with $^{238}$Pu. Representative assays by $^{238}$Pu weight percent are provided in Table 3-1.

Table 3-1. Plutonium contamination from HB Line.

| $NpO_2$ (kg) | Minimum Pu wt% | Average Pu wt% | Maximum Pu wt% | Reference |
|---|---|---|---|---|
| 21.8 | <0.05 | 0.16 | 0.6 | DuPont 1974a |
| 4.84 | 0.18 | 0.36 | 0.53 | DuPont 1974b |
| 12.65 | 0.02 | 0.18 | 0.42 | DuPont 1974c |
| 5.25 | 0.01 | 0.03 | 0.06 | DuPont 1974d |
| 2.25 | 0.25 | 0.28 | 0.32 | DuPont 1974e |

Such plutonium contamination is important to note because as the plutonium-to-neptunium ratio increases the plutonium becomes the alpha emitter of radiological concern as demonstrated in Table 3-2 assuming specific activities for $^{238}$Pu and $^{237}$Np of 17.1 Ci/g and 0.00069 Ci/g, respectively.

Table 3-2. Alpha ratio.

| Np wt% | Pu wt% | Pu:Np alpha ratio |
|---|---|---|
| 99.0 | 1.0 | 250:1 |
| 99.5 | 0.5 | 125:1 |
| 99.9 | 0.1 | 25:1 |
| 99.95 | 0.05 | 12:1 |
| 99.99 | 0.01 | 2.5:1 |
| 99.995 | 0.005 | 1.2:1 |
| 99.999 | 0.001 | 0.25:1 |

The first target, in the form of a canned slug, was fabricated at SRS in 1961. The slug consisted of a compacted blend of neptunium oxide and clad in an aluminum can. Groh, Poe, and Porter (2000) reported:

> *"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 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.*

Figure 3-1 compares the sizes of the neptunium operations (one room) with plutonium fabrication operations (Rooms 7, 8, 9, 11 and 13). Note the neptunium line, which is below Vault 100, is divided. The top part was the maintenance side while the bottom was the operations side. The two sides were separated by walls. A photograph of the production side of the neptunium billet line showing production operators and supervisors is shown in Figure 3-2. A view of the maintenance side of the line is shown in Figure 3-3.

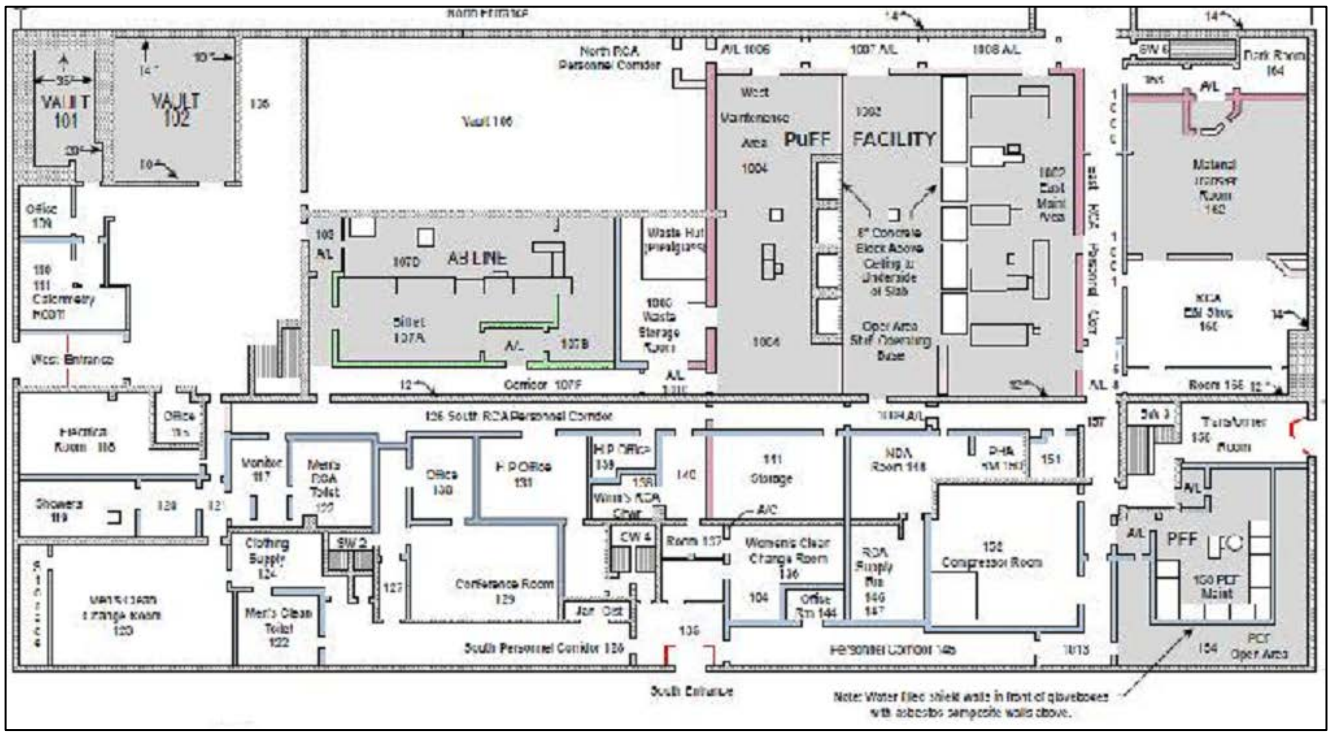



Figure 3-1. Building 235-F layout (Taylor and Phifer 2012).

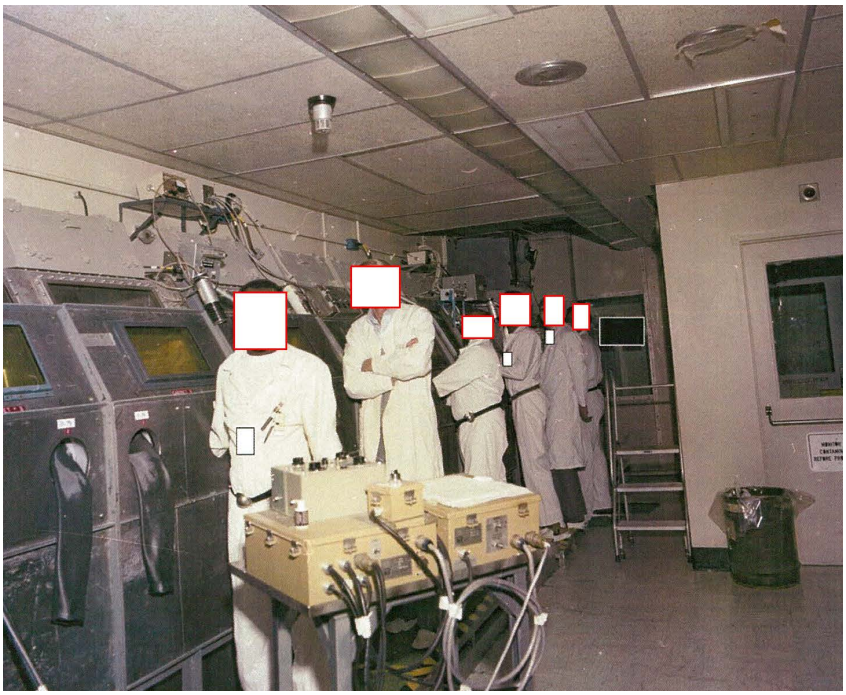


Figure 3-2. Workers on the neptunium billet line, Building 235-F (DuPont ca. 1975a).

In Building 235-F, two compacts were loaded into an impact-extruded aluminum can and capped. These components made up a slug. The slug was then loaded into an industrial die that was placed in a vacuum furnace. This furnace, enclosed in a glovebox line, consisted of a floating back-up die inside a stainless-steel sheath wrapped with resistance heaters (Figure 3-4).

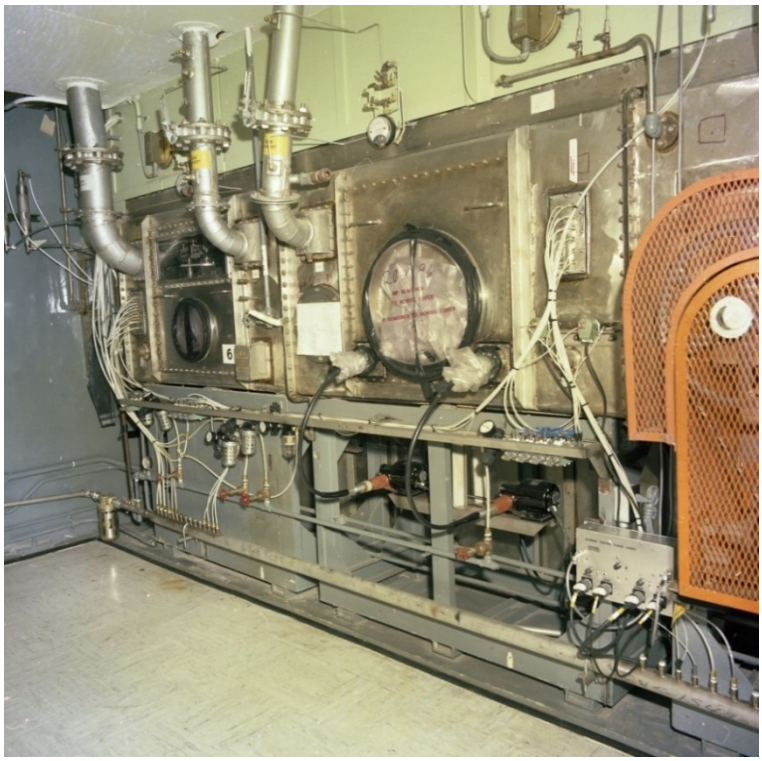


Figure 3-3. Maintenance side of the neptunium billet line, Building 235-F (DuPont ca. 1975b).

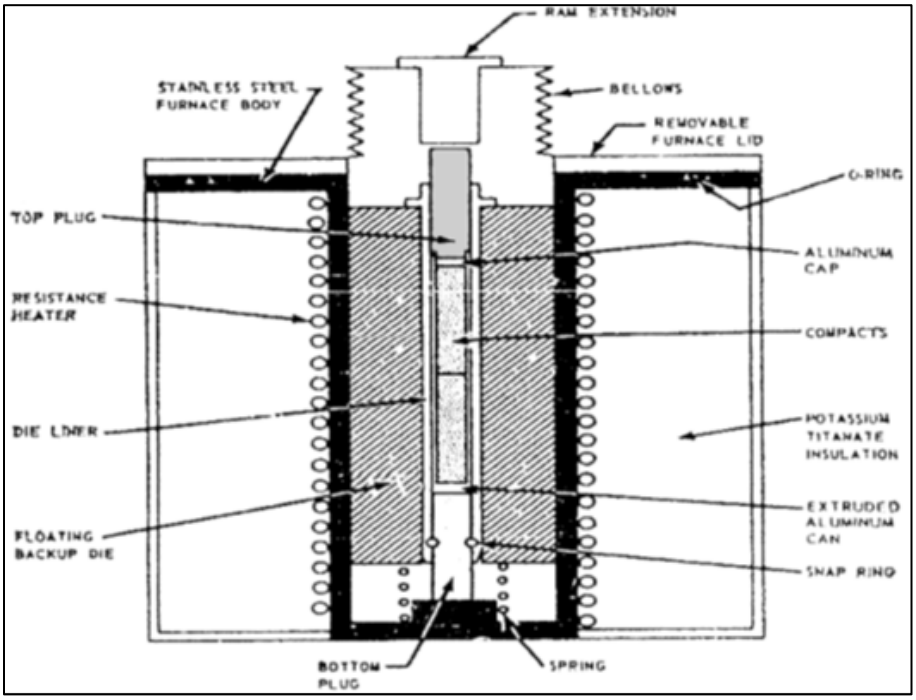



Figure 3-4. Hot-press-bond furnace for fabrication of neptunium target slugs (WSRC 2000).

After hot pressing, slugs (with the flash intact) were cleaned in a hot caustic solution to remove any residual surface lubricant, then rinsed in cold water. Slugs were dipped in nitric acid to neutralize caustic agents and to remove surface smut left by the caustic etch, then given a final rinse in cold water. The entire procedure was repeated in a second set of baths to remove residual surface

contamination. Slugs were dried before the flash was trimmed. Slugs were trimmed to remove the flash and to prepare the top end for a weld. A gauging dimple was pressed into the top end caps during hot pressing so slugs could be trimmed and rewelded several times, if necessary. The welds served two purposes: they offered an inspection step for the soundness of the top end cap and also served as a final closure on the slug (Hill 1961). All of these operations were performed in gloveboxes with contained atmospheres for worker protection. The target slugs were irradiated between 1961 and 1965 to produce $^{238}$Pu; the first $^{238}$Pu was produced at SRS in 1961 (WSRC 2000).

In the mid-1960s, the design of the neptunium target was changed to unalloyed tubular elements as more neptunium became available for irradiation (DuPont 1965a; WSRC 2000). The extrusion process for neptunium-aluminum (Np-Al) tubes was developed in Building 773-A in 1966 (DuPont 1967a). First, prototype elements containing thorium oxide were substituted for neptunium oxide and were fabricated with dimensions of the Mark VI-N element (DuPont 1984b). The tubular design improved heat transfer, which increased production and purity of the $^{238}$Pu by allowing operation at higher neutron fluxes. The exact designs of the tubular element were modified three times between 1966 and 1978. The target designs used differing amounts of neptunium so that the final tubular targets varied from 120 to 190 g of neptunium per foot of active tube length, and the tube diameter varied between 3 and 3.7 in.

Starting by 1968, billets were also fabricated into alloyed tubes in Building 235-F. Billets consisted of cylindrical inner and outer sleeves welded to the bottom fitting. To create a contamination shield, billets were placed in the space between the two sleeves. A top fitting with a breather tube was welded in place. Starting in 1966, the use of the contamination shield allowed the billet to be removed from containment cabinets in Building 235-F and transported to Building 321-M for outgassing and extrusion (DuPont 1960a, 1961b, 1961c, 1962, 1966a, 1968a, 1969a, 1969b,c, 1971a, 1975a, 1976a, 1976b, 1977e, 1977f, 1978a; Reed 1982; Roggenkamp 1987). Before extrusion, the billet assemblies were subjected to off-gassing in ovens. After extrusion, tubes would undergo fluoroscopic examination in 321-M using the fuel distribution analyzer and be re-extruded, if necessary (Osmond et al. 1984; DuPont 1977b, 1977c, 1977d, 1977g, 1977h; Reed and Swanson 2006). At times, neptunium fuel tubes underwent metallurgical testing in gloveboxes in Building 322-M (DuPont 1969d, 1974f); at other times, testing was performed in 773-A (DuPont 1974f). Fuel-density measurements were made with the fuel density analyzer in 321-M, and with the nuclear test gauge in 305-M (DuPont 1970b, 1974b; Reed and Swanson 2006). While not a normal part of the neptunium process, some of neptunium oxide tubes were sawed in 321-M (DuPont 1975a).

When neptunium was extruded in Building 321-M, the extrusion portion of the building was shut off from normal access and workers dressed in protective clothing (Reed and Swanson 2006). Billets were heated and stretched into reactor targets. Although this was a physical process, no machining such as cutting or grinding was done. If the billet maintained its integrity throughout the extrusion, no contamination would be present. However, because the Machining and Casting Area of 321-M was a known contamination area at SRS, it is plausible to assume that some failures occurred during the extrusion of materials, including those containing neptunium. Uranium and neptunium billets were processed with the same equipment and, in comparison with uranium, a relatively small amount of neptunium was processed through 321-M. Therefore, it is reasonable to assume that any neptunium contamination is not isolated from the uranium contamination. According to the Waste Information Tracking System database, in 321-M, the activity fraction for $^{237}$Np was less than 0.1 %; no $^{238}$Pu was listed (Epperson 1996).

The number of neptunium tubes extruded was only a small fraction of the tubes extruded in Building 321-M. Table 3-3 compares the number of neptunium, depleted uranium, and enriched uranium tubes extruded from 1968 through 1975. Figure 3-5 shows a photo of the front of the extrusion press.

Table 3-3. Comparison of tubes extruded in Building 321-M (Reed and Swanson 2006).

| Year | Neptunium tubes | Depleted uranium tubes | Enriched uranium tubes |
|---|---|---|---|
| 1968 | 28 | 67,739 | 11,944 |
| 1969 | 300 | 67,951 | 12,538 |
| 1970 | 0 | 144,631 | 17,735 |
| 1971 | 78 | 66,483 | 7,318 |
| 1972 | 87 | 119,170 | 5,433 |
| 1973 | 0 | 177,468 | No data available |
| 1974 | 66 | 96,344 | 3,422 |
| 1975 | 103 | 83,202 | 3,457 |


Figure 3-5. Front entrance of extrusion press, Building 321-M (Bebbington 1990).

Fabrication of neptunium targets was suspended from October 1979 through April 1980 while SRS used the neptunium billet line and related processes to make plutonium targets for the $^{242}Pu$ program (DuPont 1980b, 1980c). After being restarted, fabrication of neptunium targets was continued until 1984 (DuPont 1980a; Rogers et al. 1989; Fuller, Smith, and Goergen 2013; Jordan, Watkins, and Hensel 2009). Twelve MK 53A billets (high $NpO_2$ content) were extruded in December 1980 for testing neptunium loading (DuPont 1980a).

Figures 3-6 shows two photos. On the left, a billet is forced into the extrusion die. On the right, a billet as it emerges from the press.

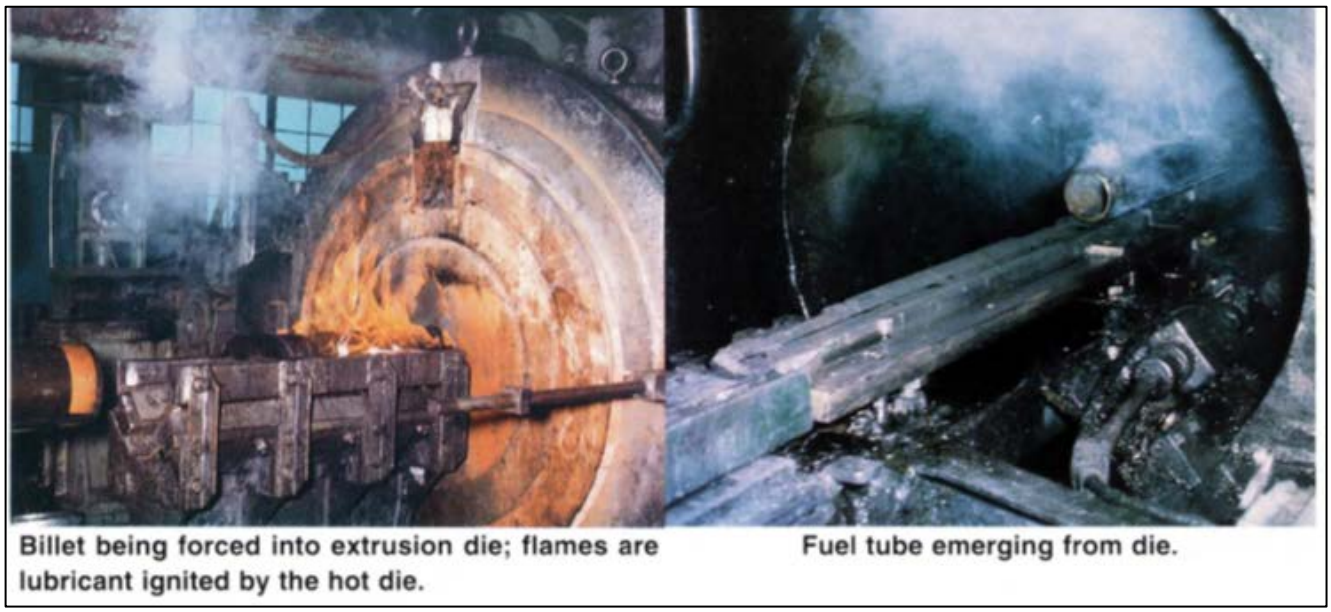



Figure 3-6. Front entrance and rear exit of extrusion press, Building 321-M (Bebbington 1990).

