

Evaluation of the Potential for Internal Dose from Np-237 at the Rocky Flats Plant After 1983

White Paper
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INTRODUCTION

In the Special Exposure Cohort Petition (SEC) Evaluation Report for Petition SEC-00192 (SRDB 132777), NIOSH proposed the following class of workers to be added to the SEC:

All employees of the Department of Energy, its predecessor agencies, and their contractors and subcontractors who worked at the Rocky Flats Plant in Golden, Colorado, from April 1, 1952 through December 31, 1983, for a number of work days aggregating at least 250 work days, occurring either solely under this employment or in combination with work days within the parameters established for one or more other classes of employees included in the Special Exposure Cohort.

This recommendation derived, in part, from NIOSH's conclusion that neptunium processing at the Rocky Flats Plant (RFP) took place at least until 1983, and that available monitoring data are insufficient for estimating potential internal exposures to neptunium (Np). Subsequent to that evaluation report, NIOSH has identified evidence that operations involving neptunium may have taken place after 1983, perhaps as late as 1988.

PURPOSE

This white paper summarizes NIOSH's extensive research on neptunium-237 (Np-237) processing at RFP after 1983 (the end of the current SEC period). It includes discussions of Np operations, inventories, available monitoring data, and an evaluation of the potential for internal exposure to Np-237 after 1983.

NATURE OF Np-237 OPERATIONS

A 1981 paper, *Neptunium Processing at the Rocky Flats Plant* (SRDB 24722, PDF p. 6), states that processing included preparation of pure neptunium oxide, metal, and metal alloys as well as Np-237 recovery from a variety of residues. Processes employed included dissolution, anion exchange, precipitation, filtration, calcination, conversion to fluoride, and reduction to metal. Fabrication steps such as casting and rolling were also sometimes performed for the production of high-purity metal shapes and foils. Neptunium was recovered from residual materials including sand, slag, crucibles, casting skulls, and various alloys containing plutonium, tin, uranium, or zirconium. This description of RFP neptunium operations is echoed in a 1984 document, *Actinide Processing at Rocky Flats* (SRDB 24798, PDF p. 323).

Np-237 OPERATIONS FROM 1962 – 1983

Neptunium was processed at Rocky Flats as early as 1962 (SRDB 24722, PDF p. 6). There is no evidence of continuous routine neptunium operations at Rocky Flats; rather, the evidence points to a series of discrete tasks performed from 1962 through 1983, involving a few grams to a few hundred grams, usually processed by request from other DOE laboratories.

The first special-order request for neptunium processing at RFP came from Lawrence Radiation Laboratory, now known as Lawrence Livermore National Laboratory (LLNL), which required high-purity neptunium with gamma-emitting impurities removed. Techniques for purifying neptunium (including the removal of other actinides) were developed as a result of this project. The project also called for preparation of neptunium-plutonium (Pu) alloys, which were prepared by co-reducing Np and PuF₄ mixtures with calcium metal; this effort led to a capability of producing pure Np metal that could be cast with Pu or uranium (U) to form alloys. The first Np-Pu alloy was produced in 1964, and several kilograms of Np metal were produced in subsequent years for preparation of U and Pu alloys.

Other specific projects involving Np-237 processing included the preparation of high-purity neptunium oxide for the Oak Ridge National Laboratory (ORNL) isotope pool, neptunium metal foils for the Savannah River Plant, and neptunium metal disks for use in the liquid-metal, fast-breeder reactor, neutron dosimeter program (SRDB 24722, PDF pp. 6-7).

Based on reviews of numerous reports in the SRDB as well as reviews of classified documents, it is evident that only relatively small quantities of Np-237 were processed at RFP at any given time. The largest single operation appears to have been a special order in 1966 that involved just over 300 grams of neptunium (SRDB 126393).

Np-237 OPERATIONS AFTER 1983

The current SEC period ends on December 31, 1983, as recommended by NIOSH in the SEC-00192 Petition Evaluation Report (SRDB 132777). Since that report was issued, NIOSH has conducted an extensive search for evidence of Np processing after 1983. The result of this effort has been the identification of a single operation described in a 1987 document, *Production-Scale Plutonium-Neptunium Separation and Residue Recovery at Rocky Flats Plant* (SRDB 129512). This document does not make clear exactly when this operation took place, although it does indicate that the campaign lasted approximately one year. NIOSH interviewed one of the authors, who was also the Principal Engineer who designed the process and directed

the activities. He estimated that the operation began around January 1985 (SRDB 130877). This is the only post-1983 Np operation that NIOSH has been able to confirm.

