

ORAU TEAM Dose Reconstruction Project for NIOSH

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ACRONYMS AND ABBREVIATIONS

AEC	U.S. Atomic Energy Commission
ARHCO	Atlantic Richfield Hanford Company
CF	commercial fuel
CFR	Code of Federal Regulations
CSREX	Cesium Strontium Rare Earth Extraction
Ci	curie
cpm	counts per minute
D&D	decontamination and decommissioning
DU	depleted uranium
DOE	U.S. Department of Energy
DOL	U.S. Department of Labor
dpm	disintegrations per minute
EEOICPA	Energy Employees Occupational Illness Compensation Program Act of 2000
FFTF	Fast Flux Test Facility
FP	fission product
ft	foot
g	gram
gal	gallon
HEPA	high-efficiency particulate air
HEU	highly enriched uranium
hr	hour
HTLTR	High Temperature Lattice Test Reactor
HTR	High Temperature Reactor
ICRP	International Commission for Radiological Protection
in.	inch
INL	Idaho National Laboratory
kVp	applied kilovoltage; peak kilovoltage
kCi	kilocurie
kg	kilogram
kW	kilowatt
L	liter
Ib	pound
m	meter
MCi	megacurie
mCi	millicurie
MeV	megaelectron-volt
mg	milligram
mi	mile
mL	milliliter
mrem	millirem
MW	megawatt

n	neutron
nCi	nanocurie
NIOSH	National Institute for Occupational Safety and Health
NU	natural uranium
ORAU	Oak Ridge Associated Universities
ORNL	Oak Ridge National Laboratory
pCi PCTR PFP PFPP POC PNNL ppb ppm PRCF PRF PRF PRTR PUREX PUREX 1WW	picocurie Physical Constants Test Reactor Plutonium Finishing Plant Plutonium Fuels Pilot Plant probability of causation Pacific Northwest National Laboratory parts per billion parts per million Plutonium Recycle Critical Facility Plutonium Reclamation Facility Plutonium Recycle Test Reactor Plutonium-Uranium Extraction (Plant) First cycle waste from the PUREX Plant
QA	quality assurance
R&D	research and development
RECUPLEX	recovery of uranium and plutonium by extraction
REDOX	Reduction-Oxidation (Plant)
RG	Rubber Glove (Line)
RMA	Remote Mechanical A (Line)
RMC	Remote Mechanical C (Line)
RU	recycled uranium
s	second
SRDB Ref ID	Site Research Database Reference Identification (number)
SRS	Savannah River Site
TBD	technical basis document
THOREX	thorium extraction
TRIGA	Training, Research, Isotopes, General Atomics (Reactor)
TTR	Thermal Test Reactor
U.S.C.	United States Code
W	watt
wt%	weight percent
α	alpha particle
μCi	microcurie
μg	microgram
§	section or sections

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2.1 INTRODUCTION

Technical basis documents and site profile documents are not official determinations made by the National Institute for Occupational Safety and Health (NIOSH) but are rather general working documents that provide historic background information and guidance to assist in the preparation of dose reconstructions at particular sites or categories of sites. They will be revised in the event additional relevant information is obtained about the affected site(s). These documents may be used to assist NIOSH staff in the completion of the individual work required for each dose reconstruction.

In this document the word "facility" is used as a general term for an area, building, or group of buildings that served a specific purpose at a site. It does not necessarily connote an "atomic weapons employer facility" or a "Department of Energy [DOE] facility" as defined in the Energy Employees Occupational Illness Compensation Program Act [EEOICPA; 42 U.S.C. § 7384I(5) and (12)]. EEOICPA defines a DOE facility as "any building, structure, or premise, including the grounds upon which such building, structure, or premise is located … in which operations are, or have been, conducted by, or on behalf of, the Department of Energy (except for buildings, structures, premises, grounds, or operations … pertaining to the Naval Nuclear Propulsion Program)" [42 U.S.C. § 7384I(12)]. Accordingly, except for the exclusion for the Naval Nuclear Propulsion Program noted above, any facility that performs or performed DOE operations of any nature whatsoever is a DOE facility encompassed by EEOICPA.