Seven of the SRS fuel element designs included $^{237}$Np for irradiation:

- Mark VII-A and VI-J (both briefly in 1961),
- Mark VI-N,
- Mark 52,
- Mark 53,
- Mark 53A (some),
- Mark 53B, and
- Mark 61 (DuPont 1961c, 1984b).

In 1987, SRS reported a $^{237}$Np inventory of about 400 kg with the capability to produce about 80 kg/yr (Roggenkamp 1987); however, active operations for separating and fabricating neptunium had been stopped in 1984 (Fuller, Smith, and Goergen 2013).

## 4.0 NEPTUNIUM OPERATIONS AFTER 1984

With the shutdown of neptunium and $^{238}$Pu separation and fabrication in 1984, the equipment used in H-Canyon was decontaminated, although other separations continued in H-Canyon until 1992. Once decontamination was completed, SRS conducted a project to upgrade the H-Canyon frames process equipment but ceased before completion. Most of the H-Canyon frames process services, instrumentation, and support equipment were upgraded, but the actual canyon frame modules were not installed. As of 2013, this equipment was still stored as spare equipment and is available for installation. The Building 235-F facilities were left in “enhanced production readiness” status from 1985 through 1990 with maintenance activities being performed by just three operators. Varying levels of maintenance activity were carried out between 1985 and 1990, primarily on the building ventilation system (DOE 1991). Neptunium was stored in 235-F through 1998.

Building 321-M was removed from operation in 1992 (Reed and Swanson 2006). In 1995, it was decommissioned, and decontamination was started in 2000 (Dewberry 2004a). During decontamination operations, $^{237}$Np hold-up contamination was measured in the outgassing ovens. SRS staff indicated the appearance at the measured level of $^{237}$Np was not inconsistent with the designated solid waste stream for the facility, but the measurements were verified (Reed and Swanson 2006) because furnace-heating of $^{237}$Np was not performed in 321-M (Dewberry 2004b).

However, outgassing of neptunium fuel was conducted in 321-M (Reed and Swanson 2006). A subsequent survey found low-level $^{237}$Np contamination on two saw bench components. The saw benches were used to cut process components, primarily U-Al product fuel tubes (Dewberry 2004c), but had also been used to cut neptunium tubes (DuPont 1975a).

Nine surplus $^{237}$Np targets were being stored in the Building 321-M vault in the 1990s, and 6,100 L of solution containing $^{237}$Np were being stored in H-Canyon holding tanks through 2003 (Brock and Craig undated; Watkins, Hensel, and Jordan 2010). The latter material represented the neptunium recovered and purified in the 1980s. The nine targets were moved to K-Area for storage until they were processed in H-Canyon for recovery of neptunium by 2004 and converted to oxide in the HB Line (Koenig 2013). Between 2004 and 2008, neptunium from the H-Canyon holding tanks was also separated in the H-Canyon and converted to an oxide in the HB Line. Fifty cans of neptunium oxide were produced and shipped to Idaho National Laboratory (Rudisill 2002; Jordan, Watkins, and Hensel 2009). Additional small quantities of $^{237}$Np resulted from H-Canyon closure operations with the processing of irradiated uranium fuel (Walker et al. 2003). These solutions were also converted to oxide. These oxide materials were shipped to Idaho National Laboratory and Oak Ridge National Laboratory in the approved type 9975 shipping containers (Watkins, Hensel, and Jordan 2010).

## 5.0 ANCILLARY WORK

From the start of operations through 2007, SRS has performed laboratory analysis, experiments and tests, fuel prototype fabrication, and test irradiations using $^{237}$Np. These activities were conducted in several buildings including 773-A, 777-M, 305-M, CMX, TNX, and 772-F. Several of these operations were performed routinely throughout the course of reactor operation; others were research and development (R&D) tasks. An overview of work items, locations, and years of operation by building are provided later in Table 5-1. Some highlights of these items include:

- Experimental equipment for testing the feasibility of recovering neptunium from HAW in the canyons (Building 773-A, 1958) (DuPont 1958a).
- Testing of the use of agitated beds of anion exchange resins (Building 773-A, 1958) (DuPont 1958a).
- Routine analyses for neptunium in process streams (Building 772-F, 1959) (DuPont 1959b).
- Evaluation of mechanical stirrers (Building 772-F, 1960) (DuPont 1960a).
- Laboratory-scale testing of neptunium separations (Building 773-A, 1961) (DuPont 1961b).
- Laboratory development and completion of a process for recovering about 90% of the neptunium from samples of stored plant wastes (Building 773-A, 1961) (DuPont 1961d).
- Measurements on 105 neptunium oxide slugs and 3,531 Mark V-B slugs (Building 773-A, 1963) (DuPont 1963a).
- Tests in the Process Development Pile to determine the reactivities and power ratios for assemblies containing tubular neptunium target elements (Building 777-M, 1965) (DuPont 1965b).
- Neptunium oxide slugs formed and fabricated into billets (Building 773-A, 1968) (Moyer 1969a).

- Requests by Separations Technology for laboratory assistance in studying possible causes of neptunium losses in the 2B bank of the second neptunium cycle (Building 772-F, 1969) (DuPont 1969d).

- Flow test of new tubular $NpO_2$ assembly (CMX, 1969) (DuPont 1969e).

- Development techniques for fabricating aluminum-clad $^{237}Np$ target tubes with cores of mixed Mg-$NpO_2$ powders (Building 773-A, 1970) (DuPont 1970b).

- Testing of techniques for improving the purity of $^{238}Pu$ from $^{237}Np$ (Building 773-A, 1971) (DuPont 1971b).

- Completion of pressurized and unpressurized anion exchange tests with macroporous 15% cross-linked resin (Building 773-A, 1972) (DuPont 1972a).

- Nondestructive X-ray fluorescence testing of residual cladding thickness on two Mark SJA ($NpO_2$) tubes (Building 773-A, 1978) (DuPont 1978b).

- Program begun to develop a method for removing $NpO_2$ from waste rags from compact line cabinets in Building 235-F. This method reduced the amount of neptunium shipped to the burial ground. Over 1, 200 g of neptunium in Building 235-F solid waste had been buried from January 1973 through July 1975 (Building 772-F, 1975) (DuPont 1975a).

- The Building 321-M extrusion press was returned to service August 16, 1976, after a 5-week shutdown that included maintenance work to correct tooling misalignment that caused excessive tube eccentricity (DuPont 1976c).

- Development of a solvent extraction flowchart for processing $^{237}Np$ and $^{238}Pu$ as a possible replacement for anion exchange processing (Building 773-A, 1977) (Thompson and Thompson 1977).

- Encapsulated neptunium tubes analyzed for fuel density at the Building 305-M nuclear test gage, a light-water moderated subcritical reactor (DuPont 1976a, 1977b, 1978e, 1979a).

- 69 g of neptunium oxide shipped from HB Line to Building 773-A in December 1980 for use by the Actinide Technology group in the evaluation of the proposed neptunium process for the new H-Area B-line facilities (HB to Building 773-A, 1980) (DuPont 1980d).

- Tests of fission product content, nuclides present, and the plutonium content in $NpO_2$ to determine the shielding required to minimize personnel exposure in design of the new HB Line facility (Building 772-F, 1980) (DuPont 1980e).

- Research performed in a mini-mixer settler to achieve adaptation of the uranium(IV) reductant to the SRS PUREX processes (Building 773-A, 1986) (Orebaugh 1986).

- The Analytical Development Section developed a method to measure $^{237}Np$ holdup in "hot spots" in Building 235-F. The method was based on exposure readings in places where the background was too high to use gamma-pulse-height analysis [Savannah River Laboratory (SRL)], 1992) (Ferrell 1992).

- SRL researched methods for, and properties of, options for dissolution of neptunium oxide residues with the goal of running the method in H-Canyon (SRL, 2008) (Kyser 2009).

- SRL performed a laboratory-scale experiment to test a method for dissolution of nine unirradiated targets with the actual dissolution to be performed in H-Canyon (SRL, 2002) (Rudisill 2002).

- 98 g of neptunium oxide were produced on a bench scale from the same feed solution source for the gas generation studies (Building 773-A, 2003) (Duffey 2003a, 2003b, 2003c).

- SRL developed a dissolution flowchart for neptunium oxide residues (i.e., various $NpO_2$ sources, HB Line glovebox sweepings, and SRL analysis samples). Samples of each type of material proposed for processing were dissolved in a closed laboratory apparatus; the rate and total quantity of offgas were measured (SRL, 2008) (Kyser 2009; Watkins, Hensel, and Jordan 2009).

- Rapid separation method developed for $^{237}Np$ and plutonium isotopes in large soil samples (Building 773-A, 2010) (Maxwell, Culligan, and Noyes 2013).

- SRL investigated the neutralization and transfer of neptunium slurry from canyon wastes to the tank farm (SRL, 2011) (Walker et al. 2003).

Table 5-1. Ancillary work.

| Building | Work | Year | Reference |
|---|---|---|---|
| 305-M (NTG) | Density analysis | 1960–1984 | DuPont 1974b, 1974d, 1976a, 1977b, 1977c, 1977g, 1978c, 1978d, 1979a; Reed and Swanson 2006 |
| 736-A | Calibration of 1.1 g neptunium source | 1960 | DuPont 1960b |
| 772-F | Routine analyses for neptunium in process streams | 1953–2007 | DuPont 1959b, 1978b; Weinheimer 2012 |
| 772-F | Analytical development work | 1958–1984 | DuPont 1958b, 1975a, 1980f, 1980g; Ferrell 1992 |
| 773-A | Cold runs of Pu-238 production using neptunium in high-level caves \ | 1959 | DuPont 1957b |
| 773-A | Chemistry work; research | 1955–2011 | DuPont 1957c, 1965c; Pollock and Walz 1958; Moyer 1966a |
| 773-A | Shipping of neptunium to other sites | 1960–1967 | Tetzlaff 1960c; Moyer 1966a; DuPont 1967b |
| 773-A | Laboratory-scale testing of canyon equipment and processes | 1958–2008 | DuPont 1957b, 1958a, 1959a, 1961b, 1961e, 1961f, 1965c, 1968a, 1969f, 1971c, 1972a, 1972c, 1974g, 1977b, 1978b, 1980d; Rudisill 2002; Kyser 2009; Caracciolo 1961; Ice 1966; Thompson 1977; Orebaugh 1986; Duffey 2003c; Burney and Karraker 1958; Walker et al. 2003 |
| 773-A | Testing of Np fabrication | 1961–1978 | DuPont 1961c, 1962, 1963a, 1970b, 1968e, 1971b, 1974f, 1979a; Moyer 1969a; Marter 1961 |
| 777-M | Procedure tested to use 20 mg Np-237 | 1955 | Hyde 1955 |
| 777-M | Irradiation of Np foils | 1965–1968 | DuPont 1965b; Copley and Moyer 1968a |
| CMX | Flow test of Np oxide fuel assembly | 1969 | DuPont 1969e |
| TNX | Tests of recovery processes | 1958–1960 | DuPont 1957b, 1958b, 1960c, 1960d |

## 6.0 TIMELINE

The major neptunium processes and associated ancillary activities are presented in Figure 6-1. The timeline of neptunium processes and ancillary tasks by area and building from 1960 through 2010 are presented in Figure 6-2.

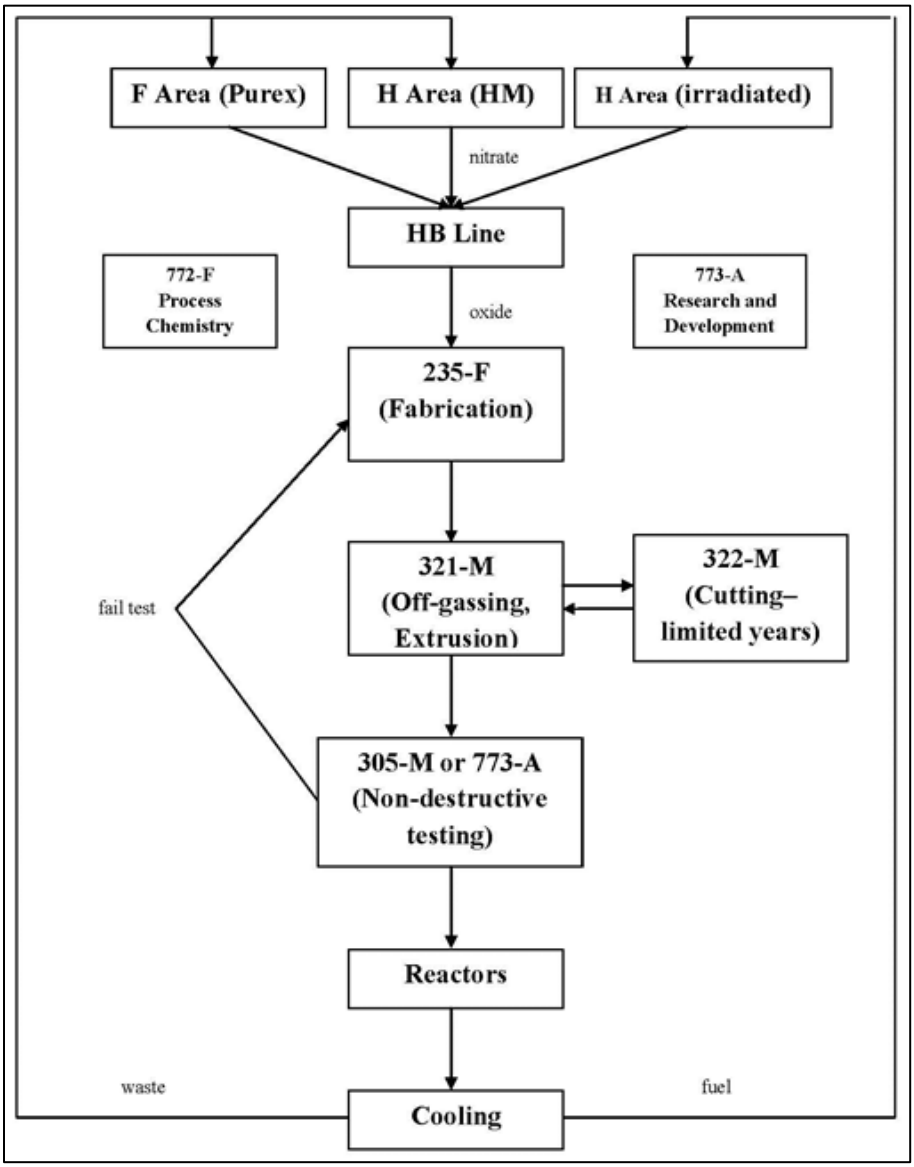



Figure 6-1. Neptunium process flowchart. (based on DuPont 1984a).

## 7.0 INVENTORY

The ORAU Team obtained data on $^{237}Np$ at SRS from DOE's Nuclear Materials Management and Safeguards System (NMMSS). The data include neptunium inventories from 1966 through 2012. The data supplied by DOE was maintained by "compcode" rather than location. The ORAU Team analyzed the data by the compcode description to subjectively map each of codes to one of the particular SRS process categories: Canyon Feed, HB Line, Fabrication, Reactor, and Lab. These categories do not include waste processes, but do include neptunium storage.

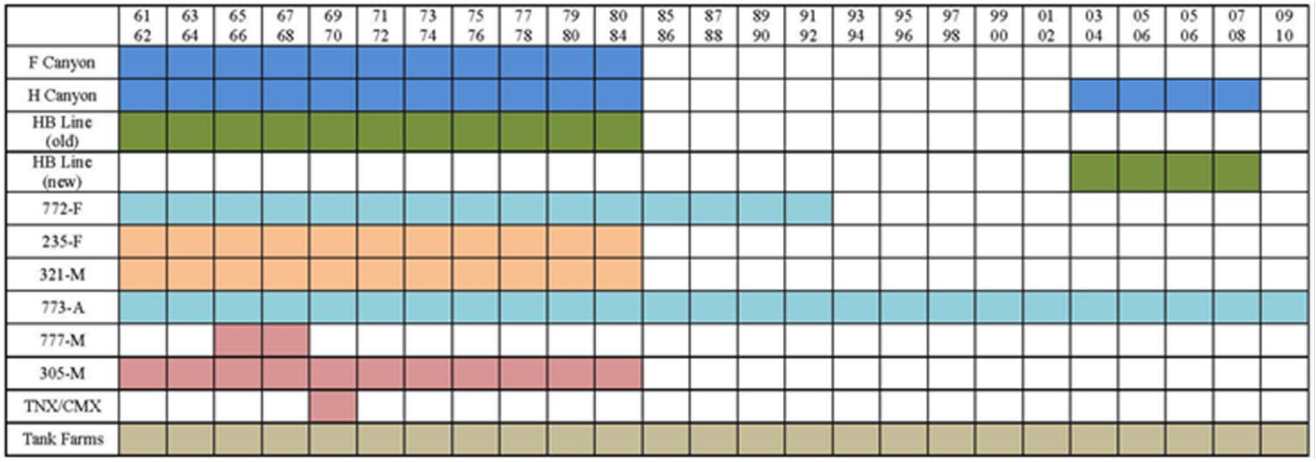



Figure 6-2. Neptunium process timeline.

The ORAU Team summed the quantity amounts (grams) across the code entries for each of the five process categories by the last month of each reported calendar quarter. Activities for each process category by month were calculated using the specific activity of $6.9 \times 10^{-4}$ Ci/g (Plexus-IEM 2016). Figure 7-1 shows inventories by major process plotted by ending date of calendar quarter. A large spike is shown for recovery from 1993 through 2007, but much of that material was in feed storage. Neptunium assigned to conversion, fabrication, and reactors after 1988 was also in feed storage until 2004 when the neptunium recovery and conversions were performed in H-Canyon and the new HB Line (Rudisill 2002). The activity amount of neptunium assigned to laboratories was quite small in relation to the feed and does not show in Figure 7-1.