This 1985 operation involved the processing of plutonium scrap containing down to 0.5% (by weight) neptunium to separate and recover the two metals. The feed material for this process consisted of 63,728 grams of Pu containing 232 grams of Np. The separation process involved oxidizing the plutonium residue, passing Pu(III) through an anion exchange resin bed, and leaving Np(IV) behind for subsequent elution, evaporation, denitrification, and calcination to oxide (SRDB 129512, PDF p. 10). The authors reported completion of 24 separations over the course of a year using this process, resulting in the purification of 58,282 grams of plutonium by the removal of 222 grams of neptunium (~96% recovery). NIOSH re-interviewed the co-author/Principal Engineer who stated that project personnel consisted of five experimental operators who performed the glove box work and the Principal Engineer (SRDB 138682).

The final purified Pu that resulted from this operation contained only 0.0069% Np. The “neptunium product” consisted of 1429 grams of Pu and 222 grams of Np, a Pu:Np mass ratio of 6.4 (SRDB 129512, PDF p. 11). This operation involved no purified neptunium; the dose from any internal exposure would have been dominated by the overwhelming amount of plutonium in the mixture, making neptunium bioassay unnecessary. Given the much greater specific activity of Pu-239, Pu bioassay would account for all organ doses, of which Pu would be the dominant component. An evaluation of the relative dose contribution from Pu and Np resulting from an inhalation intake of the Pu-Np mixture is presented later in the section titled *Potential for Np-237 Exposures After 1983*.

According to the Principal Engineer and co-author of the 1987 report, separation of Pu and Np from alloys ended in 1987 (SRDB 130877, PDF p. 5). This statement is consistent with another reference that states that the neptunium program at RFP began in 1964 and was terminated in 1988 (SRDB 33009, PDF p. 75).

Np-237 INVENTORY TOTALS AND MEASURED QUANTITIES

Documents captured by NIOSH and interviews conducted with former employees indicate that neptunium was present at RFP from 1962 to 2003. From 1963 through fiscal year 2003, neptunium quantities ranging between 29 grams and 1,318 grams are recorded in Material Balance Area accounts (SRDB 33009, PDF pp. 75-79; 126758; 131390). Neptunium-237 inventory amounts at RFP from 1963-2003, obtained from the U.S. Department of Energy/U.S. Nuclear Regulatory Commission Nuclear Materials Management & Safeguards System (NMMSS), are shown in Table 1. The neptunium program was reportedly terminated in 1988

(SRDB 33009, PDF p. 75), although neptunium remained in the RFP nuclear material inventory until 2003. Nuclear material hold-up surveys were conducted sometime after 1990 in response to concerns by the Defense Nuclear Facilities Safety Board regarding nuclear Material-Unaccounted-For at Rocky Flats. These surveys identified gram quantities of residual neptunium in previously-cleaned-out glove boxes located in Building 771, Rooms 153 and 182 (SRDB 131225, PDF p. 6).

Fiscal Year	Np-237 (g)	Fiscal Year	Np-237 (g)	Fiscal Year	Np-237 (g)
1963	29	1976A*	468	1990	937
1964	601	1977	458	1991	941
1965	1,292	1978	567	1992	941
1966	740	1979	492	1993	941
1967	1,215	1980	744	1994	941
1968	972	1981	486	1995	941
1969	1,190	1982	699	1996	935
1970	1,105	1983	869	1997	926
1971	1,318	1984	1,040	1998	930
1972	788	1985	931	1999	880
1973	768	1986	985	2000	900
1974	470	1987	995	2001	886
1975	485	1988	970	2002	636
1976*	485	1989	935	2003**	501

Source: SRDB 33009, PDF p. 76; 126758; 131390

* Inventory values reported for FY-1976 and FY-1976A correspond with a change in the federal fiscal year to begin October 1. The U.S. budget year began July 1 prior to this time (SRDB 129510, PDF p. 2; 129484, PDF p. 8). The values here agree with those reported elsewhere (SRDB 126758) for the June 30, 1976 and September 30, 1976 quarterly inventory periods. ** Last reported inventory was on June 30, 2003.

It is difficult to draw hard conclusions about neptunium processing from the inventory table above. The data are presented primarily to show that the total Np inventory at RFP was always relatively low, typically less than a kilogram (compared to much greater inventories of plutonium for the same time period). For example, the Rocky Flats TBD states that Building 771 recovery operations were processing 240 kg of Pu daily by 1965. This daily Pu throughput exceeds any single Np processing run by two to three orders of magnitude (SRDB 30013, PDF p. 15). A look at the year-to-year values reveals that inventory variability was higher in the earlier years and more stable from the mid-1980s onward.