For employees of DOE or its contractors with cancer, the DOE facility definition only determines eligibility for a dose reconstruction, which is a prerequisite to a compensation decision (except for members of the Special Exposure Cohort). The compensation decision for cancer claimants is based on a section of the statute entitled "Exposure in the Performance of Duty." That provision [42 U.S.C. § 7384n(b)] says that an individual with cancer "shall be determined to have sustained that cancer in the performance of duty for purposes of the compensation program if, and only if, the cancer ... was at least as likely as not related to employment at the facility [where the employee worked], as determined in accordance with the POC [probability of causation¹] guidelines established under subsection (c) ..." [42 U.S.C. § 7384n(b)]. Neither the statute nor the probability of causation guidelines (nor the dose reconstruction regulation, 42 C.F.R. Pt. 82) define "performance of duty" for DOE employees with a covered cancer or restrict the "duty" to nuclear weapons work (NIOSH 2007).

The statute also includes a definition of a DOE facility that excludes "buildings, structures, premises, grounds, or operations covered by Executive Order No. 12344, dated February 1, 1982 (42 U.S.C. 7158 note), pertaining to the Naval Nuclear Propulsion Program" [42 U.S.C. § 7384I(12)]. While this definition excludes Naval Nuclear Propulsion Facilities from being covered under the Act, the section of EEOICPA that deals with the compensation decision for covered employees with cancer [i.e., 42 U.S.C. § 7384n(b), entitled "Exposure in the Performance of Duty"] does not contain such an exclusion. Therefore, the statute requires NIOSH to include all occupationally-derived radiation exposures at covered facilities in its dose reconstructions for employees at DOE facilities, including radiation exposures related to the Naval Nuclear Propulsion Program. As a result, all internal and external occupational radiation exposures are considered valid for inclusion in a dose reconstruction. No efforts are made to determine the eligibility of any fraction of total measured exposures to be occupationally derived (NIOSH 2007):

Background radiation, including radiation from naturally occurring radon present in conventional structures

The U.S. Department of Labor (DOL) is ultimately responsible under the EEOICPA for determining the POC.

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Radiation from X-rays received in the diagnosis of injuries or illnesses or for therapeutic reasons

2.1.1 Purpose

The purpose of this TBD is to provide a description of the Hanford Site that contains technical basis information for use by the Oak Ridge Associated Universities (ORAU) Team to evaluate the total occupational dose for EEOICPA claimants. This TBD is part of the Hanford Site Profile; it describes the facilities and processes, the use of radionuclides significant for dose reconstruction, and historical information about the programs and operations on the Hanford Site. It contains technical information to assist dose reconstructors in the reconstruction of radiological doses workers might have received dating back to the 1940s.

The Hanford Site played an important role in the development of the U.S. nuclear weapons program. During the Manhattan Project, the site was the Hanford Engineering Works; other names during the site's history include the Hanford Works, Hanford Reservation, and most recently Hanford Site. This document refers to the Hanford Site or simply Hanford. Operations at the site included the construction of nine reactors to produce plutonium for weapons and seven physical testing, research, and demonstration reactors. Facilities for the separation of uranium and plutonium, for uranium and tritium extraction, and for many support functions evolved over the years.

2.1.2 <u>Scope</u>

Section 2.2 provides the history of the Hanford Site facilities and the activities that took place in them. Section 2.3 provides the history of certain, but not all, processes in relation to radionuclides or to special mixtures [e.g., highly enriched uranium (HEU)] that were in addition to and generally on a much smaller scale than the main processes that were involved in creation and extraction of plutonium and recycling of uranium. There is some redundancy between the two parts. Attachment A is a condensed summary of the activities and radioactive material work at the Hanford Site facilities.

Attributions and annotations, indicated by bracketed callouts and used to identify the source, justification, or clarification of the associated information, are presented in Section 2.4.

2.2 SITE ACTIVITIES AND PROCESSES

Construction of facilities at the Hanford Site, an area of approximately 600 mi², commenced in March 1943, when several major nuclear facilities were started. These included 105-B (Production Reactor), 221-T (Separation Facility), 305 (Test Pile), and 313 (Fuel Element Fabrication). The production reactors, designed to produce plutonium by irradiating metallic uranium, were constructed in the 100 Areas along the Columbia River on the north side of the site. The separation facilities were built in the 200 Areas on a high plateau in the center of the site. The fuel fabrication facilities, test reactors, and research and development (R&D) laboratories were built in the 300 Area on the south side of the site. The facilities discussed in the following sections represent those facilities out of more than 500 major facilities where nuclear activities were conducted.