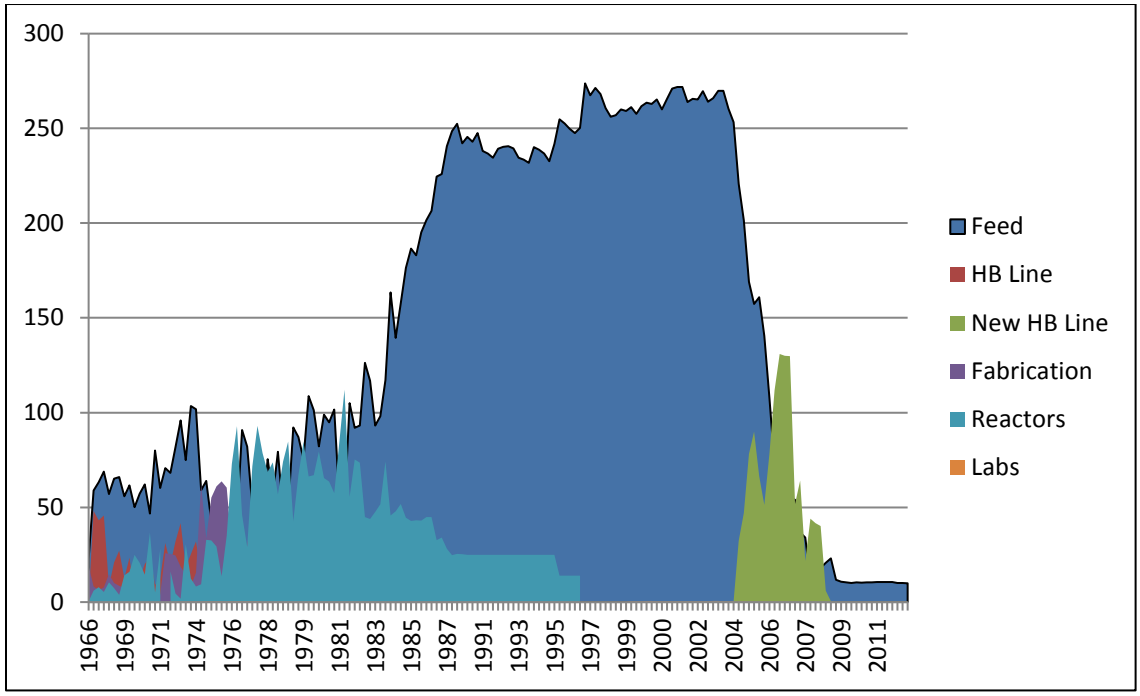



Figure 7-1. Neptunium by activity (Ci) by major process.

The NMMSS data cannot be used to determine specific amounts of $^{237}$Np by exact work location at a given time. However, it is apparent from other inventory data that some portions of neptunium inventories reported for dissolution and recovery and the HB Line were used in the actinide technology program at Building 773-A, Rooms B114, B115, and C155 (Brown 1983; Jowers 1984). Furthermore,

some portions reported for fabrication were used and stored at both Buildings 321-M and 773-A (Brown 1983). Actinide technology was largely involved with the chemistry of uranium, plutonium, and neptunium; ion exchange and solvent extraction development; mixing, filtration, and precipitation research; chemical and engineering studies of separation processes; process waste minimization; and power source development for space application.

## 8.0 RADIATION SAFETY AND MONITORING

SRS used radiological controls with the neptunium handling processes to reduce the potential for radiation exposure. The SRS Health Physics organization monitored neptunium work since its inception. The ORAU Team has reviewed SRS Works Technical Division monthly reports and health physics periodic reports to identify radiological controls and specific instances of occupational radiation monitoring. Routinely, highlights and summaries of information were included in such reports, while more detail and data from additional monitoring was provided on survey data forms. Radiological control of neptunium and other radionuclides was mandated by procedure DPSOP-40 and Special Hazards Bulletins (DuPont 1961h, 1972b).

### 8.1 RADIOLOGICAL CONTROLS

All canyon maintenance work was designed to be done remotely (ORAUT 2013a, 2013b). New equipment was installed in F- and H-Canyon facilities. The original equipment and subsequent replacement parts were first tested in a nonradioactive mock-up shop section of each canyon (Fernandez 2000). Small modular units called “frames” were installed by skilled workers using novel techniques in a canyon space approximately 10 ft square by 17 ft high with a standard arrangement of pipe nozzles to supply services in both F- and H-Canyons to process waste streams and irradiated targets. Within the frames in F-Canyon, four ion exchange columns were installed.

In H-Canyon, five columns and a dissolver were installed for neptunium-plutonium separation from irradiated neptunium targets. The size of the ion exchange columns was small in comparison with regular canyon equipment; this permitted development of a concept in which a number of columns and tanks could be mounted in a frame that could be installed as one modular unit. The canyon frames were installed in modules 10 ft square by 17 ft high with standard services (i.e., inlet and outlet piping, steam, cooling water, electricity, sampling, and instruments). The modules were isolated by a low curb to contain liquid spills or leaks, and had a sump and a permanently installed transfer jet to move spilled liquids or leakage to a rework system. Installing multiple items in a frame was considered an economical use of canyon space, but it also entailed a burden when providing services to a module. Pipe and electrical connections were made to the frame using conventional canyon jumpers rather than to individual pieces of equipment. Electrical and other services were piped to equipment pieces as part of the permanent frame structure. Additional services were required though, primarily for pneumatic liquid level and specific gravity instrumentation for the several vessels in each frame. Bundles of up to six small, stainless-steel tubes were drawn through 3-in. pipes embedded in the canyon shielding walls. No significant problems were reported for the installation of these lines or their use (Poe, Joyce, and Martens 1963; WSRC 2000; Overbeck, Ice, and Dessauer 1965). In 2000, Groh, Poe, and Porter (2000) reported that the useful life of the frames was equivalent to the life of embedded piping. A photo of a canyon frame is shown in Figure 8-1.

The ion exchange columns used in neptunium recovery were either 12- or 24-in. in diameter and normally contained 25 or 100 L of resin. By design, they were used as conventional settled-bed ion exchange columns, but they could also be used as agitated ion exchange bed columns. One unit was used routinely as an agitated bed column. All maintenance operations were performed remotely without mechanical valves, including resin replacement. There were sometimes events that resulted in contamination. Those were resolved remotely. For example, in June 1962, solution from a $^{237}$Np-$^{238}$Pu raw metal process filter drained through an embedded line to a blanked flange on the outside canyon

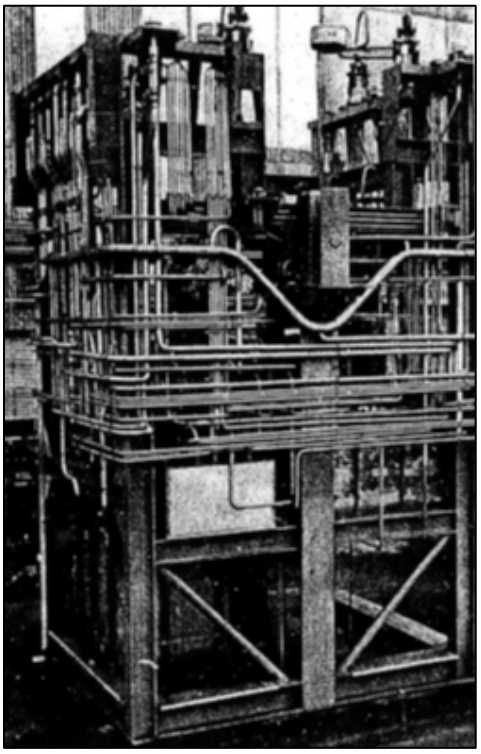


Figure 8-1. Neptunium-plutonium frame without ion exchange columns (Groh, Poe, and Porter 2000).

wall. A prefabricated 3-in. lead shield was remotely installed over the portion of the line, which extended 6 in. from the canyon wall. The shield reduced the radiation level from 5 R/hr to 30 mR/hr at 2 ft from the flange. Equipment to permit remote decontamination of the embedded pipe from inside the hot canyon was used to clean up the contamination (DuPont 1980h).

Two weirs determined flow paths. Air pressure was applied to either or both to direct flow properly for feed, wash, elution, or resin removal depending on the task. Dowex 1 and Permutit SK anion exchange resins were used in the columns (Hill 1961). Dowex 1 resin provided higher decontamination from fission products, while Permutit SK resin was more stable with alpha radiation. Selected use of smaller particle resins were used where fast kinetic absorption was required, such as in the ion exchange column used for separation of $^{238}$Pu from $^{237}$Np. A diagram of an ion exchange column used in the process is presented in Figure 8-2.

The original HB Line was also modified to provide facilities to convert the purified $^{238}$Pu product and the recovered $^{237}$Np to the final oxide forms. Neptunium-237 and $^{238}$Pu finishing operations were performed in HB Line gloveboxes. The $^{238}$Pu operations were performed in a new small glovebox line, but the neptunium was finished in the old B-Line cabinets and gloveboxes. A new HB Line, constructed over Sections 2 through 6 of H-Canyon, was designed with three functional areas: (1) purification of neptunium; (2) purification of $^{238}$Pu; and (3) recovery of neptunium and plutonium from scrap. The new neptunium oxide line, placed in use by 1984, was constructed over Sections 4 and 5 of the 221-H-Canyon. All work with $^{237}$Np was performed in two glovebox lines constructed with adjacent operating and maintenance rooms to minimize spread of contamination. The gloveboxes contained ion exchange columns and equipment to precipitate, finish, and package neptunium oxide for shipment to Building 235-F for target fabrication.

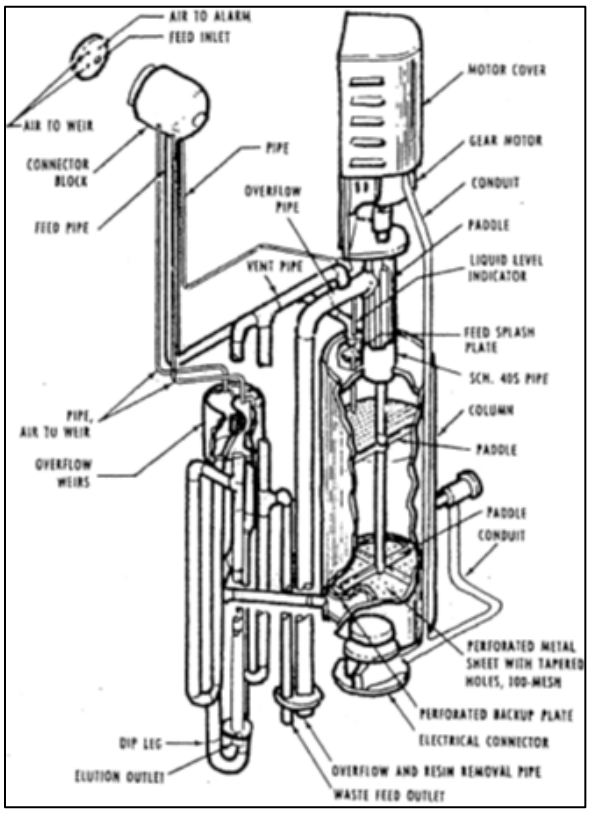



Figure 8-2. Ion exchange column (Groh, Poe, and Porter 2000).

A structure referred to as a cabinet (a small glovebox line) was used in Building 235-F to process neptunium oxide. A cabinet was a ventilated, walled, shielded glovebox. The neptunium billet room was posted as a Regulated Radiation Area. Shadow shielding was used due to high gamma dose rates. All process work in 235-F was done through glove ports. Workers donned anticontamination clothing in 235-F as shown in the example in Figure 3-2. Maintenance of the glovebox cabinets was performed on the reverse side of the process line in a room walled off from the process area.

Maintenance was performed inside cabinets, but contamination control huts were constructed and used (DuPont 1974–1984). Workers dressed in protective clothing and gloves appropriate for the task to be conducted, as assigned by Health Physics (ORAUT 2013c). Job task analyses were done for all nonroutine work. Health Physics reviewed those analyses and added measures necessary to keep radiation exposures as low as possible. Measures included but were not limited to contamination huts, additional shielding, protective clothing, and health physics surveys of the job. Workers checked their hands and feet for contamination before exiting areas where such work was performed.

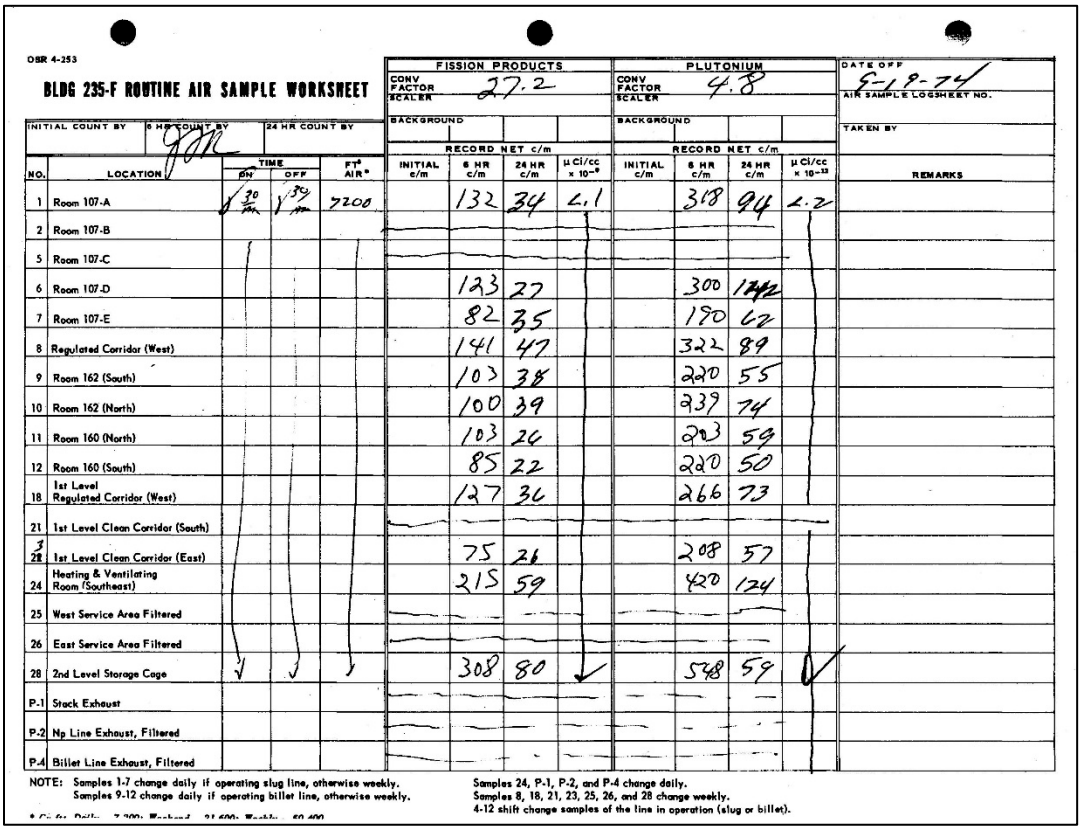
SRS continued to evaluate radiation exposures and make process and equipment upgrades over time. For example, additional shielding was installed on the lubricating cabinet in the neptunium billet line in Building 235-A in 1977. At that time only the adjacent die-press cabinet was still unshielded. The lead shield and leaded-glass viewing window reduced the body exposure dose rate from 35 to 5 mR/hr (DuPont 1977i). Access was controlled to each of the buildings where neptunium was processed (ORAUT 2013b, 2013c). Entry into canyon buildings and 235-F was by approved worker only. Special nuclear materials were stored and processed in both 235-F and 321-M; as such, those buildings had secured access. Workers were required to have Health Physics approval to enter specific neptunium work locations (ORAUT 2013b, 2013c).

The Health Physics organization implemented a radiation work permit process in 1990. Job requirements and control of access to the job area were carried out with the use of radiation work permits. SRS implemented new radiation control practices in 1990 to comply with DOE Order 5480.11 (DOE 1988), and updated them over time to comply with:

- 1992 DOE Radiological Control Manual, DOE N 5480.6 (DOE 1992),
- 1994 DOE Radiological Control Manual, DOE/EH-0256T (DOE 1994), and
- 1995 Occupational Radiation Protection (10 CFR Part 835).

## 8.2 WORKPLACE CONTROLS

SRS Health Physics routinely monitored F- and H-Canyons, HB-Line, the 235-F neptunium line, 321-M and 773-A for surface and airborne contamination, and they did special monitoring of certain jobs and monitoring of contamination incidents. Radiation survey and air monitoring data for each area where neptunium was processed are available. Primary areas of radiological concern were the neptunium billet line in 235-F, HB-Line, and the extrusion furnace in 321-M. There are extensive air monitoring data for Building 235-F. The data that includes results of fixed process, routine, and special samples shows that that air concentrations of neptunium were controlled to less than $2 \times 10^{-13}$ µCi/cm$^3$, which was 5% of the Radioactivity Concentration Guide (RCG) for neptunium, $4 \times 10^{-12}$ µCi/cm$^3$ (DuPont 1971–1974). Representative routine, special, and process examples of 235-F air monitoring are given in Figures 8-3, 8-4 and 8-5, respectively.



OBR 4-253

**BLDG 235-F ROUTINE AIR SAMPLE WORKSHEET**

FISSION PRODUCTS — CONV FACTOR SCALER 27.2 — BACKGROUND — RECORD NET c/m

PLUTONIUM — CONV FACTOR SCALER 4.8 — BACKGROUND — RECORD NET c/m

DATE OFF 5-19-74 — AIR SAMPLE LOGSHEET NO. — TAKEN BY

INITIAL COUNT BY — 6 HR COUNT BY — 24 HR COUNT BY

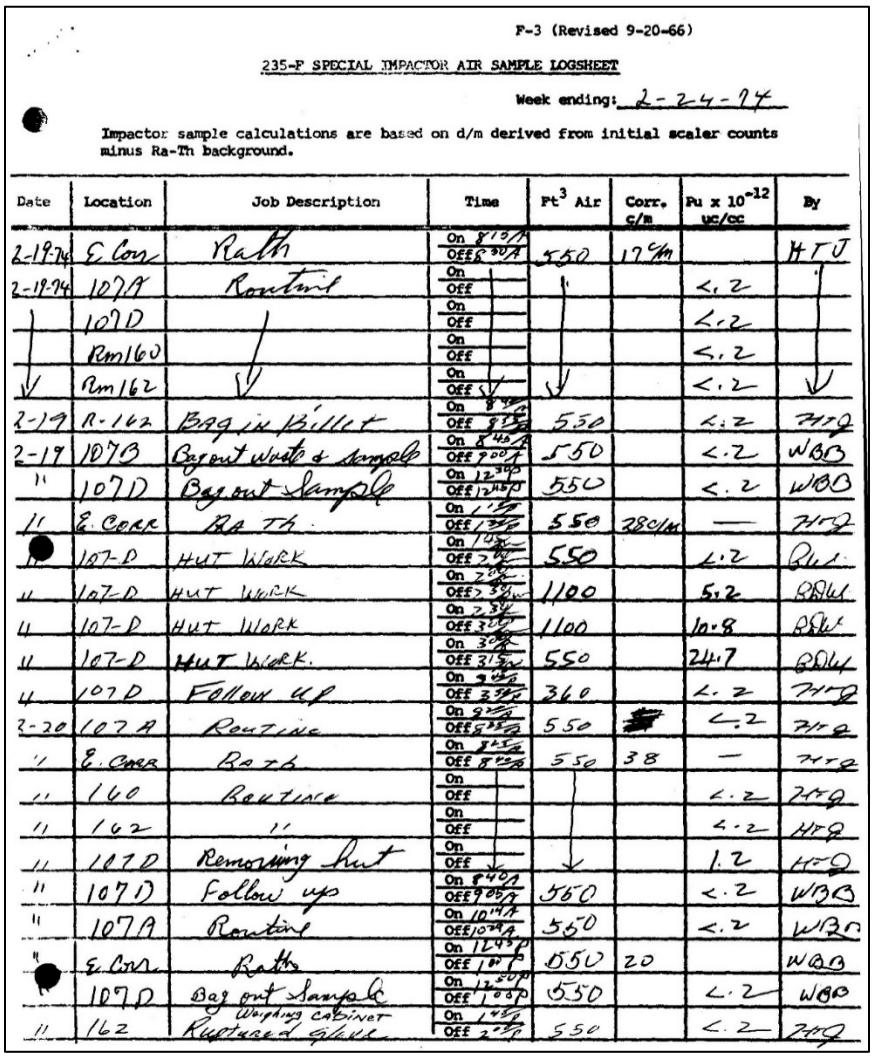
| NO. | LOCATION | TIME ON | TIME OFF | FT³ AIR* | FISSION INITIAL c/m | FISSION 6 HR c/m | FISSION 24 HR c/m | FISSION µCi/cc × 10⁻⁹ | PLUTONIUM INITIAL c/m | PLUTONIUM 6 HR c/m | PLUTONIUM 24 HR c/m | PLUTONIUM µCi/cc × 10⁻¹³ | REMARKS |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | Room 107-A | 7:30 am | 7:39 am | 7200 | | 132 | 34 | 2.1 | | 318 | 94 | 2.2 | |
| 2 | Room 107-B | | | | | | | | | | | | |
| 5 | Room 107-C | | | | | | | | | | | | |
| 6 | Room 107-D | | | | | 123 | 27 | | | 300 | 1742 | | |
| 7 | Room 107-E | | | | | 82 | 35 | | | 180 | 62 | | |
| 8 | Regulated Corridor (West) | | | | | 141 | 47 | | | 322 | 89 | | |
| 9 | Room 162 (South) | | | | | 103 | 38 | | | 220 | 55 | | |
| 10 | Room 162 (North) | | | | | 100 | 39 | | | 239 | 74 | | |
| 11 | Room 160 (North) | | | | | 103 | 26 | | | 293 | 59 | | |
| 12 | Room 160 (South) | | | | | 85 | 22 | | | 220 | 50 | | |
| 18 | 1st Level Regulated Corridor (West) | | | | | 127 | 36 | | | 266 | 73 | | |
| 21 | 1st Level Clean Corridor (South) | | | | | | | | | | | | |
| 23 | 1st Level Clean Corridor (East) | | | | | 75 | 26 | | | 208 | 57 | | |
| 24 | Heating & Ventilating Room (Southeast) | | | | | 215 | 59 | | | 420 | 124 | | |
| 25 | West Service Area Filtered | | | | | | | | | | | | |
| 26 | East Service Area Filtered | | | | | | | | | | | | |
| 28 | 2nd Level Storage Cage | ✓ | ✓ | ✓ | | 308 | 80 | ✓ | | 548 | 59 | ✓ | |
| P-1 | Stack Exhaust | | | | | | | | | | | | |
| P-2 | Np Line Exhaust, Filtered | | | | | | | | | | | | |
| P-4 | Billet Line Exhaust, Filtered | | | | | | | | | | | | |

NOTE: Samples 1-7 change daily if operating slug line, otherwise weekly.
Samples 9-12 change daily if operating billet line, otherwise weekly.