MEASUREMENTS OF RFP NP WASTE AT INEEL

As shown later in Table 5, Rocky Flats shipped neptunium to several DOE sites, including Np-containing waste to the Idaho National Engineering and Environmental Laboratory (INEEL). A recent interviewee (SRDB 138666) identified two volumes of a 2004 document titled *Validation of the Rocky Flats Plant Radionuclide Inventory in the Historic Data Task Using SWEPP Assay Data* (SRDB 104511; 138530). Volume 1 of that document (SRDB 104511) provides non-destructive assay (neutron and gamma) measurements of Rocky Flats Plant waste stored at INEEL’s Radioactive Waste Management Complex (RWMC). Table 2 presents measurements showing that, in drums containing Np-237, Pu-239 was also present at Pu-239/Np-237 mass ratios ranging from 105 to 6,450. Table 3 presents measurements showing that the Pu-239/Np-237 mass concentration ratios ranged from 109 to 5,820. The predominance of Pu-239 in the waste reflects the prevailing ratios in the Rocky Flats operating environment where neptunium was processed. Potential organ doses to workers packaging and handling this waste would, as previously discussed, be dominated by the plutonium contribution.

Table 2: Mean Masses and Ratios of Pu-239 and Np-237 in Rocky Flats Waste Drums

Waste Matrix	Pu-239			Np-237			Pu-239/Np-237 Isotopic Mass Ratio
	<i>n</i>	Mean (g)	σ (g)	<i>n</i>	Mean (g)	σ (g)	
Graphite	1,307	2.60×10^1	2.75×10^1	4	4.03×10^{-3}	5.78×10^{-5}	6.45×10^3
Filters	68	2.76×10^1	3.16×10^1	54	5.07×10^{-3}	3.19×10^{-3}	5.44×10^3
Mixed Metals	523	1.46×10^1	3.14×10^1	14	1.82×10^{-2}	3.64×10^{-2}	8.02×10^2
1 st and 2 nd Stage Sludge	3,095	6.17	8.74	281	5.86×10^{-2}	8.01×10^{-2}	1.05×10^2

Source: SRDB 104511: graphite (PDF p. 66); filters (PDF p. 74); mixed metals (PDF p. 80); 1st and 2nd Stage Sludge (PDF p. 86)
 Note: Data in this table are compiled from measurements of radioactive waste stored at the INEEL RWMC. The symbols *n* and σ are the number of measured drums and standard deviation, respectively, for the log-transformed data.

Table 3: Mean Mass Concentrations and Ratios of Pu-239 and Np-237 in Rocky Flats Waste Drums

Waste Matrix	Pu-239			Np-237			Pu-239/Np-237 Isotopic Mass Concentration Ratio
	<i>n</i>	Mean (g/kg waste)	σ (g/kg waste)	<i>n</i>	Mean (g/kg waste)	σ (g/kg waste)	
Graphite	1,307	3.67×10^{-1}	4.16×10^{-1}	4	6.31×10^{-5}	5.78×10^{-5}	5.82×10^3
Filters	68	1.06	1.03	54	2.96×10^{-4}	3.80×10^{-4}	3.58×10^3
Mixed Metals	523	2.43×10^{-1}	4.75×10^{-1}	14	6.92×10^{-4}	1.99×10^{-3}	3.51×10^2
1 st and 2 nd Stage Sludge	3,095	3.44×10^{-2}	5.10×10^{-2}	281	3.15×10^{-4}	4.05×10^{-4}	1.09×10^2

Source: SRDB 104511: graphite (PDF p. 68); filters (PDF p. 75); mixed metals (PDF p. 81); 1st and 2nd Stage Sludge (PDF p. 87)
 Note: Data in this table are compiled from measurements of radioactive waste stored at the INEEL RWMC. The symbols *n* and σ are the number of measured drums and standard deviation, respectively, for the log-transformed data.

INTERNAL DOSE MONITORING

Captured documents contain only two reported neptunium bioassays. A urinalysis result in July 1966 was “Below Significant Level” (SRDB 24691, PDF p. 74; 26259, PDF p. 3). A urinalysis result in August 1966 reported 0.9 dpm/24h; a follow-up body count showed no detectable uptake (SRDB 24691, PDF p. 78; 26259, PDF p. 5).

WORKPLACE AIR MONITORING

NIOSH has found no neptunium-specific workplace air monitoring.