Throughout this document, an asterisk (*) after a date indicates either that there were two dates found during the research or that the actual year during the decade was not found in the documents. The dates this document gives in these cases are the most favorable to claimants. In addition, if "19UNK" is used, a date has not been found.

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The main sources of information for the following sections are several Hanford Site histories. They include Author unknown (undated), Ballinger and Hall (1991), Carlisle and Zenzen (1996), DOE (undated), Gerber (1992, 1993a,b,c, ca. 1994, 1996, 2005), Marceau et al. (2002), Selby and Soldat (1958), Wilson (1987) and Wilson et al. (1990). Other sources are noted as appropriate.

2.2.1 <u>Reactors</u>

Nine graphite-moderated, light-water-cooled reactors were constructed near the Columbia River in the Hanford 100 Areas over a period of 20 years commencing in 1943 [1]. The production reactors were used to produce plutonium by irradiating metallic uranium fuel elements with neutrons during the fission reaction in the reactor core. Other defense-related radionuclides that were experimented with included irradiation of thorium to produce ²³³U, irradiation of depleted uranium to produce ²⁴⁰Pu, irradiation of neptunium targets to produce ²³⁸Pu, and irradiation of americium to produce ^{equine}Pu, irradiation of neptunium targets to produce ²³⁸Pu, and irradiation of americium to produce ^{equine}Pu, were constructed in the B, D, F, H, K, and N Areas, respectively, from 1944 to 1963. The first eight reactors at Hanford, designated 105-B, -C, -D, -DR, -F, -H, -KW, and -KE, were similar in design, using a once-through, light-water cooling system. The ninth reactor, 105-N, used a closed-loop, light-water cooling system. The ninth reactor, 105-N, used a closed-loop, light-water cooling system. The segnetive are primarily due to the presence of activation and fission products (FPs). During operation, entry was controlled into areas where high direct dose rates from photons and neutrons could occur. Internal exposures to uranium, activation products, and FPs were primarily limited to maintenance and refueling activities.

In addition, seven physical testing, research, and demonstration reactors were operated at Hanford over a period of 40 years [2]. Six research and test reactors in the 300 Area were constructed and operated from 1943 to 1978. These include the 305 Hanford Test Reactor (HTR), Physical Constants Test Reactor (PCTR), Thermal Test Reactor (TTR), Plutonium Recycle Test Reactor (PRTR), Plutonium Recycle Critical Facility (PRCF), and High Temperature Lattice Test Reactor (HTLTR) (Newman 1989). The Fast Flux Test Facility (FFTF) Reactor was constructed and operated from 1979 to 1993 in the 400 Area. The years of operation and radionuclides of concern are listed in Tables A-3 and A-4, respectively, in Attachment A.

2.2.1.1 105-B, -C, -D, -DR, -F, -H, -KW, and -KE Production Reactors

The first three reactors (100-B, -D, and -F) were built in B, D, and F Areas starting in August, November, and December 1943, respectively. B Reactor commenced operation in September 1944. The D and F Reactors commenced operation in December 1944 and February 1945, respectively. In addition to producing plutonium for nuclear weapons, the reactors produced polonium for use as a neutron source in the weapons. By May 1945, four of the tubes in D Reactor were charged with bismuth slugs for polonium production. Construction of DR Reactor, in D Area, was started in December 1947, with startup commencing in October 1950. H Reactor was constructed in H Area starting in March 1948, with startup in October 1949. Construction of C Reactor in B Area commenced in June 1951, with startup in November 1952. The KW and KE Reactors were constructed in the KW and KE Areas starting in November 1952 and January 1953, respectively. They commenced operation in December 1954 and February 1955, respectively. These eight reactors were graphite-moderated and single-pass-cooled with treated river water. The first three reactors (B, D, and F) had the same design. The remaining five reactors (DR, H, C, KW, and KE) were similar in design, with the KE and KW reactors differing primarily in the number, size, and type of process tubes; the size of the graphite stack; and the type of shielding.

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The fuel elements were metallic uranium clad with aluminum [3]. In 1960, confinement systems were incorporated in the first eight reactors. These reactors were shut down over a period of years starting in 1964 and ending in 1971. See Table A-1 for shutdown dates.