Samples 24, P-1, P-2, and P-4 change daily.
Samples 8, 18, 21, 23, 25, 26, and 28 change weekly.
4-12 shift change samples of the line in operation (slug or billet).

Figure 8-3. Routine air monitoring results, Building 235-F (DuPont 1973–1974).

Airborne activity in 235-F was generally controlled to <0.2 x $10^{-12}$ µC/cm$^3$. Routine and process samples were first counted 6 hours after collection and recounted at 24 hours to account for radon.



F-3 (Revised 9-20-66)

235-F SPECIAL IMPACTOR AIR SAMPLE LOGSHEET

Week ending: 2-24-74

Impactor sample calculations are based on d/m derived from initial scaler counts minus Ra-Th background.

| Date | Location | Job Description | Time | Ft³ Air | Corr. c/m | Pu x 10⁻¹² uc/cc | By |
|---|---|---|---|---|---|---|---|
| 2-19-74 | E. Corr. | Rath | On 8:10A Off 8:30A | 550 | 17 c/m | | HTJ |
| 2-19-74 | 107A | Routine | On Off | | | <.2 | |
| | 107D | | On Off | | | <.2 | |
| | Rm 160 | | On Off | | | <.2 | |
| | Rm 162 | | On Off | | | <.2 | |
| 2-19 | R-162 | Bag in Billet | On 8:35A Off 8:55A | 550 | | <.2 | HTJ |
| 2-19 | 107B | Bag out Waste & Sample | On 8:40A Off 9:00A | 550 | | <.2 | WBB |
| " | 107D | Bag out Sample | On 12:30P Off 12:50P | 550 | | <.2 | WBB |
| " | E. Corr. | Ra Th | On 1:15P Off 1:35P | 550 | 38 c/m | — | HTJ |
| | 107-D | Hut Work | On 1:40P Off 2:00P | 550 | | <.2 | RLL |
| " | 107-D | Hut Work | On 2:05P Off 2:35P | 1100 | | 5.2 | RRLL |
| " | 107-D | Hut Work | On 2:35P Off 3:00P | 1100 | | 10.8 | RRLL |
| " | 107-D | Hut Work. | On 3:00P Off 3:15P | 550 | | 24.7 | RRLL |
| " | 107D | Follow up | On 3:00P Off 3:30P | 360 | | <.2 | HTJ |
| 2-20 | 107A | Routine | On 9:20A Off 5:35A | 550 | [redacted] | <.2 | HTJ |
| " | E. Corr. | Rath | On 8:15A Off 8:35A | 550 | 38 | — | HTJ |
| " | 160 | Routine | On Off | | | <.2 | HTJ |
| " | 162 | " | On Off | | | <.2 | HTJ |
| " | 107D | Removing Hut | On Off | | | 1.2 | HTJ |
| " | 107D | Follow up | On 8:40A Off 9:05A | 550 | | <.2 | WBB |
| " | 107A | Routine | On 10:14A Off 10:29A | 550 | | <.2 | WBB |
| " | E. Corr. | Rath | On 12:45P Off 1:05P | 550 | 20 | | WBB |
| | 107D | Bag out Sample | On 12:50P Off 1:05P | 550 | | <.2 | WBB |
| " | 162 | Weighing Cabinet Ruptured Glove | On 1:45P Off 2:05P | 550 | | <.2 | HTJ |

Figure 8-4. Special air monitoring results, Building 235-F (DuPont 1973–1974).

Health Physics performed daily control surveys in the neptunium billet room. A representative survey log is shown in Figure 8-6.

Representative dose rate surveys for $^{237}Np$ exposures in Building 773A are provided in Table 8-1.

Completed billets were bagged for transfer to Building 321-M for extrusion. Billets were smeared and surveyed by Health Physics before the transfer. A representative billet survey is shown in Figure 8-7. The survey shows a high gamma dose rate with a neutron component of about 1%.

Billets were also surveyed at 321-M when received. A representative survey of received billets shown in Figure 8-8.

Extruded tubes were surveyed for dose rate once they came from the oven; a representative survey is shown in Figure 8-9.

Tubes made from extrusion of neptunium billets were surveyed before being transferred to the reactors for irradiation. A representative survey of neptunium tubes is shown in Figure 8-10.

OSR 4-323
(Rev 9-75)

**PROCESS AIR SAMPLE RESULTS, F-AREA**
**BUILDING 235-F**
D/M X .00101

SAMPLE: P-2 BLDG. PROCESS EXHAUST HDR. (FILTERED)
SAMPLER LOCATION: FILTER RM. | EXHAUST STREAM: 9,000 cfm
TYPE FILTER PAPER: HV-70 | SAMPLING RATE: 5 cfm

| | DATE | TIME | FT³ AIR | SCALER CONV FACTOR | BETA-GAMMA SAMPLE RESULTS MRADS/HR AT 2" | INITIAL c/m | 24 HR c/m | d/m* | BETA-GAMMA µCi RELEASED DAILY | MONTH TO DATE | SCALER CONV FACTOR d/m/c/m | ALPHA SAMPLE RESULTS INITIAL c/m OVER d/m | 6 HR c/m OVER d/m | 24 HR c/m OVER d/m | $c_p$* | ALPHA µCi RELEASED DAILY | MONTH TO DATE |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| ON | 10/12/77 | 8:30a | | | | | | | | | | | | | | | |
| OFF | 10/13/77 | 8:30a | 7200 | 19.6 | — | 122 | 29 | 568 | .5741 | 5.0869 | 4.6 | 166 | 129 | 25 | 0 | 0 | .1579 |
| ON | 10/13/77 | 8:30a | | | | | | | | | | | | | | | |
| OFF | 10/14/77 | 8:30a | 7200 | 19.6 | — | 131 | 29 | 568 | .5741 | 5.6610 | 4.6 | 169 | 139 | 28 | 0 | 0 | .1579 |
| ON | 10/15/77 | 8:30a | | | | | | | | | | 371 | 216 | 22 | | | |
| OFF | 10.16 | 8:30a | 7200 | 19.6 | — | 123 | 23 | 451 | .4553 | 6.1163 | 4.6 | | | | 0 | 0 | .1579 |
| ON | 10.16 | 8:30a | | | | | | | | | | 188 | 147 | 30 | | | |
| OFF | 10.16 | 8:30a | 7200 | 19.6 | — | 135 | 33 | 647 | .6533 | 6.7696 | 4.6 | | | | 0 | 0 | .1579 |
| ON | 〃 | 〃 | | | | | | | | | | 229 | 133 | 40 | | | |
| OFF | 10-17 | 〃 | 7200 | 〃 | | 151 | 20 | 392 | .3959 | 7.1663 | 〃 | | | | 0 | 0 | .1579 |
| ON | 10/17 | 11 | | | | | | | | | | 242 | 135 | 37 | | | |
| OFF | 10/18 | 11 | 7200 | 19.6 | | 143 | 27 | 529 | .5345 | 7.7 | 4.6 | | | | 0 | 0 | .1579 |
| ON | 10/18 | 11 | | | | | | | | | | 205 | 140 | 41 | | | |
| OFF | 10/19 | 11 | 7200 | 19.6 | | 128 | 22 | 431 | .4353 | 8.1353 | 4.6 | | | | 0 | 0 | .1579 |
| ON | 〃 | 〃 | | | | | | | | | | 225 | 160 | 49 | | | |
| OFF | 10-20 | 〃 | 7200 | 〃 | | 133 | 21 | 412 | .4157 | 8.551 | 〃 | | | | 0 | 0 | .1579 |
| ON | 〃 | 〃 | | | | | | | | | | 259 | 113 | 19 | | | |
| OFF | 10-21 | 〃 | 〃 | 〃 | | 129 | 21 | 412 | .4157 | 8.967 | 〃 | | | | 0 | 0 | .1579 |
| ON | 10-21 | 11 | | | | | | | | | | 362 | 131 | 38 | | | |
| OFF | 10-22 | 〃 | 〃 | 〃 | | 199 | 20 | 392 | .3959 | 9.363 | 〃 | | | | 0 | 0 | .1579 |
| ON | 〃 | 〃 | | | | | | | | | | 290 | 129 | 33 | | | |
| OFF | 10-23 | 〃 | 〃 | 〃 | | 177 | 19 | 372 | .3761 | 9.7391 | 〃 | | | | 0 | 0 | .1579 |
| ON | 10/23 | 〃 | | | | | | | | | | 160 | 108 | 37 | | | |
| OFF | 10/24 | 〃 | 〃 | 〃 | | 82 | 18 | 353 | .3563 | 10.0954 | 〃 | | | | 5 | .02372 | .1811 |
| ON | 〃 | 〃 | | | | | | | | | | 170 | 99 | 18 | | | |
| OFF | 10-25 | 〃 | | 〃 | | 99 | 19 | 372 | .3761 | 10.4715 | 〃 | | | | 0 | 0 | .1811 |
| ON | 〃 | 〃 | | | | | | | | | | 155 | 121 | 28 | | | |
| OFF | 10-26 | 〃 | | 〃 | | 112 | 21 | 412 | .4157 | 10.8872 | 〃 | | | | 0 | 0 | .1811 |
| ON | 〃 | 〃 | | | | | | | | | | 161 | 114 | 34 | | | |
| OFF | 10-27 | 〃 | | 〃 | | 114 | 21 | 412 | .4157 | 11.3029 | 〃 | | | | 0 | 0 | .1811 |

* Use this number in the following formula to determine release rate. See DPSOL 193-119 for determining $C_p$.

µCi RELEASED = ——— × ———— , cfm (EXHAUST STREAM)
———— , cfm (SAMPLING RATE) × 0.80 × 2.22 × $10^6$ d/m/µCi

Figure 8-5. Process air monitoring results, Building 235-F (Brown 2016).

Revised 2/83

Week Ending 3/27/83

235-F
DAILY CONTROL SURVEYS

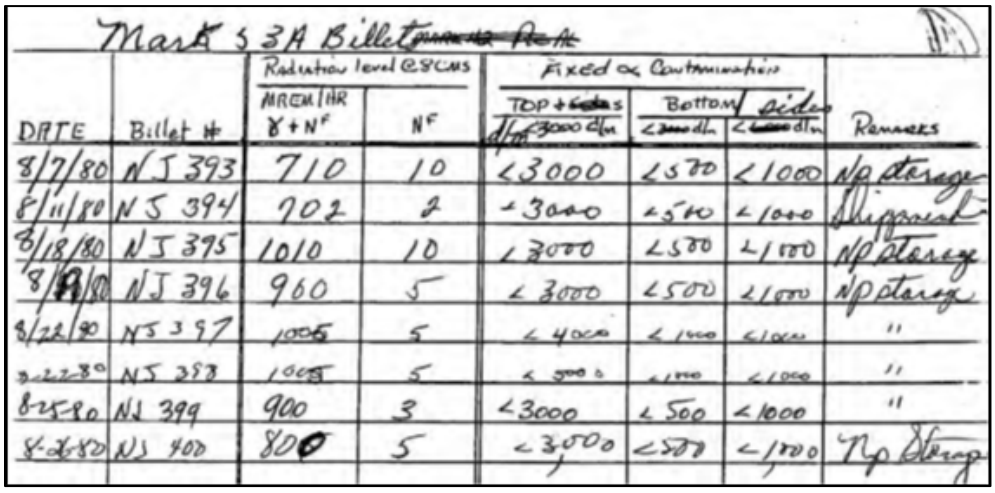
A signature in the proper space indicates a complete survey was made. Notify supervision and prepare a diagram showing conditions if clean area has transferable contamination above 10 c/m beta-gamma and 10 d/m alpha and regulated area has contamination above 1000 c/m beta-gamma and 500 d/m alpha. Note all unusual radiation levels.

| ITEM | SHIFT | M | T | W | T | F | S | S |
|---|---|---|---|---|---|---|---|---|
| Source check scalers, record results | 8-4 | JCD | JCD | JCD | JCD | JCD | X | X |
| Source check H & F monitors | 8-4 | SFE | JCD | JCD | JCD | JCD | X | X |
| Change High Volume CAM Samples - Count: Sat. & Sun. as needed | 8-4 | JCD | JCD | JCD | JCD | JCD | | |
| Change planchets on portable CAMs and source check | 8-4 | LDP | LDP | LDP | JCD | JCD | X | X |
| Change P-1 (Process) and P-2 (Room) duct air samples - give proper counts | 8-4 | SFE | SFE | SFE | LDP | JCD | | |
| Change all Filter Paper Samples - Give proper count | 8-4 | JCD SFE | JCD SFE | JCD SFE | 〃 | JCD | X | X |
| PUFF Control Rm - Manipulator Collars | 8-4 | JCD | JCD | JCD | JCD | JCD | X | X |
| PUFF Control Rm (clean) | 8-4 | JCD | JCD | JCD | JCD | JCD | X | X |
| | 4-12 | 〃 | 〃 | 〃 | 〃 | 〃 | X | X |
| SOPs to clean areas (clean) | 8-4 | JCD | JCD | JCD | JCD | JCD | X | X |
| | 4-12 | 〃 | 〃 | 〃 | 〃 | 〃 | X | X |
| Lunch rms - (instrument and disc smears) (clean) (prior to lunch period) | 8-4 | JCD | JCD | JCD | JCD | JCD | X | X |
| | 4-12 | 〃 | 〃 | 〃 | 〃 | 〃 | X | X |
| Source check CAMS: 4-12 Areas in Use | 8-4 | JCD | JCD | JCD | JCD | JCD | X | X |
| | 4-12 | 〃 | 〃 | 〃 | 〃 | 〃 | X | X |
| Floors in Process Rms and Reg Corridors (Reg) | 8-4 | JCD | JCD | JCD | JCD | JCD | X | X |
| | 4-12 | 〃 | 〃 | 〃 | 〃 | 〃 | X | X |
| Gloves - at start of shift; 1. PUFF E. Maint Rm - all Cell No. 1 lower 2. Met Lab - all gloves (if operating) 3. NP - all gloves in operating rm | 8-4 | SFE | SFE | SFE | SFE | SFE | X | X |
| | 4-12 | 〃 | 〃 | 〃 | 〃 | 〃 | X | X |
| Regulated Change rms and toilets (toilets - clean) | 4-12 | 〃 | 〃 | 〃 | 〃 | 〃 | X | X |
| Disc smear Bioassay Stations, (clean) | 4-12 | 〃 | 〃 | 〃 | 〃 | 〃 | X | X |

Figure 8-6. Daily control sheet, Building 235-F (DuPont 1981–1984, p. 87).

Table 8-1. Health physics surveys in Building 773-A (mrem/hr).