CONTAINMENT MEASURES EMPLOYED DURING Np-237 OPERATIONS

The neptunium processing descriptions described in the 1981 document, *Neptunium Processing at the Rocky Flats Plant* (SRDB 24722) supports NIOSH’s understanding that, by 1983, all Np operations were being performed in glove box facilities. For example, the document states:

A typical glove box for aqueous Np processing consisted of a “wet” section (for aqueous processes) and a “dry” section (for calcining precipitates and weighing powders) separated by an air lock. Each section had separate air inlet and exhaust filters. A door in the air lock was used to pass equipment and material between the wet and dry sections. A 1/8-inch-thick lead sheet was bonded to the stainless steel portion of the glove box and 1/4-inch leaded glass was placed over the glove box windows as a shield against gamma radiation from the neptunium and its impurities (SRDB 24722, PDF pp. 7-8).

The single neptunium processing operation identified by NIOSH after 1983 involved the separation of neptunium from plutonium residues (see *Np-237 Operations After 1983*). According to the Principal Engineer who designed the process and directed activities, this operation was performed in glove boxes and tanks. The tanks contained feed material (plutonium and neptunium nitrate solution) that was piped directly into the glove boxes (SRDB: 138682).

WORKPLACE INCIDENTS

No radiological incidents at RFP involving neptunium have been identified.

Np-237 SHIPMENT AND RECEIPT DATA

NIOSH obtained and analyzed NMMSS records of neptunium shipments and receipts by Rocky Flats from 1983 until site closure in 2003 (SRDB 131384; 131387; 131388; 131389). Table 4 shows the fiscal years during which grams of neptunium were received at RFP from other DOE sites (LANL, SRS, ORNL, and LLNL). No material was received after FY 1986. The three-letter designations in parentheses after the site acronyms are the DOE Reporting Identification Symbol (RIS) codes for the sites. Neptunium shipments from Rocky Flats to other DOE sites are shown in Table 5 on the next page.

Table 4: Neptunium Receipts (g) by Rocky Flats				
Fiscal Year	Originating Site			
	LANL (AUA)	SRS (DZA)	ORNL (FZC/FZG)	LLNL (LZB)
1983	---	556	199	105
1984	---	---	199	1
1985	99	---	---	228
1986	1	113	---	---

(Continued)

Table 5: Neptunium Shipments (g) from Rocky Flats								
Fiscal Year	Receiving Site							
	LANL (AUA)	SRS (DZA)	ORNL (FZC/FZG)	Hanford (HRA)	LLNL (LZB)	Nevada Test Site (NAB)	WIPP (VPA)	INEEL Waste (VSB)
1983	---	---	378	---	220	40	---	---
1984	---	---	---	---	159	---	---	---
1985	---	---	---	---	146	26	---	22
1986	73	---	---	2	---	---	---	---
1987	1	---	---	---	---	---	---	---
1988	2	8	---	---	---	---	---	---
1989	---	36	---	---	---	---	---	---
1990	---	---	---	---	---	---	---	---
1991	---	---	---	---	---	---	---	---
1992	---	---	---	---	---	---	---	---
1993	---	---	---	---	---	---	---	---
1994	---	---	---	---	---	---	---	---
1995	---	---	---	---	---	---	---	---
1996	---	---	---	---	---	---	---	---
1997	5	---	---	---	7	---	---	---
1998	---	---	---	---	---	---	---	---
1999	---	46	---	---	---	---	1	---
2000	---	18	---	---	---	---	---	---
2001	---	---	---	---	---	---	174	---
2002	---	297	---	---	---	---	324	---
2003	---	394	---	---	---	---	11	---

POTENTIAL FOR Np-237 EXPOSURES AFTER 1983

In the SEC-00192 Petition Evaluation Report (SRDB 132777), NIOSH acknowledged that from 1962 through 1983 there existed the potential for unmonitored worker exposures to Np-237. NIOSH further concluded that there was insufficient data available to estimate intakes of Np-237 during that time period.

For the post-1983 period, NIOSH has identified only one operation involving neptunium (see *Np-237 Operations After 1983*). Workplace monitoring for this project included continuous air monitoring (CAM) and contamination surveys typical of the plutonium processing areas. Radiation Monitors (technicians) were available to provide radiological safety support. Personnel involved in the project were on bioassay programs typical of the plutonium processing environment (i.e., routine urinalyses and body counts) (SRDB 138682).

The previous discussion also noted that this single post-1983 operation did not involve purified Np, but rather Pu with Np as a contaminant. Even the most highly-concentrated Np product produced by this separation process was still mostly Pu, with a Pu:Np mass ratio of 6.4. Since the specific activity of Pu-239 is about 90 times greater than that of Np-237, the activity ratio of this Pu:Np mixture is greater than 500. As a result, all organ doses resulting from an intake from such a mixture would be dominated by the Pu component. The Np dose component would comprise only about 0.1% for Type M Pu for most organs, and only about 1% for Type S Pu (SRDB 137075).