2.2.1.2 105-N Production Reactor

Construction of 105-N (the ninth production reactor) was started in N Area in 1959. The N Reactor design was based on the original graphite production reactors. However, it differed substantially from the first eight because it incorporated closed-loop cooling and it was the first Hanford reactor to incorporate a confinement system in the original design. It was designed to be a dual-purpose reactor (i.e., to produce both plutonium and steam to be used to generate power). The dual-purpose reactor started producing plutonium in March 1964 and electrical power some time later. The N Reactor fuel was a tube-in-tube design that consisted of a metallic uranium core with thin Zircaloy-2 cladding. The core consisted of an inner tube of 0.95%-enriched ²³⁵U and an outer tube of either 0.95%- or 1.25%-enriched ²³⁵U. From 1965 to 1967, tritium was produced at N Reactor using fuel elements that were manufactured in the 333 Building. The N Reactor was shut down in 1987.

2.2.1.3 305 Hanford Test Reactor

The 305 HTR (also called the Test Pile) in the 305 Building was the first reactor to operate at Hanford, starting in 1943. It was operated until 1972 at a very low critical level (usually less than 50 W) to test fuel elements, fuel configuration, graphite samples, and other materials for the production reactors. It was used as a quality assurance (QA) tool to house testing of samples of each lot of graphite, uranium, aluminum jacketing material, and other materials that were used in the large production reactors. The reactor consisted of a graphite pile and was air-cooled. It was removed from the 305 Building in 1976 and 1977.

2.2.1.4 305-B Physical Constants Test Reactor

The PCTR started operation in 1954. The 800-W test reactor was in a shielded room in the 305-B Building. The mission of the PCTR was to measure reactor changes as a result of use of different reactor fuels. Some plutonium contamination that occurred in the reactor room as a result of an accident could have been the source of minor internal exposure during cleanup. External exposure was negligible because the reactor was in a shielded underground room and remotely operated. Operation was terminated in 1970.

2.2.1.5 305-B Thermal Test Reactor

The TTR started operation in 1954. The 1-KW reactor was in a shielded underground room in the 305-B Building and remotely operated. The mission of the TTR was to measure thermal impact on fission cross-sections. It functioned as an early and small version of the HTLTR. External exposure was minimized because the reactor was remotely operated. Operation was terminated in 1978.

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2.2.1.6 308 TRIGA Reactor

A Training, Research, Isotopes, General Atomics (TRIGA) reactor operated in the 308 Building. See 2.2.5.7.

2.2.1.7 309 Plutonium Recycle Test Reactor

The PRTR, in the 309 Building, started operating in 1960. The PRTR was a 40-MW, heavy-watercooled and heavy-water-moderated reactor. The reactor was chosen for a large fuels diversification program known as the Plutonium Fuels Utilization Program. Tests were to be performed with various mixed oxide fuels using various methods of manufacture. As an example, tests were performed on a variety of powdered and pelletized fuels using a blend of plutonium oxide, uranium oxide, and other metallic oxides. The fuel was nominally 96% depleted uranium oxide and 4% plutonium oxide powder that was vibration-compacted within the fuel pin. External exposure was minimized as a result of the remote operation and the shielding. Maintenance operations resulted in external exposure from FPs and activation products. A high extremity exposure occurred when a worker picked up an irradiated steel pin. Tritium (³H) oxide was the principal internal exposure contaminant during normal operation. This whole-body exposure was added into the external whole-body dose summary. (ORAUT-TKBS-0006-6, Hanford Site – Occupational External Dose (ORAUT 2010a), and ORAUT-TKBS-0006-5, Hanford Site - Occupational Internal Dose (ORAUT 2010b) provide details.) The reactor was shut down in 1969 after an accident resulted in contamination in the reactor. A sample of the contamination showed the major contaminant was ⁹⁵Zr/Nb. Other contaminants included ⁶⁰Co, ¹⁰³Ru, ¹⁰⁶Ru, ¹⁰⁶Rh, ¹⁴⁰Ba/La, ¹⁴¹Ce, ¹⁴⁴Ce, and ¹⁴⁴Pr.

2.2.1.8 309 Plutonium Recycle Critical Facility

The PRCF, which was in the 309 Building, began operating in 1962. Tests were conducted in the PRCF to determine which geometric arrangement of fissionable materials would work in a reactor. It was shut down in 1976.