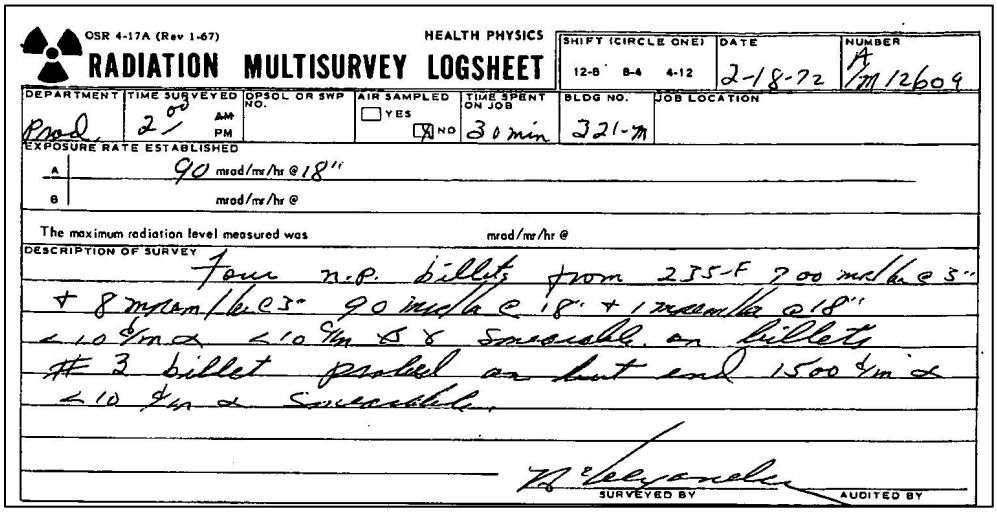
| Month | Location | Gross exposure rate (γ) at 3 in. | Fast neutron exposure rate at 3 in. | Reference |
|---|---|---|---|---|
| June 1968 | B-130 glovebox | 35 | Not measured | Copley and Moyer 1968b |
| July 1968 | B-130 glovebox | 15 | Not measured | Copley and Moyer 1968c |
| August 1968 | B-130 glovebox | 50 | Not measured | Copley and Moyer 1968d |
| September 1968 | B-130 glovebox | 50 | Not measured | Copley and Moyer 1968e |
| October 1968 | B-130 glovebox | 60 | Not measured | Copley and Moyer 1968g |
| November 1968 | B-130 glovebox | 60 | Not measured | Copley and Moyer 1968f |
| December 1968 | B-130 glovebox | 30 | Not measured | Copley and Moyer 1969a |
| January 1969 | B-130 glovebox | 40 | Not measured | Copley and Moyer 1969b |
| February 1969 | B-130 glovebox | 40 | Not measured | Copley and Moyer 1969c |
| April 1969 | B-130 glovebox | 40 | 5 | Copley and Moyer 1969d |
| May 1969 | B-130 glovebox | 40 | 10 | Copley and Moyer 1969e |
| June 1969 | B-130 glovebox | 30 | <1 | Moyer 1969b |
| June 1969 | B-151 glovebox | 40 | Not measured | Moyer 1969a |



Mark 53A Billets

| DATE | Billet # | Radiation level @ 8 CMS MREM/HR γ+N^F | N^F | Fixed Contamination TOP + sides | Bottom | Sides | Remarks |
|---|---|---|---|---|---|---|---|
| 8/7/80 | NJ 393 | 710 | 10 | <3000 | <500 | <1000 | Np storage |
| 8/11/80 | NJ 394 | 702 | 2 | <3000 | <500 | <1000 | Shipped |
| 8/18/80 | NJ 395 | 1010 | 10 | <3000 | <500 | <1000 | Np storage |
| 8/19/80 | NJ 396 | 960 | 5 | <3000 | <500 | <1000 | Np storage |
| 8/22/80 | NJ 397 | 1005 | 5 | <4000 | <1000 | <1000 | " |
| 8-22-80 | NJ 398 | 1005 | 5 | <3000 | <1000 | <1000 | " |
| 8-25-80 | NJ 399 | 900 | 3 | <3000 | <500 | <1000 | " |
| 8-26-80 | NJ 400 | 800 | 5 | <3000 | <500 | <1000 | Np Storage |

Figure 8-7. Survey of neptunium billets to be shipped to Building 321-M (Brown 2014, p. 22).



OSR 4-17A (Rev 1-67) HEALTH PHYSICS

RADIATION MULTISURVEY LOGSHEET

SHIFT (CIRCLE ONE): 12-8 8-4 4-12 | DATE: 2-18-72 | NUMBER: A M12609

DEPARTMENT: Prod. | TIME SURVEYED: 2 00 AM PM | OPSOL OR SWP NO. | AIR SAMPLED: YES NO | TIME SPENT ON JOB: 30 min | BLDG NO.: 321-M | JOB LOCATION

EXPOSURE RATE ESTABLISHED

A: 90 mrad/hr @ 18"

B: mrad/hr @

The maximum radiation level measured was mrad/hr @

DESCRIPTION OF SURVEY

Four n.p. billets from 235-F 700 mr/hr @ 5" + 8 mrem/hr @ 3" 90 mr/hr @ 18" + 1 mr/hr @ 18" < 10 γ/m² < 10 γ/m² & & Smearable on billets # 2 billet packed on butt end 1500 γ/m² < 10 γ/m² & Smearable.

SURVEYED BY AUDITED BY

Figure 8-8. Survey of neptunium billets when received at Building 321-M (DuPont 1972d).

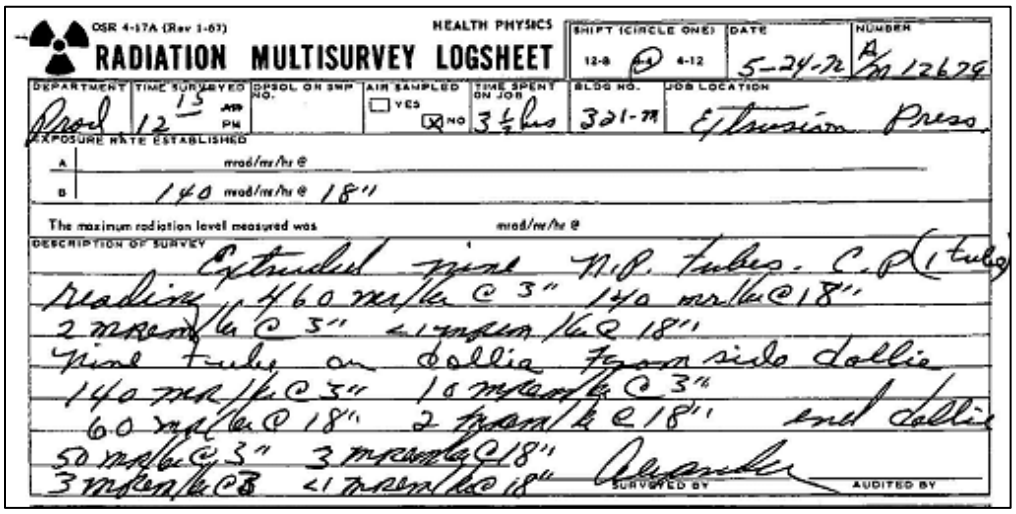

OSR 4-17A (Rev 1-67) HEALTH PHYSICS
RADIATION MULTISURVEY LOGSHEET
SHIFT (CIRCLE ONE) 12-8 (8) 4-12 DATE 5-24-72 NUMBER A 12679
DEPARTMENT Prod TIME SURVEYED 12 15 AM PM DPSOL OR SWP NO. AIR SAMPLED YES NO TIME SPENT ON JOB 3½ hrs BLDG NO. 321-M JOB LOCATION Extrusion Press
EXPOSURE RATE ESTABLISHED
A mrad/mr/hr @
B 140 mrad/mr/hr @ 18"
The maximum radiation level measured was mrad/mr/hr @
DESCRIPTION OF SURVEY
Extruded final N.P. Tubes. C.P. (1 tube)
reading 460 mr/hr @ 3" 140 mr/hr @ 18"
2 mrem/hr @ 3" <1 mrem/hr @ 18"
final tube on dollie from side dollie
140 mr/hr @ 3" 10 mrem/hr @ 3"
60 mr/hr @ 18" 2 mrem/hr @ 18" end dollie
50 mr/hr @ 3" 3 mrem/hr @ 18"
3 mrem/hr @ 3" <1 mrem/hr @ 18"
SURVEYED BY AUDITED BY

Figure 8-9. Survey of freshly extruded neptunium tubes at Building 321-M (DuPont 1972e).

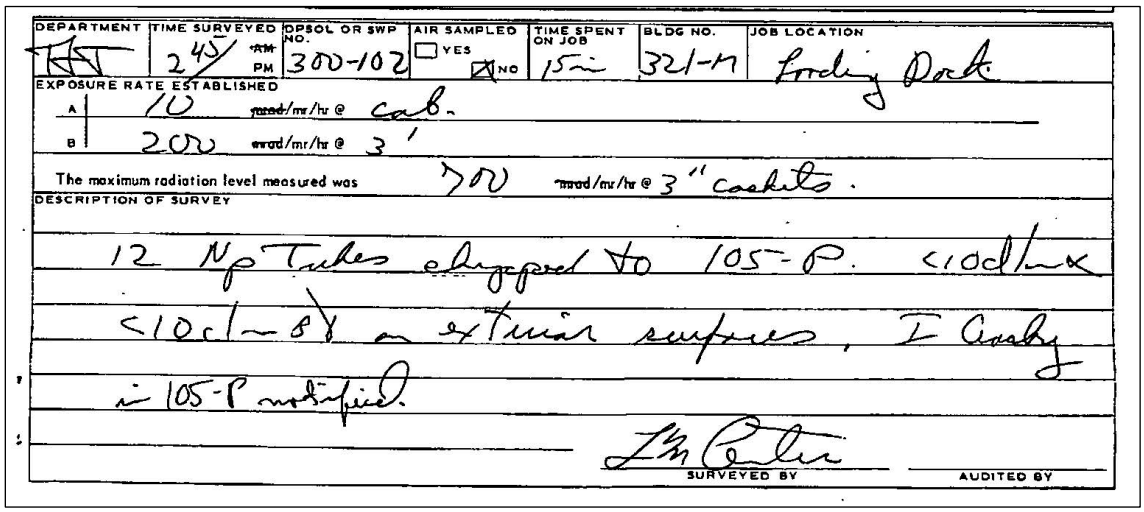

DEPARTMENT TAT TIME SURVEYED 245 AM PM DPSOL OR SWP NO. 300-102 AIR SAMPLED YES NO TIME SPENT ON JOB 15 min BLDG NO. 321-M JOB LOCATION Loading Dock
EXPOSURE RATE ESTABLISHED
A 10 mrad/mr/hr @ cab.
B 200 mrad/mr/hr @ 3'
The maximum radiation level measured was 700 mrad/mr/hr @ 3" casketts.
DESCRIPTION OF SURVEY
12 Np Tubes shipped to 105-P. <10 dpm x
<10 dpm-β on exterior surfaces. T Cosby
in 105-P notified.
SURVEYED BY AUDITED BY

Figure 8-10. Survey of neptunium billets when received at Building 321-M (DuPont 1972e).

At times, the neptunium billet line was used to fabricate $^{242}$Pu billets using the same procedures and controls. These billets were also bagged for transfer. A representative survey of $^{242}$Pu (Mark-42) billets is shown in Figure 8-11. Those surveys reveal a lower gamma dose rate but a higher neutron component (about 25% of the total dose rate). Operators on the neptunium billet line wore white regulated clothing with both beta/gamma and neutron dosimeters (DuPont 1971–1974). Supervisors wore white laboratory coats with shoe covers; they were monitored by beta/gamma dosimetry.

SRS monitored the workplace, equipment, neptunium parts, and jobs to control exposures to neptunium. Additional representative examples are provided in Attachment A, Table A-1, for 1966 through 1980.

## 8.3 WORKER MONITORING

Monitoring for potential intakes of radionuclides at SRS was mandated and controlled by DPSOL 193-302 from 1971 through 1988 (DuPont 1971d) and DPSOL 193-211 through 1989 (DuPont 1989). Bioassay monitoring was prescribed by work area with sampling frequencies based on exposure potential. Twenty-four-hour urine samples were collected from workers for $^{237}$Np analysis when a

MARK 42 Pu Al

| DATE | Billet # | Radiation level @ 8 CMS MREM/HR β+N$^F$ | N$^F$ | Fixed or Contamination Top + Sides < 1000 dpm | Bottom < 3000 dpm | Bottom < 6000 dpm | Remarks |
|---|---|---|---|---|---|---|---|
| 4-16-80 | 6063 | 120 | 20 | ✓ | ✓ | | shipped to 3-700 |
| 4-16-80 | 3060 | 140 | 20 | ✓ | ✓ | | " " |
| 4-16-80 | 3063 | 160 | 30 | — | — | — | vault only |
| 4-17-80 | 3064 | 180 | 50 | ✓ | ✓ | — | vault only |
| 4-17-80 | 3065 | 165 | 15 | — | — | — | Vault only |
| 4/18/80 | 2064 | 120 | 20 | ✓ | — | — | vault only |
| 4/18/80 | 6075 | 115 | 15 | ✓ | — | — | Vault only |
| 4/20/80 | 3066 | 140 | 40 | ✓ | ✓ | ✓ | Vault only |
| 4/23/80 | Pm 3060 | 140 | 40 | ✓ | ✓ | ✓ | Shipped |

Figure 8-11. Survey of plutonium billets to be shipped to Building 321-M (Brown 2014, p. 19).

positive $^{238}$Pu result was obtained, or when requested by Health Physics. According to DuPont (1971d), DuPont workers in HB-Line and 235-F were sampled twice per year by plutonium urinalysis and annually by chest count. From 1978 through 1989, neptunium process workers in Building 235-F were sampled twice per year for plutonium and annually for neptunium intakes. During this period, workers in HB Line and radiation workers in Building 773-A were sampled four times per year for plutonium by urinalysis. Workers in Building 321 M were sampled annually by plutonium urinalysis. From 1990 forward, with SRS implementation of alpha spectroscopy, all F- and H-Area workers were sampled by urinalysis for neptunium intakes.

DuPont (1971d) prescribed bioassay requirements for Construction Division workers: one sample every year for plutonium and a sample when terminating. That requirement is shown in Figure 8-12.

DO NOT REMOVE
FROM SRP WITHOUT APPROVAL

DPSOL 193-302
REV 5 PAGE 3 OF 14

4. BIOASSAY SAMPLING FREQUENCIES — Construction Division

a. Routine Urine Samples

- Fission Products and/or Induced Activity — one sample per year and when terminating.
- Tritium — sample frequency is outlined in Radiation and Contamination Control DPSOP 40-1 or Construction Job Plans.
- Plutonium — one sample every 3 years and when terminating.
- Other Nuclides — as specified by area Health Physics in Construction Job Plans.

[NOTE] Construction Division Medical Department annually provides each employee with a sample bottle and label and instructs the employee to submit a one-liter urine sample. Samples are also obtained from new employees who worked in Radiation Zones at another installation where radioactive materials were handled. Personnel Monitoring will forward requests for resamples through Construction Medical.

b. Special Sampling (See Division B and Construction Division Safety Procedure 58)

Figure 8-12. Bioassay requirements, Construction Division (Dupont 1971d).

While the bioassay control procedures in force from 1971 through 1989 required urine sampling, the ORAU Team has obtained data to demonstrate that neptunium workers were monitored for internal intakes of $^{237}$Np intakes since 1961 (DuPont 1963b, 1964, 1965d, 1966a, 1967a, 1968b, 1970c, 1971e, 1974h, 1975b, 1980a, 1980h, 1980g, 1980e, 1980d, 1984c, 1985a, 1985b, 1985c, 1985d, 1985e; Reinig 1984; Reed 1982). The number of neptunium analyses reported by SRS in Works Technical Department reports and the number of results obtained by the ORAU Team from logbooks are listed in Table 8-3. The number of $^{237}$Np results by area provided in Table 8-4.

Table 8-3. Neptunium urinalysis data.

| Year | No. of Np-237 samples identified in Works Technical Reports | No. of Np-237 Samples Identified in other Pu, Eu, and Am Logbooks |
|---|---|---|
| 1972 | 22 | 20 |
| 1973 | 31 | 17 |
| 1974 | 42 | 18 |
| 1975 | Not listed | 0 |
| 1976 | Not listed | 0 |
| 1977 | Not listed | 0 |
| 1978 | Not listed | 0 |
| 1979 | 11 | 1 |
| 1980 | 48* | 49 |
| 1981 | 57 | 19 |
| 1982 | 146 | 78 |
| 1983 | 22* | 25 |
| 1984 | 37* | 48 |
| 1985 | 13* | 14 |
| 1986 | No summary report | 0 |
| 1987 | No summary report | 0 |
| 1988 | No summary report | 30 |
| 1989 | No summary report | 14 |

* Only partial data currently available for year.

Table 8-4. Number of $^{237}$Np samples by area.

| Year | 235-F | HB Line | 321-M | 773-A/772-F | Other |
|---|---|---|---|---|---|
| 1972 | 20 | 0 | 0 | 0 | 0 |
| 1973 | 16 | 0 | 1 | 0 | 0 |
| 1974 | 17 | 0 | 0 | 1 | 0 |
| 1975 | 0 | 0 | 0 | 0 | 0 |
| 1976 | 0 | 0 | 0 | 0 | 0 |
| 1977 | 0 | 0 | 0 | 0 | 0 |
| 1978 | 0 | 0 | 0 | 0 | 0 |
| 1979 | 0 | 0 | 1 | 0 | 0 |
| 1980 | 36 | 0 | 8 | 3 | 2 |
| 1981 | 3 | 2 | 9 | 3 | 2 |
| 1982 | 36 | 32 | 6 | 3 | 1 |
| 1983 | 12 | 4 | 0 | 7 | 2 |
| 1984 | 23 | 5 | 16 | 4 | 0 |
| 1985 | 7 | 2 | 0 | 3 | 2 |
| 1986 | 0 | 0 | 0 | 0 | 0 |
| 1987 | 0 | 0 | 0 | 0 | 0 |
| 1988 | 5 | 5 | 0 | 16 | 4 |
| 1989 | 3 | 0 | 0 | 9 | 2 |
| **Totals** | **178** | **50** | **41** | **49** | **15** |

In addition to urinalysis data, the ORAU Team has obtained results of in vivo counting which may also be used to reconstruct potential $^{237}$Np doses.

Positive intakes of radionuclides at SRS were recorded and maintained over the years in an intake registry. Through 2002, the registry contained over 5,400 intakes. Of those 5,400 intakes, 14 workers received positive intakes of $^{237}$Np, with 2 workers receiving more than one neptunium intake. Information available for those intakes is presented in Table 8-2 (ORAUT 2008). The first intake was by wound in March 1960 by an operator in F-Area. The same worker received two other neptunium intakes by wound in Building 235-F. The last intake of neptunium recorded in the version of the intake database available to the ORAU Team was also from a wound in May 2000; the location of that incident is not available.

Table 8-2. Neptunium intakes (ORAUT 2008).[a]

| Worker | Date | Type of Assimilation | Area | Job Title | Comments |
|---|---|---|---|---|---|
| 1 | 02/15/1972 | Wound | 235-F | Separations Operator | Exposed to Pu-238 |
| 1 | 07/05/1973 | Inhalation | 235-F | Separations Operator | Exposed to Pu-238 |
| 2 | 11/08/1974 | Inhalation | 235-F | No information | Exposed to Pu-238 |
| 3 | 11/1988 | Wound | CS | Boilermaker | Exposed to Pu-238 |
| 4 | 04/1989 | Inhalation | Old HB Line | Carpenter | Exposed to Pu-238 |
| 5 | 06/1968 | Inhalation | 235-F | Separations Operator | None |
| 6 | 03/1968 | Inhalation | F | Operator | None |
| 6 | 06/1967 | Inhalation | 235-F | Separations Operator | None |
| 6 | 06/1968 | Inhalation | 235-F | Separations Operator | None |
| 7 | 08/1991 | Inhalation | 235-F | Operator | Exposed to Pu-238 |
| 8 | 10/1961 | Inhalation | F | R&D Supervisor | None |
| 9 | 04/1967 | Wound | F | Technician | Possibly 772-F |
| 10 | 03/1991 | Inhalation | No information | No information | D&D period |
| 11 | 12/1998 | Inhalation | No information | No information | D&D period |
| 12 | 08/1999 | Inhalation | No information | No information | D&D period |
| 13 | 05/2000 | Inhalation | No information | No information | D&D period |
| 14 | 09/1999 | Inhalation | No information | No information | D&D period |

a. D&D = decontamination and decommissioning.

Internal doses from potential exposure to $^{237}$Np may be reconstructed using results of neptunium bioassay available for the worker. For unmonitored internal dose, coworker neptunium intakes may be used to derive internal dose. The methods for derivation of the initial neptunium coworker data models are discussed in ORAUT-OTIB-0081, *Internal Coworker Dosimetry Data for the Savannah River Site* (ORAUT 2013d). The ORAU Team used several data sources for the neptunium coworker models. For years before 1970, some neptunium urinalysis bioassay data were obtained from SRS laboratory notebooks (DuPont 1961–1969, 1969g). These data were assumed to be a complete dataset for this period. Neptunium bioassay results available in HPRED were used for 1991 through 1995. The ORAU Team has no neptunium bioassay data for the year 1990. Four data sources were available to develop a coworker model for use in estimating neptunium exposures for the period from 1970 through 1990: (1) limited neptunium bioassay, (2) ratio of neptunium from the plutonium bioassay, given that plutonium activity is between 2 and 10 times greater than the neptunium activity, (3) interpolation, and (4) whole-body count (WBC) data.