As mentioned previously, NIOSH conducted two interviews with the Principal Engineer/co-author of the 1987 document, *Production-Scale Plutonium-Neptunium Separation and Residue Recovery at Rocky Flats Plant* (SRDB 129512). As Principal Engineer, he was closely involved with the 1985 Pu-Np separation operation under discussion. During those interviews, he stated that everyone who worked in a hot building had a urinalysis every few months as well as annual whole body counts (SRDB 130877; 138682). If this individual or any of his co-workers had received Np intakes from this operation, the resulting organ doses would be adequately accounted for by the Pu bioassay program. Pu bioassay data are readily available for RFP claimants.

NIOSH has not identified any specific operations involving neptunium other than the 1985 operation. If other operations of a similar nature (i.e., Np associated with Pu) did take place, Pu bioassay would have similarly accounted for any doses due to Np.

CONCLUSIONS

In conclusion, NIOSH finds no evidence that Np-237 intakes occurred at RFP after December 31, 1983. If intakes had occurred during this period from the single identified Np operation, the resulting organ doses would be adequately accounted for by the available Pu bioassay data.

A recent interviewee provided to NIOSH additional names of individuals who may be knowledgeable about RFP Np operations (SRDB 138666). NIOSH will follow-up on these individuals and continue to evaluate any new information that becomes available regarding Np operations after 1983. If such information suggests that intakes of purified Np may have occurred during this time, NIOSH will reconsider its position, as appropriate. For the 1962 through 1983 period, NIOSH maintains its existing conclusion, as stated in the SEC-00192 Petition Evaluation Report (SRDB 132777), that unmonitored Np intakes may have occurred and insufficient data are available for estimating resulting doses.

REFERENCES

- SRDB 24691: *Status Reports: Industrial Hygiene & Bio-Assay – June and July, 1966*
- SRDB 24722: *Neptunium Processing at the Rocky Flats Plant*
- SRDB 24798: *Actinide Processing at Rocky Flats; Chapter 15: Neptunium Recovery Process*
- SRDB 26259: *Special Analyses Logbook*
- SRDB 30013: *Technical Basis Document: Rocky Flats Plant – Site Description*
- SRDB 33009: *Summary of Rocky Flats Plant Waste Buried in the Subsurface Disposal Area: Section 10. Special-Order Work*
- SRDB 104511: *Validation of the Rocky Flats Plant Radionuclide Inventory in the Historic Data Task Using SWEPP Assay Data, Volume 1*
- SRDB 126393: *Research and Development Quarterly Progress Report for October, November, and December 1966 – Chemistry and Physics*
- SRDB 126758: *RFP MT82 Inventories Export to Excel*
- SRDB 129484: *Fiscal Year 2012, Historical Tables, Budget of the U.S. Government*
- SRDB 129510: *Documented Communication with [name redacted] on Neptunium Inventory Values at Rocky Flats*
- SRDB 129512: *Production-Scale Plutonium-Neptunium Separation and Residue Recovery at Rocky Flats Plant*
- SRDB 130877: *Documented Communication SEC-00192 with [name redacted] on Neptunium Handling at Rocky Flats*
- SRDB 131225: *Documented Communication SEC-00192 with [name redacted] on Neptunium Handling at Rocky Flats*
- SRDB 131384: *RIS Code ARF Np Shipments and Receipts 1983 to 1988*

SRDB 131387: *RIS Codes ARFH Np Shipments and Receipts January 31, 1983*

SRDB 131388: *RIS Code ORF Np Shipments and Receipts 1990 to 2003*

SRDB 131389: *RIS Code ORW Np Shipments and Receipts 2002 to 2003*

SRDB 131390: *Np Inventory 1989 to 2003, All RIS Codes*

SRDB 132777: *SEC Petition Evaluation Report for Petition SEC-00192, Rocky Flats Plant*

SRDB 137075: *PDF of Excel Spreadsheet Calculations from [name redacted] SEC-00192 Rocky Flats Neptunium*

SRDB 138530: *Validation of the Rocky Flats Plant Radionuclide Inventory in the Historic Data Task Using SWEPP Assay Data, Volume 2*

SRDB 138666: *Interview on Neptunium with [name redacted]*

SRDB 138682: *Documented Communication SEC-00192 Re-interview with [name redacted] on Neptunium at Rocky Flats*