2.2.1.9 318 High Temperature Lattice Test Reactor

The HTLTR was a 2-MW test reactor that was built in the 318 Building in 1967 and commenced operation in 1968 (Newman 1989). The reactor consisted of a graphite cube in a large shielded room; it was operated at elevated temperatures up to 1,000 °C. The mission was to advance reactor physics technology. The reactor was shut down in 1971 after 3 years of operation over 4 years, during which time it operated with six different cores (i.e., three ²³³U, one ²³⁵U, one ²³⁸U, and one ²³⁹Pu). Because the reactor was heavily shielded and operated remotely, external exposure was at background levels. There were no contamination incidents, so internal exposure was negligible. The building and the shielded room were modified in the early 1980s and used as the new Hanford Radiation Calibration Facility (see Section 2.2.5.8).

2.2.1.10 405 Fast Flux Test Facility

The FFTF was a 400-MW sodium-cooled test reactor in the 405 Building. It began startup operations in 1979 and first went critical in August (Briggs 2001, p. 34) in support of the Fast Breeder Reactor program. The FFTF was used to test the irradiation of fuels and materials that would be used in a breeder reactor and to perform long-term testing of reactor components and systems. It was also used for the production of medical isotopes and research on space power systems. The external and internal exposures were at background levels. The facility was shut down in December 1993 and currently is awaiting decontamination and decommissioning (D&D).

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2.2.2 Fuel Separations Facilities

Seven separation facilities – B Plant, T Plant, U Plant, Reduction-Oxidation (REDOX), Plutonium-Uranium Extraction (PUREX) Plant, UO₃, and C Plant – were constructed and operated in the 200-W and 200-E Areas from 1943 to 1993 [4]. B Plant, PUREX, and C Plant were in 200 East; T Plant, U Plant, REDOX, and UO₃ were in 200 West. The tritium processing facility was operated from 1949 to 1955 in the 108-B Building in the 100-B Area. Natural uranium and thorium fuel elements and lithiumaluminum target elements were irradiated to produce ²³⁹Pu, ²³³U, and tritium, respectively. The spent reactor fuel was transported to the 200 Areas (i.e., Separations Areas) for processing. Spent fuel was initially stored in the reactor storage basins and the 212-N, 212-P, and 212-R Buildings in the 200-N Area before processing in the fuel separations facilities (e.g., 221-B, 221-T, REDOX, and PUREX). The irradiated lithium-aluminum target elements were transferred to the 108-B Building in the 100-B Area for processing. Irradiated thorium fuel elements were processed in the PUREX Plant to recover ²³³U in two separate campaigns that occurred in 1966 and 1970. External exposures were primarily high-energy betas and photons from FPs in the separations facilities. Internal exposures, primarily due to contamination incidents, could be due to either FPs or plutonium. The years of operation and radionuclides of concern are listed in Tables A-5 and A-6, respectively, in Attachment A.

2.2.2.1 Original Fuel Separations Facilities

Three large canyon buildings – 221-T, 221-B, and 221-U – were originally constructed in the 200 Area starting in 1943 to chemically separate plutonium from uranium and FPs. These buildings were massive rectangular canyons that housed the chemical separations equipment. This equipment was operated remotely to provide protection to the workers from the high radiation fields. The T Plant started up in December 1944 and shut down in August 1956, while B Plant started up in April 1945 and shut down in October 1952. Because the capacity was not needed, the startup of U Plant was delayed until 1952 when the mission was changed (see Section 2.2.2.4).

The 221-T and 221-B Buildings, known as the T and B Plants, used a bismuth phosphate precipitation batch process to separate plutonium from the uranium and FPs after the aluminum jackets were dissolved in sodium hydroxide solution and the uranium metal slugs were dissolved in nitric acid. The dilute plutonium solution from T and B Plants was transferred to the 224 Concentration Buildings (e.g., 224-B and 224-T) behind each canyon building, where the product was purified, the product volume was reduced, and the carrier was changed from bismuth phosphate to lanthanum fluoride. The final plutonium product was concentrated in the 231-Z Building. The B Plant was closed in 1952. Starting in 1954 and 1955, the processing capacity was increased in both facilities. The 221-B and 221-T Plants were not restarted because of the success of PUREX.

In March 1956, 221-T was converted to perform equipment decontamination (Gerber ca. 1994). In 1959, the 2706 T Decon Annex was added to the 221-T Building to enhance the equipment decontamination capabilities, particularly for pieces of equipment too large to take into T Plant. The T Plant is still in operation.