For the analysis of the ratio of neptunium from plutonium bioassay, the ORAU Team reviewed monthly issues of SRS Works Technical Reports available for 1968 through 1982. In each monthly report, $^{238}$Pu by weight or weight percent was reported as a primary contaminant in $^{237}$Np feeds from the frames from HB Line and in the product from 235-F. By procedure, SRS considered $^{237}$Np to be contaminated with $^{238}$Pu, generally at 0.01 wt% (100 ppm) or greater. The available records indicate only one instance where SRS produced $^{237}$Np with less than 0.01 wt% $^{238}$Pu. SRS reported that one

special product of 200 g $^{237}$Np with about 10 ppm (0.001%) $^{238}$Pu contamination was produced in September 1974 (DuPont 1974i). By July 1973, SRS mostly reported the minimum weight percent of $^{238}$Pu as less than 0.05% in monthly Works Technical Reports. The concentration of $^{238}$Pu contamination found in $^{237}$Np feeds was reduced starting in spring 1974 with the gradual implementation of a new macroporous resin for anion exchange into 1975 (DuPont 1974c, DuPont 1974e). The use of the new resin resulted in less $^{238}$Pu contamination in the final $^{237}$Np product.

To establish Pu:Np ratios and the plutonium isotopic composition, the ORAU Team collated the information on plutonium contamination in the neptunium product from the Works Technical Department reports (see the DuPont section of the References list). The Pu:Np ratios for 1968 through 1982 (excluding 1971) were analyzed on an annual basis. The plutonium isotopic composition data was evaluated for the entire period. The annual Pu:Np ratios were evaluated to determine the 5th percentile of the ratio (favorable to claimants as is use of a 95th-percentile intake rate) and converted from a mass ratio to an activity ratio using the isotopic composition of the plutonium mixture and the isotopic specific activities.

For each year, random values were drawn from the fitted monthly truncated normal distributions (proportionately to the estimated sample size in each month) to create an annual distribution. The empirical 5th percentiles of the annual distributions of the ratios were used in subsequent analyses (see Figures 8-13 and 8-14). The calculated Pu:Np ratio for 1982 was used for all years after 1982.

The coworker study plutonium intake rates (ORAUT 2013d) were divided by the Pu:Np ratios to determine neptunium intake rates for the period from 1968 through 1990 (see Figure 8-15). There are two plutonium intake rates in the coworker study, one each for types M and S. The corresponding neptunium intake rates for each solubility type are shown, although neptunium itself is only evaluated as a type M material. For comparison, the coworker study intake rates are also shown. During the 1970 to 1990 period when WBC data were used, intake rates based on interpolation of the urinalysis- based coworker intake rates before and after this period are also presented for comparison. The 1980 to 1984 period has more neptunium urinalysis data available than any other portion of the 1970s or 1980s. A neptunium intake rate was calculated in accordance with the normal internal coworker study methodology (ORAUT 2013d, ORAUT 2014). That intake rate is also shown in Figure 8-15 and falls directly along the interpolation line of the neptunium urinalysis-based coworker study intake rates.. Urinalyses data available for 1970, 1972, 1973, 1974, 1985, and 1988 was insufficient to permit calculation of intake rates with sufficient confidence in the calculated excretion rate. The measured excretion rates from these years were included for comparison purposes only. Figure 8-16 shows a plot of these values along with the predicted excretion rates from all the neptunium urinalysis-based intake rates and the individual data points used to calculate these intake rates. The coworker study intake rate for 1968 to 1969 was extrapolated through 1979, and the 1980 to 1984 intake rate calculated above was extrapolated through 1989 to generate the predicted excretion rates. As can be seen, the excretion rates are generally consistent.

The use of neptunium in ratio to plutonium reported in plutonium bioassay results in calculated intake rates substantially lower than the intake rates before and after that time period which are based on actual neptunium urinalysis data.

WBC data was selected as the source data for the coworker model as detailed in ORAUT-RPRT-0056, *A Comparison of Neptunium Coworker Models at the SRS* (ORAUT 2012). The ORAU Team has confirmed that workers in neptunium processing areas were required to have WBCs. Neptunium doses calculated using in vivo data are favorable to claimants but not so unreasonably high as to be deemed insufficiently accurate. While WBC data was selected as the source of neptunium coworker intake distributions, another reasonable approach – and one directly based on neptunium bioassay data – would be to extend the 1968 to 1969 intake rate through 1979, use the calculated 1980 to 1984

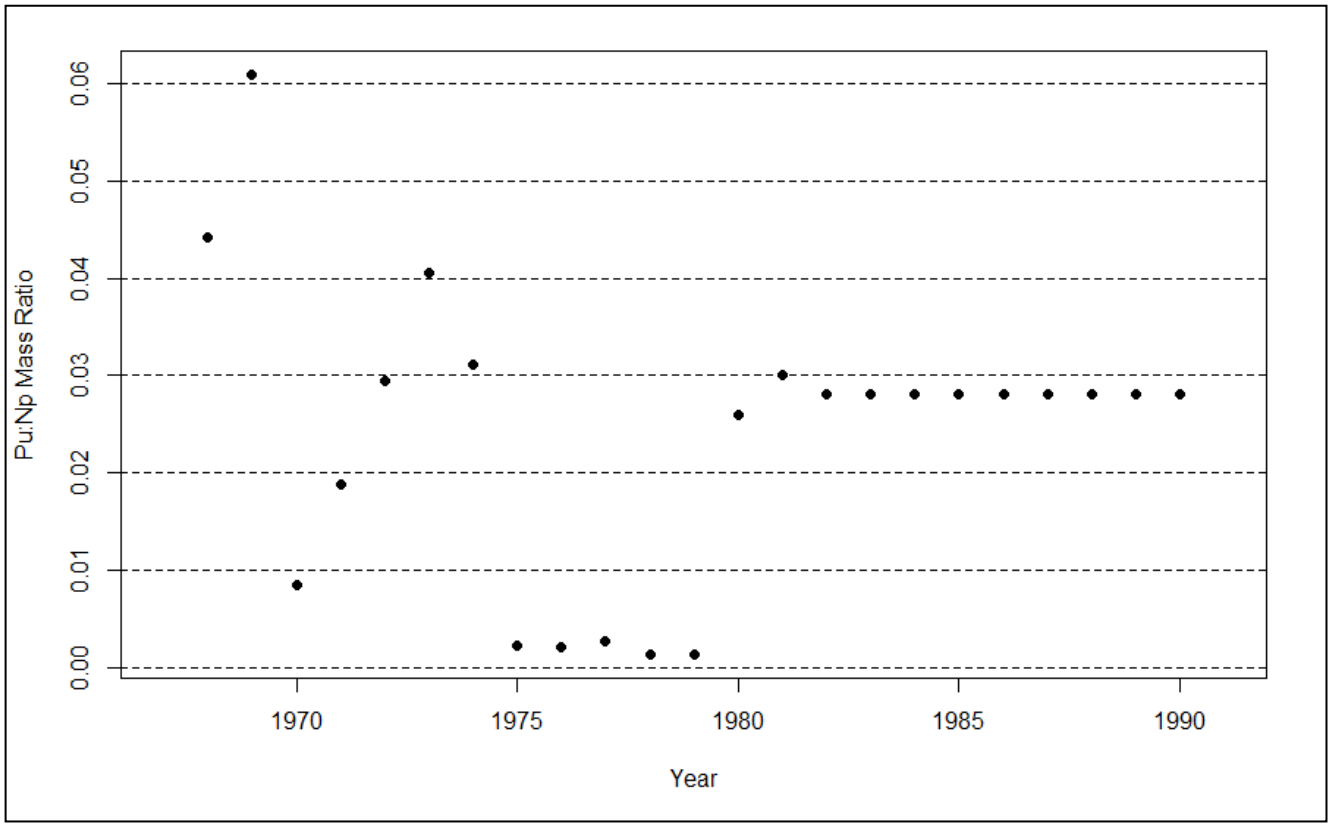



Figure 8-13. Pu:Np mass ratios.

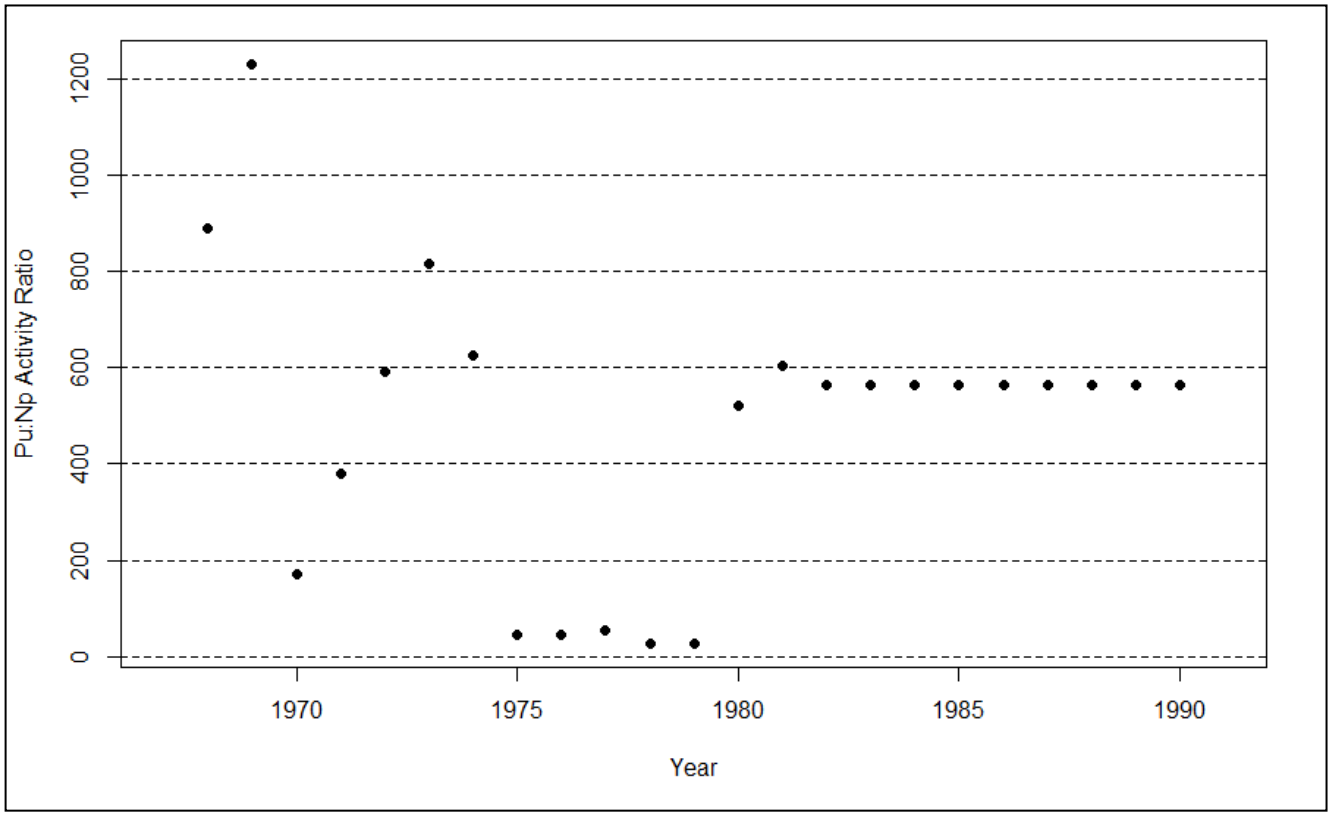



Figure 8-14. Pu:Np activity ratios.

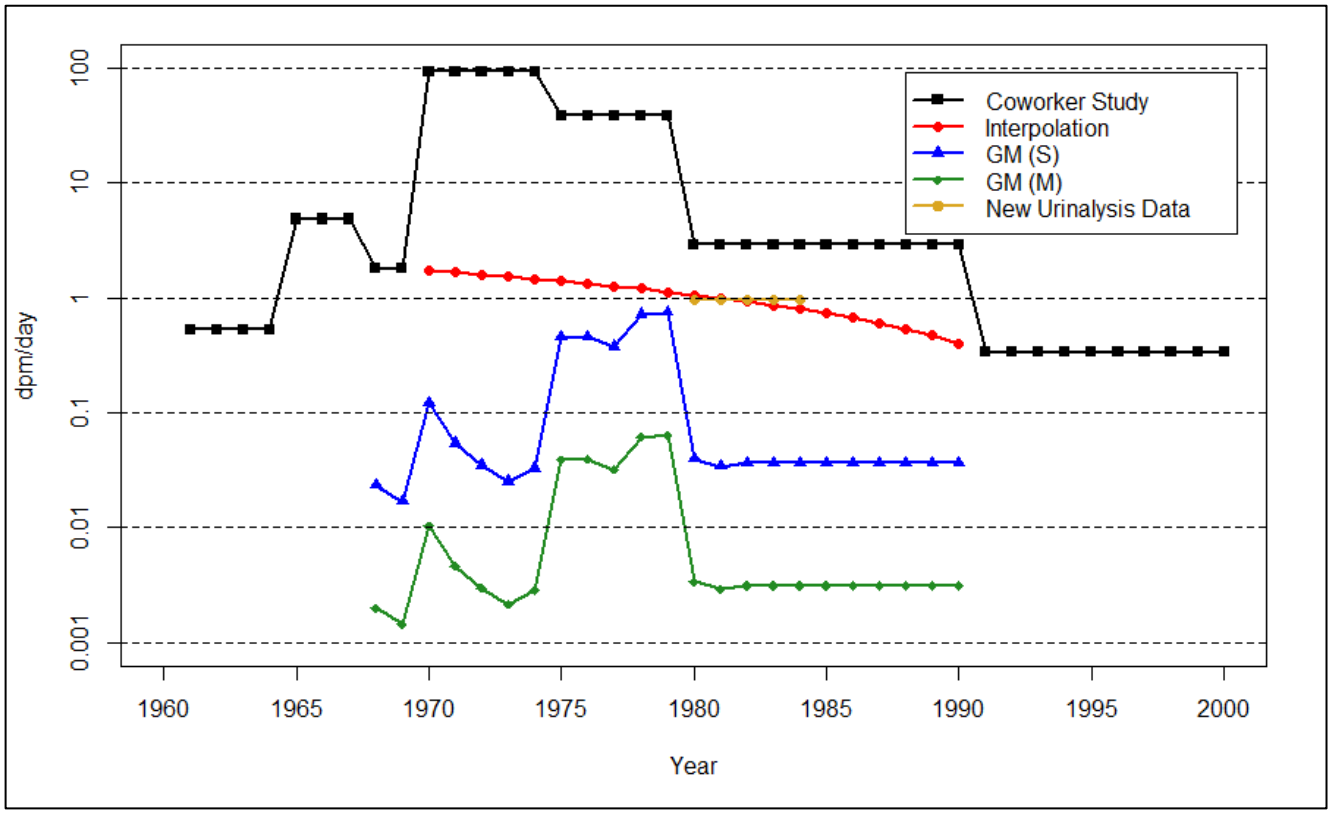



Figure 8-15. Neptunium intake rates.

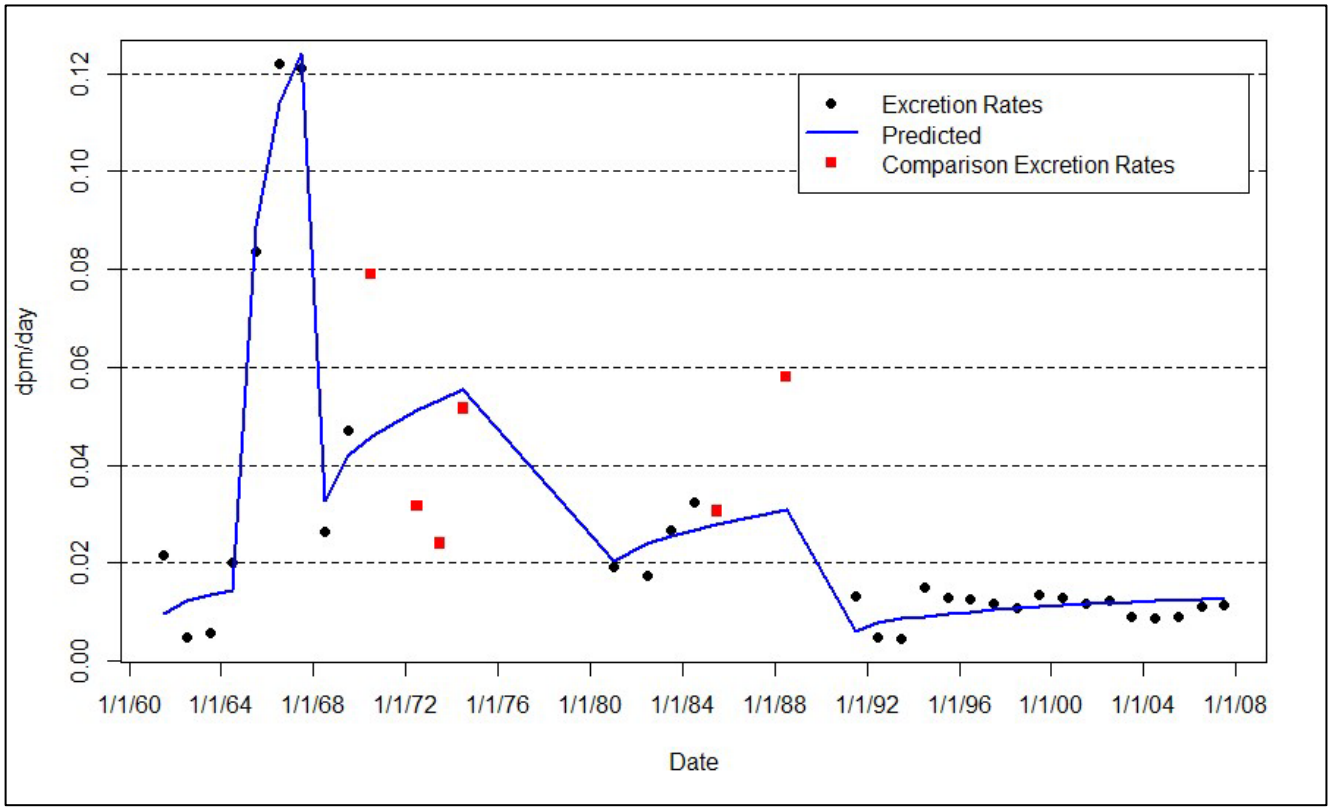



Figure 8-16. Neptunium excretion rates.

neptunium intake rate for that period, then extend the 1980 to 1984 intake rate through 1990. This approach would provide an approximation of the interpolation line that is favorable to claimants, is consistent with gradual improvement in contamination control and worker safety and dose limitation, and is more favorable to claimants in comparison with using intakes based on the Pu:Np ratios.