The B Plant facilities were modified in 1968 to remove ⁹⁰Sr and ¹³⁷Cs from high-level wastes from the single- and double-shell waste tanks. This was in support of a new mission to isolate the longer-lived FPs by fractionalization for storage in a safer form and to return the short-lived fractions to underground storage and ultimate solidification. The purified ⁹⁰Sr and ¹³⁷Cs solutions were transferred to the 225-B Waste Encapsulation Storage Facility (see Section 2.2.2.6). The ¹³⁷Cs and the ⁹⁰Sr recovery operations at the B Plant were completed in September 1983 and February 1985, respectively [5].

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2.2.2.2 202-S REDOX Plant

Construction of the REDOX Facility (S Plant) was started in 1949. In January 1952, REDOX started up in the 200-W Area using the REDOX process as a replacement for the bismuth phosphate process. This new REDOX process differed from the process that was used in B and T Plants in that, with the exception of the metal dissolving and feed preparation steps, all steps were continuous rather than batch. The new process, which involved solvent extraction using hexone, resulted in improved throughput, recovery of uranium, improved recovery of plutonium, and a decrease in the amount of high-level liquid waste that was generated. The aqueous plutonium product was concentrated for shipment to the plutonium finishing facilities. In 1950*, processing of irradiated plutonium metals (0.947 enrichment) was assigned to REDOX. Shutdown of this facility occurred in December 1967.

In an effort to enhance production capacity, the 233-S Building was constructed in 1957 to contain equipment for the third and final concentration cycle at 202-S. This building was shut down in July 1967 after a major fire resulted in extensive plutonium contamination. The facility is currently undergoing D&D [6].

2.2.2.3 202-A PUREX Plant

Construction of the PUREX Facility (A Plant) commenced in 1953. It was started up in January 1956 using the PUREX process (Gerber 1993a; Marceau et al. 2002). The PUREX process was an organic solvent extraction process that used tributyl phosphate in kerosene rather than hexone, nitric acid as a salting agent, pulse column contractors, and nitric acid recovery by distillation. The nominal design capacity was 8.33 tons of uranium per day as opposed to 2.5 tons for the REDOX Plant. In 1955, due to the development of more restrictive ¹³¹I release limits, the 293-A facility was constructed between PUREX and the 291-A stack. In 1958, PUREX initiated ²³⁷Np recovery activities. Equipment was added in 1962 to permit continuous recovery of ²³⁷Np. In 1963, the processing of E-metal fuel that contained ²³⁵U enriched to 1.75% began. The processing of irradiating powdered thorium oxide targets to recover ²³³U commenced in 1965 and 1966. In 1970, the targets were changed to pelletized thorium oxide. The PUREX Plant was shut down in June 1972 and restarted in November 1983. During the shutdown, a new process was added to convert plutonium nitrate to plutonium oxide. The Plant was started up and shut down several times between 1983 and 1992; final closure was announced in December 1992.

A 500-ft railroad tunnel that extended south from PUREX was designed and constructed to permit the remote storage of highly contaminated processing equipment. In 1964, a new 1,500-ft tunnel was added to PUREX to accommodate the storage of additional equipment.

2.2.2.4 221-U U Plant

As noted in Section 2.2.2, the U Plant was constructed in 1943. The U Plant startup was delayed until March 1952, when the Plant was reconfigured for use in the recovery of uranium from process waste. The high-level liquid wastes from the B and T Plants, which were stored in single-shell tanks, contained large quantities of uranium. The uranium-bearing sludge from the underground storage tanks was pumped to process vessels in the U Plant. The sludge was dissolved in nitric acid, and the uranium was extracted by solvent extraction using tributyl phosphate in kerosene. The recovered uranium was shipped off the site for reuse. The U Plant was shut down in January 1958.

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2.2.2.5 224-U UO₃ Plant

The 224-U Bulk Reduction Building (known as the UO₃ Plant) was constructed in the 1940s. In 1951, it was converted to process the liquid uranium nitrate solution from U Plant to produce a powdered uranium oxide (UO₃) using a calcination process. It was started up in January 1952. The product was shipped off the site for reuse. A major addition (224-UA) was added to the UO₃ Plant in the fourth quarter of 1956 to house three new calciners. It was shut down in 1972 and opened again in 1983. There were 17 startups and shutdowns between 1984 and 1992 that corresponded with activities at PUREX. The plant was deactivated in the summer of 1993.