The ORAU Team analyzed the neptunium coworker data from 1961 through 1989 to determine if there was a statistical difference in stratum from construction trade workers (CTWs) versus non-CTWs. The analysis revealed that the CTW strata did not differ from the strata for all monitored workers between 1961 and 1989 (ORAUT 2012). Final stratified neptunium coworker statistics will be generated using time-weighted one-person, one-sample (TWOPOS) calculation methods documented in ORAUT-RPRT-0053, *Analysis of Stratified Coworker Datasets* (ORAUT 2014), and *Draft Criteria for the Evaluation and Use of Coworker Datasets* (Neton 2015).In summary, doses from neptunium can be bounded using bioassay available for the claimant and from coworker distributions generated for CTWs and non-CTWs using WBC results for 1971 through 1990 and sufficient neptunium urinalysis results for 1991 through 1995.

## 8.4 INCIDENTS

Some incidents involving work with radioactive materials were categorized for official review and tracked in the Special Hazard Incidents (SHI) reports available through 1989 (DuPont 1954–1959, 1962–1974, 1974–1984, 1983–1989). Reports of these incidents were retained in SRS Dosimetry SHI files. In addition to SHIs, SRS investigated incidents resulting from any of the following, as required by DPSOP-40, Special Hazards Bulletin 2 (DuPont 1972b, p. 183):

- Acts or conditions that caused or could have caused radiation or contamination hazards;
- Incidents of contamination that required costly clean-up or that concerned the Health Physics group,
- Incidents that caused internal body contamination or that concerned the Health Physics or Medical groups, and
- Incidents exceeding criticality control limits given in Technical Standards, Operating Procedures, or Test Authorizations.

Bulletin 2 required investigations to be conducted (1) to determine pertinent facts and conditions about unsafe practices or unusual incidents involving radiation or contamination and (2) to recommend measures to prevent to prevent recurrence. The bulletin required any individual who was aware of a circumstance like one of those listed to promptly report it to supervision or Health Physics supervision.

Selected incidents occurring when neptunium was processed are listed in Attachment A, Table A-2. While none of the incidents involved the use of neptunium, they help demonstrate management and Health Physics support and monitoring of locations where neptunium tasks were performed.

The numbers of incidents recorded by location where neptunium was processed or handled, regardless of incident type, are presented in Table 8-5. Several of the incidents recorded for M-Area (Buildings 305-M and 321-M) involved storage and criticality issues. No incidents of the level requiring inclusion in the SRS dosimetry SHI files involved the use of neptunium.

Table 8-5. Special hazard incidents by location.

| Location | 1960s | 1970s | 1980s |
|---|---|---|---|
| Building 305-M | 2 | 2 | 0 |
| Building 321-M | 3 | 10 | 10 |
| Building 773-A | 14 | 13 | 5 |
| Building 235-F | 1 | 0 | 2 |
| Building 772-F | 5 | 3 | 12 |
| HB Line | 1 | 3 | 6 |
| F-Canyon | 10 | 2 | 5 |
| H-Canyon | 8 | 5 | 6 |

The ORAU Team examined a sampling of records in the NIOSH-Division of Compensation Analysis and Support Claims Tracking System (NOCTS) for SRS workers to determine if personal incident records were available where workers received contamination in areas where neptunium was processed.

- April 1966: A technician working in C-155, Building 773-A was not properly wearing rubber gloves while working in a glovebox, resulting in personal contamination. The worker's hand was contaminated, but nasal smears were negative (NOCTS).

- June 1967: Air activity increased to $2 \times 10^{-12}$ µCi/cm$^3$ $^{237}$Np while a worker was changing glovebox cabinet glove ports in Room 160, Building 235-F. Personnel were wearing assault mask (NOCTS). One Separations Operator received neptunium intake (ORAUT 2008).

- March 1968: A glove ruptured during slug line work in Room 107A, Building 235-F (NOCTS). One Separations Operator received a neptunium intake (ORAUT 2008).

- April 1968: A Separations Operator was contaminated when a process cabinet glove failed in Room 107A, Building 235-F. The operator's coveralls were contaminated. Bioassay did not detect an intake of radioactivity (NOCTS).

- June 1968: A Separations Operator was contaminated when a process cabinet glove failed in Room 107A, Building 235-F (NOCTS). Bioassay indicated operator received an intake of $^{237}$Np (ORAUT 2008).

- July 1968: A Separations Operator received a puncture wound on the left ring finger during cabinet glove work in Room 107A, Building 235-F. The wound monitor showed either 621 dpm $^{238}$Pu or 42 dpm $^{237}$Np. The wound was excised by the plant doctor and no intake was detected (NOCTS).

- February 1970: A Separations Operator without respiratory protection erected a containment hut over an area already contaminated in HB Line (NOCTS). The worker received an inhalation intake of $^{237}$Np (ORAUT 2008).

- August 1971: A technician working in L-136, Building 772-F was contaminated while handling contaminated equipment. Bioassay revealed an intake of $^{238}$Pu (NOCTS; ORAUT 2008).

- January 1972: One construction worker continued installation of Pu-Np partitioning equipment in the JT·3 process cabinet in Room 311. Transferable contamination was measured up to 108 dpm/ft$^2$. Gamma radiation exposure rates up to 100 mR/hr were measured in the cabinet. Plastic suits were worn for personnel protection and containment huts were used for contamination control. Even with these precautions, as one employee undressed after work he contaminated his hair to 80,000 dpm and his right cheek to 30,000 dpm. A second

employee contaminated his coveralls to 40,000 dpm when a seam in his plastic suit failed. Bioassay analyses indicated that neither employee assimilated radioactive materials (DuPont 1972f).

- December 1972: A Separations Operator received a puncture wound while attempting to remove a prefilter from the filter housing in the H & V Room, Building 235-F (NOCTS). The worker received a $^{237}$Np intake via the wound.

- July 1973: A Separations Operator was contaminated while welding and decontamination of uranium and neptunium billets in Room 107D, Building 235-F (NOCTS). Follow-up bioassay indicated the worker received an intake of $^{237}$Np (ORAUT 2008).

- November 1974: While working in the Neptunium Compact Operating Room, two Maintenance Mechanics and a Separations Operator received nasal contamination (to 420 dpm, 25 dpm, and 30 dpm, respectively) due to a process cabinet glove failure. The Maintenance Mechanic with the highest nasal contamination also had contamination to 10,000 dpm on his wrist. There were five other persons in the room at the time; their nasal smears were negative. Bioassay sampling for the three persons with positive nasal smears was initiated. A survey of the room identified 1 failed glove and 14 contaminated gloves. The failed glove was contaminated to $8 \times 10^6$ dpm alpha; the other 14 gloves were contaminated with levels to $1 \times 10^5$ dpm. The floor was contaminated up to 10,000 dpm/ft$^2$ (DuPont 1974h).

- August 1975: A maintenance worker was contaminated with plutonium in L-130, Building 772-F. Contamination was measured on the worker's glove. Bioassay for internal intakes was negative (NOCTS).

- November 1978: An airborne release occurred in Building 235-F, room not given. No intakes were detected by bioassay (NOCTS).

- July 1979: A maintenance worker's right forearm was contaminated to 20,000 dpm alpha after a glove failed in Cell 2 in the east maintenance room, Building 235-F. Nasal smears for intake of radioactivity were negative (NOCTS).

- June 1980: After completion of neptunium billet outgassing, transferable contamination to $3 \times 10^6$ dpm alpha/1,000 cm$^2$ was detected on the hood, furnace floor, valves, and manifold fittings. No particulate airborne radioactivity was detected in the work area. Employees wore appropriate respiratory protection. All equipment was decontaminated to less than 500 dpm alpha/1,000 cm$^2$ (DuPont 1980f).

- November 1979: The cladding on an extruded neptunium tube cracked in 321-M. Alpha contamination to $2.0 \times 10^5$ dpm was detected. No transferable or airborne contamination was detected. Radiation exposure rates to 200 mR/hr were measured 45 cm from the tube (DuPont 1979d).

- July 1981: A technician was contaminated after spilling a sample of plutonium in L-127, Building 772-F. Bioassay revealed an inhalation intake of $^{238}$Pu (ORAUT 2008; NOCTS).

- October 1981: A maintenance worker's left wrist was contaminated to 40,000 dpm alpha after tape came from glove in the east maintenance room, Building 235-F. Nasal smears for intake of radioactivity were negative (NOCTS).

- March 1982: A technician working in L-147, Building 772-F was exposed to high airborne radioactivity. Bioassay revealed intakes of $^{238}$Pu, $^{239}$Pu, and $^{241}$Pu (NOCTS).

- April 1982: A technician working in L-126, Building 772-A was contaminated. Bioassay detected an intake of $^{238}$Pu (ORAUT 2008; NOCTS).

- July 1983: A construction worker was contaminated while drilling through a wall in High-Level Cell 16, Building 773-A. Bioassay for intakes was negative (NOCTS).

- January 1984: A technician working in B-142-146, Building 773-A was contaminated after a glovebox glove ruptured. Contamination was detected on the worker's laboratory coat, pants, and hands. Bioassay indicated an intake of $^{244}$Cm (ORAUT 2008; NOCTS).

- February 1988: A maintenance worker was exposed to airborne plutonium in Building 235-F when exhaust fans stopped. Follow-up bioassay did not detect an intake of radioactivity (NOCTS).

- June 1988: A technician working in L-123, Building 772-F was contaminated while mixing a solution as the top came off. Bioassay revealed the worker sustained an intake of $^{238}$Pu (ORAUT 2008; NOCTS).

- February 1989: A maintenance worker was exposed to airborne plutonium in Building 235-F when exhaust fans stopped. Follow-up bioassay did not detect an intake of radioactivity (NOCTS).

- April 1989: A carpenter was briefly exposed to $^{238}$Pu and $^{237}$Np in Room 307 of the old HB Line when the fresh air suit failed momentarily. The worker took off the suit top and shell before he could be stopped. Health Physics quickly intervened putting on a new suit. The worker was placed on fecal and in vivo bioassay, which noted intakes of both $^{238}$Pu and $^{237}$Np (ORAUT 2008; NOCTS).

- August 1989: An operator was exposed to high airborne activity in the old HB Line. Bioassay did not detect an intake of radioactivity (NOCTS).

- April 1990: A construction worker was contaminated while trying to break through the floor in L-143, Building 772F. Bioassay for internal intakes was negative (NOCTS).

- August 1991: Workers were erecting a contamination hut when a plug fell out of the cabinet exhaust duct in Building 235-F neptunium billet maintenance. Bioassay confirmed that one of the workers received intakes of $^{238}$Pu and $^{237}$Np (NOCTS).

- March 1992: A Separations Operator was exposed to airborne plutonium when an airflow reversal occurred in Room 308, old HB Line. Bioassay did not detect an intake of radioactivity (NOCTS).

- September 1992: An operator received 20,000 dpm alpha-beta contamination on his shoe cover in the old HB Line. Bioassay did not detect an intake of radioactivity (NOCTS).

Contamination incidents did occur in neptunium processing areas, some with worker intakes of neptunium or plutonium. However, the available evidence shows that such incidents were monitored by Health Physics with follow-up surveys and bioassay when appropriate.

## 9.0 CONCLUSION

In this report, the ORAU Team details primary and ancillary processes SRS used to separate neptunium from process flows into a nitrate, purify the nitrate product to neptunium oxide, and fabricate

$NpO_2$ into reactor-ready components. Using the available records, the ORAU Team demonstrates that Health Physics provided workplace monitoring of all areas where primary and ancillary work tasks were performed, using data from neptunium and other processes. The preponderance of documentation demonstrates that work with neptunium was designed and planned to prevent or reduce exposures to radiation but that when such exposures occurred health Physics was there to monitor and remediate contamination events. Routine air concentrations were effectively controlled to 5% RCG for neptunium, or less than $2 \times 10^{-13}$ µCi/cm$^3$. When off-normal occurrences occurred SRS took appropriate action with follow-up bioassay. Access to areas where neptunium work was conducted was controlled and required Health Physics approval before entry. In the report, the ORAU Team shows there were no incidents with $^{237}$Np significant enough to be included in the SRS Dosimetry Special Hazards Investigations files.

SRS has routinely monitored neptunium workers for radiation exposure. Potential intakes and internal doses of $^{237}$Np to neptunium workers, those that would have been exposed to neptunium in F- and H-Canyons, HB Line, 235-F, 321-M, 772-F, 773-A, and ancillary locations, can be reconstructed using actual worker data and coworker intakes when monitoring data is missing. Final sets of stratified coworker intakes will be derived using neptunium urinalysis through 1969 and 1990 through 1995, and WBC data through 1989 using TWOPOS calculation methods (ORAUT 2014; Neton 2015). ORAUT-OTIB-0081, *Internal Coworker Dosimetry Data for the Savannah River Site* (ORAUT 2013d), will be revised once the final stratified coworker models are generated.

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# ATTACHMENT A
# HEALTH PHYSICS MONITORING OF SPECIFIC EVENTS

Table A-1. Health physics monitoring of specific events.

| Date | Location | Event |
|---|---|---|
| January 1966 | 773-A, A-122 | A 200-mg source of Np-237 used in Mossbauer studies leaked, resulting in transferable contamination of 10,000 dpm/cm$^2$ on the source holder. The source was subsequently bagged and moved to a hood. No additional contamination was detected (Moyer 1966c). |
| March 1966 | 773-A, B-114 | A liquid sample of Np-237 was spilled in the hood; a small amount splashed on an employee and the laboratory floor. The employee was not wearing protective clothing. An alpha survey of laboratory coat gave 24,000 dpm. The coat was discarded to radioactive waste. The floor had been contaminated to 30,000 dpm/ft$^2$ but was subsequently decontaminated to less than 10 dpm/cm$^2$. Survey of the worker revealed no skin contamination; nasal smears were negative (DuPont 1966b). |
| March 1966 | 773-A, B-114 | A worker detected 6,000 dpm on his shoe after working in the hood (later identified as Np-237 by gamma spectroscopy). An immediate follow-up survey detected transferable contamination of 6,000 dpm/100 cm$^2$ on the floor by the hood. The shoe and floor were cleaned (Moyer 1966d). |
| July 1966 | 773-A, B-133 | After preparing Np-237 source, an employee detected low-level contamination on his shoes (less than 1,000 dpm). A survey indicated contamination of the employee's laboratory coat and rubber gloves. The floor and shoes were cleaned to background and the gloves were discarded to waste (Moyer 1966a). |
| August 1966 | 773-A, A-122 | Np-237 source used in the Mossbauer studies leaked resulting in contamination of the source holder (60,000 dpm), laboratory floor (24,000 dpm), and personal shoes (2,000 dpm). The contaminated items were decontaminated. Nasal smears showed no intakes (Moyer 1966b). |
| September 1966 | 773-A, B-114 | An employee working with Np-237 contaminated his hands, shoes, and personal clothing, and spread contamination to the laboratory floor, personnel corridor, and his office (B-109). The worker's hands were contaminated to 6,000 dpm but cleaned to background. The worker's rubber gloves were contaminated to 9E+05 dpm and were discarded to waste. All work surfaces and equipment were decontaminated to background levels (Moyer 1966e). |
| September 1966 | 773-A, B-065 | An alpha count rate meter and a permanent air sampler were installed when about 500 mg of Np-237 was introduced into a glovebox (Moyer 1966e). |
| October 1967 | 235-F | Radioactive material trapped between retaining rings was released during replacement of a lathe cabinet glove. Floor contamination ranged up to 4E+06 dpm/ft$^2$. An operator and a Health Physics inspector both had contamination in their hair (20,000 dpm and 500 dpm, respectively), but were decontaminated. Respiratory equipment was worn. Airborne radioactivity was 9E-12 Ci/cm$^3$. Special bioassay samples were collected (DuPont 1967b). |
| November 1967 | 235-F | Separations Operators processing Np-237 received exposures above prorated guide values throughout 1967 that necessitated movement of personnel to other facilities before the end of the year. Forecasts of increased production prompted a thorough study of radiation problems and methods of reducing exposure. Options considered were better coordination of process steps and additional shielding (DuPont 1967c). |
| January 1968 | 777-M | Contamination was detected in the Nuclear Physics laboratory after a sealed Np-237 source was removed from the thermal column of the SP reactor. Three graphite plugs were contaminated and were sent to burial ground. The column was decontaminated (Moyer 1968a). |

## ATTACHMENT A
## HEALTH PHYSICS MONITORING OF SPECIFIC EVENTS (continued)

| Date | Location | Event |
|---|---|---|
| May 1968 | 235-F | Four Np-237 foils reading 1,000 mR/hr at 3 in. were transferred from a shipping container to the vault (Copley and Moyer 1968a). |
| July 1968 | HB Line | Neptunium nitrate, produced at Hanford, was mixed with SRS-produced Np-237 nitrate for processing. The Hanford material was apparently contaminated with Th-234 and its progeny, which increased exposure rates from about 20 to 150 mR/hr when the mixture was removed from the line as an oxide (DuPont 1968c). |
| August 1968 | 321-M | Forty Np-Al tubes were fabricated from core compacted billets. The measured maximum radiation dose rates for these were 1,200 mrem/hr gamma with 7 mrem/hr fast neutron. The maximum radiation dose accumulated by an individual during production of the tubes was 110 mrem. Nine of the tubes had fixed contamination to 2,000 dpm alpha on the end cuts. Two of the tubes had transferable contamination to 100 dpm alpha (DuPont 1968d). |
| August 1968 | P Assembly | Gamma exposure does rates to 300 mrem/hr at 18-in. (3 mrem/hr fast neutron) were measured during the assembly of Np-Al tubes into reactor components (DuPont 1968d). |
| October 1968 | 773-A, B-114 | A beaker containing a solution of Np-237 broke inside a hood. Contamination inside the hood was measured at 1.5E+06 dpm, and contamination measured on the floor was 3E+05 dpm/ft$^2$. Both were decontaminated (Copley and Moyer 1968g). |
| April 1969 | 321-M | Twenty Np-Al tubes were fabricated from core compacted billets. No fixed or transferable contamination was detected. The maximum measured dose rate was 500 mrem/hr (2 mrem/hr fast neutron) (DuPont 1969h). |
| June 1969 | 321-M | Thirty-five Np-Al tubes were fabricated from core compacted billets. One tube had a break in the cladding of 4 in.$^2$ during extrusion and was surveyed at 1E+06 dpm. The die on the press read 15,000 dpm transferable and was decontaminated. No other contamination was detected. The process was monitored for airborne contamination, but results revealed no increase during extrusion. The maximum radiation dose received by an individual on all billet extrusions was 200 mrem (Reed 1982). |
| August 1969 | 321-M | Forty-eight Np-Al tubes were fabricated from core compacted billets with typical radiation intensities from the extruded tubes with maximum exposure rate 80 mrem/hr. Breaks in the cladding were observed in three of the tubes. The maximum contamination measured during surveys of these breaks was 30,000 dpm alpha transferable. Ten tubes were fabricated from cores of hot-press-bonded $NpO_2$-Al slugs tubes with maximum exposure rate of 100 mrem/hr. Breaks were observed in the cladding of four of the tubes. The maximum contamination measured during surveys of these breaks was 45,000 dpm transferable. The maximum radiation dose received by an individual worker on all billet extrusions was 715 mrem (Reed 1982). |
| August 1969 | 773-A, D-165 | Two operators who fabricated and decontaminated special neptunium oxide slugs had gamma exposures of 50 mR/hr. The slugs were formed and fabricated into billets with body exposures of 1,465 mR and 810 mR, respectively (Marter 1961). |

## ATTACHMENT A
## HEALTH PHYSICS MONITORING OF SPECIFIC EVENTS (continued)

| Date | Location | Event |
|---|---|---|
| August 1969 | 773-A, D-037 | Machining the aluminum end caps from five neptunium oxide-Al slugs resulted in air activity of 125E-12 µCi/$cm^3$ for a sampled period of thirty minutes. As the core material was lathed, it crumbled and contaminated the lathe, floor, other equipment, and the machinist. Contamination was highest on the lathe at 100,000 dpm/$ft^2$. All equipment and floors were decontaminated to less than 25 dpm/100 $cm^2$. Nasal swabs of the machinist showed some nasal contamination (400 dpm), but a follow-up WBC showed no measureable intake. Subsequent machining in 773-A was moved to a hot cell, D-049 (Moyer 1969b, 1969c). |
| August 1969 | 773-A, D-059 | The exposure rate for fabricating neptunium billets ranged from 12 to 25 mR/hr. Six workers received a total estimated body exposure of 215 mrem. Transferable contamination to 600 dpm/$ft^2$ was spread to the papered laboratory table but was cleaned (Moyer 1969c). |
| September 1969 | 773-A, D-165 | The measured exposure rate for Reactor Engineering personnel to work with Np-Al tubes was 100 mrem/hr. Two employees accumulated a total estimated exposure of 150 mrem. No contamination from the work was measured (Hayes 1969a). |
| October 1969 | 321-M | Twenty Np-Al tubes were fabricated from core compacted billets with typical radiation intensities from the extruded tubes with maximum exposure rate 200 mrem/hr. Six of the tubes had fixed alpha contamination with a maximum 30,000 dpm alpha on the end cuts. The maximum radiation dose received by an individual worker on all billet extrusions was 120 mrem (Reed 1982). |
| November 1969 | 773-A, B-114 | An obsolete glovebox was disconnected and removed from B-114. It contained an estimated 5 mCi Np-237-Am-241. The glovebox was removed with monitoring without incident and sent for burial (Hayes 1969b). |
| January 1970 | 321-M | Thirty-seven tubes were fabricated from cores of hot-press-bonded $NpO_2$-Al slugs with the maximum exposure rate 300 mrem/hr. Contamination was read on 10 of the tubes with a maximum 2,000 dpm on the end cuts (Reed 1982). |
| July 1970 | 321-M | Eleven tubes were extruded from cores of hot-press-bonded $NpO_2$-Al slugs with the maximum exposure rate 600 mrem/hr. The maximum radiation dose received by an individual on all billet extrusions was 285 mrem (Reed 1982). |
| September 1970 | 321-M | Forty-one tubes were extruded from cores of hot-press-bonded $NpO_2$-Al slugs with the maximum exposure rate 360 mrem/hr (Reed 1982). |
| September 1970 | 321-M | Transferable contamination to 6E+05 dpm/$ft^2$ alpha was detected on an Np-Al tube and on the press die when a tube was pulled apart at the draw bench. Air activity was measured at 1E-12 Ci alpha/$cm^3$ immediately following the failure. Two additional tubes had cladding defects with transferable contamination to 1,000 dpm. The maximum transferable contamination on equipment in contact with the tube was 100 dpm/100 $cm^2$ (DuPont 1970c). |
| October 1970 | 321-M | Fixed contamination up to 1E+05 dpm was detected with cladding defects on two Np-Al tubes after drawing; transferable contamination was less than 15 dpm/100 $cm^2$ (DuPont 1970d). |
| December 1970 | 321-M | Forty-three tubes were fabricated from cores of hot-press-bonded $NpO_2$-Al slugs with maximum exposure rate 300 mrem/hr. Fixed contamination was detected at cladding defects on one tube at 4000 dpm. Transferable contamination was measured at less than 15 dpm/100 $cm^2$ (Reed 1982). |

## ATTACHMENT A
## HEALTH PHYSICS MONITORING OF SPECIFIC EVENTS (continued)

| Date | Location | Event |
|---|---|---|
| January 1971 | 321-M | Forty-three tubes were fabricated from cores of hot-press-bonded $NpO_2$-Al slugs with maximum exposure rate 300 mrem/hr. One of the tubes separated during the stretch-straightening process, resulting in transferable contamination to 1,000 dpm on the tube and 8,000 dpm/ft$^2$ on the floor (Reed 1982; DuPont 1971a). |
| March 1971 | 321-M | Ten tubes were fabricated from cores of hot-press-bonded $NpO_2$-Al slugs with maximum exposure rate 300 mrem/hr (Reed 1982). |
| April 1971 | 321-M | Nine tubes were fabricated from cores of hot-press-bonded $NpO_2$-Al slugs with maximum exposure rate 300 mrem/hr (Reed 1982). |
| May 1971 | 321-M | Twenty-seven tubes were fabricated from cores of hot-press-bonded $NpO_2$-Al slugs with maximum exposure rate 700 mrem/hr (Reed 1982). |
| June 1971 | 321-M | Twenty-four tubes were fabricated from cores of hot-press-bonded $NpO_2$-Al slugs with maximum exposure rate 400 mrem/hr (Reed 1982). |
| July 1971 | 321-M | Twenty-four Np-Al tubes were fabricated from cores of hot-press-bonded $NpO_2$-Al slugs with maximum exposure rate 600 mrem/hr (Reed 1982). |
| December 1971 | 321-M | Seventeen tubes were fabricated from cores of hot-press-bonded $NpO_2$-Al slugs with maximum exposure rate 80 mrem/hr unshielded but reduced to 55 mrem/hr with shielding (Reed 1982; DuPont 1971f). |
| January 1972 | 321-M | Ten tubes were fabricated from cores of hot-press-bonded $NpO_2$-Al slugs with maximum exposure rate 140 mrem/hr (Reed 1982). |
| January 1972 | HB Line | One construction worker was installing Pu-Np partitioning equipment in the JT·3 process cabinet in Room 311. Transferable contamination was measured up to 108 dpm/ft$^2$ and gamma radiation exposure rates to 100 mR/hr were measured in the cabinet. Plastic suits were worn for personnel protection and containment huts were used for contamination control. Even with these precautions, as one employee undressed following work, he contaminated his hair to 80,000 dpm and his right cheek to 30,000 dpm. A second employee contaminated his coveralls to 40,000 dpm when a seam in his plastic suit failed. Bioassay analyses indicated that neither employee assimilated radioactive materials (DuPont 1972f). |
| March 1972 | 321-M | Nine tubes were fabricated from cores of hot-press-bonded $NpO_2$-Al slugs with maximum exposure rate 100 mrem/hr unshielded but reduced to 60 mrem/hr with shielding (Reed 1982). |
| May 1972 | 321-M | Twenty tubes were fabricated from cores of hot-press-bonded $NpO_2$-Al slugs with maximum exposure rate 150 mrem/hr unshielded but reduced to 60 mrem/hr with shielding (Reed 1982). |
| December 1973 | 322-M | Two Np-237 tubes were introduced into the new glovebox facility. Exposure dose rates to 250 mrem/hr were detected while workers sawed, grinded, and polished tube sections. Workers received a total exposure of about 1 rem for that work. No contamination control problems were encountered. This work was reported as temporary, with the possibility of becoming routine in the future (DuPont 1973). |
| 1974 | 322-M | Alpha contamination up to 5E+07 dpm/ft$^2$ was detected in the glovebox, Room 126, after cutting two neptunium tubes for metallurgical examination (DuPont 1974b). |

## ATTACHMENT A
## HEALTH PHYSICS MONITORING OF SPECIFIC EVENTS (continued)

| Date | Location | Event |
|---|---|---|
| November 1974 | 235-F | Two maintenance mechanics and a Separations Operator received nasal contamination to 420 dpm, 25 dpm, and 30 dpm, respectively, due to a process cabinet glove failure while working in the Neptunium Compact Operating Room. The maintenance mechanic with the highest nasal contamination also had contamination to 10,000 dpm on his wrist. There were five other persons in the room at the time; their nasal smears were negative. Bioassay sampling for the three persons with positive nasal smears were initiated. A survey of the room one failed glove and 14 contaminated gloves. The failed glove was contaminated to 8E+06 dpm alpha with levels to 1E+05 dpm on the other 14. The floor was contaminated up to 10 dpm/ft (DuPont 1974h). |
| 1975 | 321-M | High external exposure resulted from improper storage of neptunium (Ferrell 1992). |
| November 1975 | HB Line | The Health Physics office and Cold Feed Preparation Room were contaminated on October 3 by a process liquid spill in Process Vacuum Room 454. It was decontaminated and painted. Decontamination efforts in Room 454 and Air Lock 411 generally reduced transferable contamination levels from 4E+08 dpm/ft$^2$ to 5E+04 dpm/ft$^2$ (DuPont 1975c). |
| 1977 | 772-F | A technician contaminated her upper right thigh to 2E+06 dpm and personal shirt to 40,000 dpm when technician dropped a process sample in Lab 158. Skin decontamination efforts were successful. A special bioassay sample was requested (DuPont 1977c). |
| October 1978 | 235-F | An operator working in the neptunium line incurred nasal contamination of 190 dpm when a cabinet glove failed. The glove was contaminated to 1E+04 dpm and the floor to 2E+03 dpm/0.1 m$^2$. Room airborne activity remained less than the RCG during the incident. Nasal irrigation promptly removed the contamination. A follow-up chest count of the operator indicated less than the minimum detectable amount and urine sample results were negative (DuPont 1978f). |
| December 1978 | 321-M | Eight employees collectively received an estimated 900-mrem whole-body exposure while processing 22 neptunium tubes. Exposure rates ranged to 300 mR/hr at 30 cm (DuPont 1978g). |
| January 1979 | 321-M | Worker exposure rates ranged to 300 mR/hr at 30 cm during the extrusion and processing of neptunium tubes. Ten employees collectively received about 1,800·mrem exposure performing the work (DuPont 1979b). Radiation dose rates ranged up to 200 mR/hr at 30 cm from the tubes (DuPont 1979c). |
| November 1979 | 321-M | Alpha contamination to 2E+05 dpm was detected in a 1.5-mm wide, 1.5-cm-long crack on the cladding of an extruded neptunium tube. No transferable or airborne contamination was detected. Gamma exposure rates to 200 mR/hr were measured 45 cm from the tube (DuPont 1979d). |
| June 1980 | 321-M | Transferable contamination to 3E+06 dpm alpha/1,000 cm$^2$ was detected on the hood furnace floor, valves, and manifold fittings after completion of neptunium billet outgassing. No particulate airborne radioactivity was detected in the work area. Employees wore appropriate respiratory protection. All equipment was decontaminated to less than 500 dpm alpha/1,000 cm$^2$ (DuPont 198fd). |

## ATTACHMENT A
## HEALTH PHYSICS MONITORING OF SPECIFIC EVENTS (continued)

| Date | Location | Event |
|---|---|---|
| June 1980 | 772-F | Contamination to 2.5E+05 dpm/100 $cm^2$ was detected on the floor of L-138 following housekeeping inside a laboratory bench where neptunium samples were processed. Contamination ranging from 4,000 to 20,000 dpm/100 $cm^2$ was spread to the utility corridor and three other laboratory modules. Shoe covers of several technicians were contaminated to 20,000 dpm. No airborne activity was detected and nasal smears from personnel were negative. Bioassay samples were collected from five technicians (DuPont 1980f). |
| 1980 | 325-F | A nuclear incident monitor (NIM) alarm occurred in the Neptunium Line Maintenance Room when mechanic checked a gamma monitor adjacent to the NIM without bypassing the NIM as required by procedure. All personnel evacuated the building according to procedure and there was no spread of contamination (DuPont 1980a). |
| August 1980 | 321-M | Exposure rates to 205 mrem/hr (5 mrem/hr fast neutron) were measured at 30 cm from the core section of neptunium tubes during extrusion. Personnel accumulated exposures were approximately 1.5 rem for the extrusion work (DuPont 1980i). |

Table A-2. Special hazards incidents.

| SHI# | Date | Location | Description |
|---|---|---|---|
| 273 | Mar-73 | Building 773-A Decontamination Facility: | A General Service Operator released airborne activity to the decontamination room environment while removing solid radioactive waste from the Decontamination Chamber. An Eberline alpha count rate meter alarmed and alerted him to the presence of alpha activity. He obtained assistance to remove his outer protective clothing and went with a Radiation Control Inspector to the Personnel Decontamination Room. It was found he had a high nasal smear of about 3.6E+04 dpm alpha. Later a whole-body count indicated a positive uptake of curium-244 (DuPont 1962–1974). |
| 283 | Feb-71 | Building 773-A Room B-150 | An Analytical Chemistry Division chemist assimilated some curium while removing samples from storage. Decontamination of the chemist was accomplished under the surveillance of Radiation Control. He was sent to Medical where an aerosol chelation·treatment was given. A chest count and subsequent bioassay data confirmed an uptake of curium less than the maximum permissible body burden (DuPont 1962–1974). |
| 297 | Feb-74 | Building 773-A Room B-061 | A Nuclear Materials Division technician received a tritium assimilation of about 140 µCi/L during a physical inventory of metal specimens that contained absorbed tritium and that were stored in packages in a freezer (DuPont 1962–1974). |
| 304 | Oct-74 | HB Line | Plutonium contamination from a Freon cooling system spread to nearby rooms during maintenance work (DuPont 1974–1984). |
| 310 | Oct-75 | HB Line | Liquid was observed seeping under the door from Room 454 into Air Lock 4llA at 2:00 p.m. on October 3, 1975. The solution transfer vacuum system was immediately shut down. Further inspection revealed liquid seeping through the building expansion joints and down the walls in Room 302 and a section of Corridor 308 (Health Physics Office area) Liquid also spread under transite wall panels into Air Lock 4llA and Room 4l0S (DuPont 1974–1984). |
| 324 | Mar-77 | Building 772-F Laboratory | A decontamination facility attendant received a confirmed Pu-238 assimilation while disposing of excess radioactive solution samples (DuPont 1974–1984). |

## ATTACHMENT A
## HEALTH PHYSICS MONITORING OF SPECIFIC EVENTS (continued)

| SHI# | Date | Location | Description |
|---|---|---|---|
| 330 | Nov-77 | Building 773-A Rooms E-063 and E-065 | Four Laboratory Operations and Services Division employees (two maintenance mechanics, a laboratory technician, and a decontamination facility assistant) were exposed to high concentrations of airborne curium contamination while working in the Manipulator, Decontamination, and Repair Facility in the High Level Caves. Two separate releases of contamination occurred as a result of work being performed on two Manipulator Thru-Tubes from the Target Fabrication Facility. The four employees received nasal contamination and two had indications of internal assimilation (DuPont 1974–1984). |
| 331 | Jan-78 | Building 773-A High Level Caves | A Laboratory Operations and Services Division employee (Laboratory Technician Number 1) unsuspectingly released airborne curium contamination while taking measurements in High-Level Caves Cell Block A. Accompanying him into Cell Block A were three other employees (Laboratory Technician Number 2 and two maintenance mechanics). While taking smear surveys during a follow-up of the incident, a radiation control inspector, accompanied by the first technician, released further airborne curium contamination. The inspector and the technician received nasal contamination and internal assimilations. The other technician and one of the mechanics received skin contamination (DuPont 1974–1984). |
| 343 | Jan-80 | HB Line | When a cabinet exhaust high-efficiency particulate air filter was introduced into the HB-Line Recovery Facility (in its polyvinylchloride bag port bag) it was placed on the top flange of the RD-l scrap dissolver. The dissolver was operating at 120°C, and after approximately 10 min the bag caught fire. The fire was apparently extinguished by the cabinet Halon fire suppression system but reignited several minutes later. It was finally extinguished by the Halon system. Radioactivity releases to the process rooms were minimal, and there was no increase in radioactivity released to the environment during or after the fire. There were no injuries, personnel assimilations, or capital equipment damage resulting from the incident (DuPont 1974–1984). |
| 344 | Feb-80 | Building 773-A Rooms B-147 | A Construction Division sheet metal craftsman removing an installed wooden hood in Laboratory B-147 of Building 773-A noticed dust smeared on the surgeon's gloves he was wearing. Monitoring revealed the presence of contamination. The gloves were removed and Radiation Control was immediately summoned Radiation Control personnel, who effected immediate decontamination of the individual. Subsequent bioassay data confirmed that an assimilation of plutonium estimated at less than 10% of the maximum permissible body burden had occurred (DuPont 1974–1984). |
| 352 | Feb-81 | Building 321-M Machining Room | A raw materials operator had a confirmed assimilation of enriched uranium while working in the machining room during the period February 3, 1981, through March 2, 1981 (DuPont 1974–1984). |
| 358 | Mar-77 | Building 772-F Fan Room | A Power Operator received a low-level plutonium assimilation from airborne contamination when he started a vacuum pump in the 772-F fan room (DuPont 1974–1984). |
| 362 | Sep-81 | Building 772-F Room 156-A | A Laboratories Shift Supervisor received Pu-239 skin contamination when inventorying emission standards in 772-F (DuPont 1974–1984). |
| 365 | Sep-81 | HB Line | A mechanic received a Pu-238 contaminated puncture wound from an aluminum electrical wire while inspecting a failed agitator in a process cabinet (DuPont 1974–1984). |

## ATTACHMENT A
## HEALTH PHYSICS MONITORING OF SPECIFIC EVENTS (continued)

| SHI# | Date | Location | Description |
|---|---|---|---|
| 391 | Oct-83 | HB Line Plutonium Finishing, Room 410 | A mechanic received a Pu-238 assimilation during purge of level dip tubes in Filtrate Catch Tank EP 25 (DuPont 1974–1984). |
| 401 | Jan-84 | Building 773-A Rooms B-142-146 | An Analytical Development Division analyst received Cm-244 skin and nasal contamination while preparing alpha mounts of solution samples submitted by the Target Fabrication Facility. The laboratory was also contaminated. Subsequent bioassay results confirmed a biological uptake by the analyst (DuPont 1983–1989). |
| 414 | Aug-84 | HB Line Room 410 | A construction pipe fitter received a confirmed plutonium assimilation when he ran his contaminated gloved hand under his plastic suit top and removed his cloth hood (DuPont 1983–1989). |
| 440 | Oct-85 | Building 772-F Laboratory L-174 | A technical analyst received a confirmed plutonium assimilation while handling Pu-239 metal samples in preparation for impurity analyses by emission spectroscopy (DuPont 1983–1989). |
| 444 | Jan-86 | HB Line (old) | Two construction electricians, one construction electrical foreman, and one health protection inspector received nasal contamination while the two electricians were drilling a 1-in. hole through a concrete wall. One of the electricians received a confirmed plutonium assimilation (DuPont 1983–1989). |
| 470 | June 1987 | Building 772-F Laboratory L-126 | A Laboratories Department analyst received total nasal contamination of 13,005 dpm Pu-238 while discarding Pu-239 standard solutions in a radiobench. The Pu-239 standards had been stored in a glovebox in L-126, which was contaminated with Pu-238. Bioassay results indicated that the analyst did not receive an uptake (DuPont 1983–1989). |
| 485 | Apr-89 | HB Line (Old) Room 307 | A Separations Operator received a puncture wound to his knee while inadvertently kneeling on a sharp object. Initial surveys conducted in H-Area resulted in no detectable contamination. Follow-up surveys the next day detected minor contamination of the wound site. The wound was then excised. The bioassay program confirmed that no assimilation was experienced (DuPont 1983–1989). |
| 486 | Apr-89 | Building 773-A B-141 Office and B-194 Personnel Corridor | Personal shoes contaminated with 10,000 dpm of plutonium. No other activity was detected. Office and corridor were contaminated to 50,000 dpm/ft$^2$ (DuPont 1983–1989). |