2.2.2.6 225-B Waste Encapsulation and Storage Facility

The 225-B Waste Encapsulation and Storage Facility is in the 200-E Area near the 221-B Building. In 1974, as a part of the effort to isolate the longer-lived FPs from high-level waste tanks, the ¹³⁷Cs and ⁹⁰Sr solutions were transferred to 225-B Building from 221-B Building for conversion to a solid, encapsulation, and storage (see Section 2.2.2.1 for more details) (Briggs 2001, p. 33). A large amount of encapsulated ¹³⁷Cs and ⁹⁰Sr was stored in the 225 Building as a result of this operation. External exposures that were associated with the high-energy photons and beta particles from the large quantities of strontium and cesium that were processed were minimized as a result of remote operation. Internal exposures from the operation were also minimal due to the remote operation.

2.2.2.7 201-C C Plant or Hot Semiworks

The Hot Semiworks (also known as "hot sloppy") started up in 1949 as a pilot plant (Marceau et al. 2002) to develop REDOX reprocessing technologies. The REDOX process was tested in the 201-C reprocessing pilot plant starting in 1952, with work continuing until 1954. In 1954, the C Plant was switched to being the PUREX process pilot plant; it was shut down in 1956. In 1962, C Plant was restarted as a pilot plant for the recovery of cesium and strontium from FP waste. The C Plant was retired in 1967.

2.2.2.8 108-B Tritium Extraction Facility

The 108-B Building (known as P-10 Plant and the Mint Works) in the 100-B Area started in August 1949 to extract tritium [7]. The lithium-aluminum (for a short period lithium fluoride) target elements were transferred to the 108-B facility after irradiation in one of the production reactors. Tritium extraction operations were performed intermittently between August 1949 and 1955. The 108-B tritium operation was terminated in 1955. The lithium-aluminum target irradiation was continued with transportation to an offsite facility for processing until September 1964. A total of 10.6 kg of tritium was produced at Hanford. Internal exposure was related to ³H. This whole-body exposure was added to the whole-body dose summary (see ORAUT 2010a,b).

2.2.3 <u>Fuel Fabrication Facilities</u>

The Hanford Fuel Fabrication facilities that are described in subsequent sections were built and operated in the 300 Area from 1943 to 1988 [8]. These include three fuel fabrication facilities – 313 (Uranium Metal Fuels Fabrication), 314 (Uranium Metal Extrusion), and 333 (Fuel Cladding) Buildings – and two support facilities – 303 (Uranium Storage and Oxide Burner) and 306 (Reactor Fuel Manufacturing Pilot Plant). External exposures related to betas and photons that are associated with uranium. Due to the pyrophoric nature of uranium turnings and chips, internal exposure related primarily to inhalation of uranium, although DRs should also consider ingestion as a possible pathway.

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The years of operation and radionuclides are listed in Tables A-10 and A-11, respectively, in Attachment A.

2.2.3.1 313 Uranium Metal Fuels Fabrication Facility

The first extruded uranium rods arrived in October 1943 and machining began in December. The aluminum-clad fuel for the first eight production reactors was produced in the 313 Building starting in March 1944. Natural uranium billets or bars were heated to a red heat and shaped into long rods using an extrusion press (see Section 2.2.3.2). These were cut into shorter lengths called slugs using a turret lathe. After receiving a finish cut in another turret lathe, the slugs were cleaned then dipped in a succession of molten baths (i.e., the triple-dip method) and pushed into an aluminum can. An aluminum cap was pushed into the can and the top edge of the cap was welded to the can. Bismuth fuel targets were fabricated in the 313 Building beginning in 1944. These were irradiated in the production reactors to produce ²¹⁰Po, which was used as an initiator in some atomic weapons. Leadcadmium fuel rods, which were welded into nonbonded aluminum cans, were produced for use as "poison" elements for the production reactors. Lithium targets were manufactured for tritium production from 1949 to 1952 and again from 1965 to 1967. In the 1950s, production reactor fuel changed from natural uranium to low-enriched uranium. In March 1954, a new simpler canning method that involved a lead-dip process was initiated to replace the old method. All uranium scrap and waste from these processes were salvaged as uranium oxide and shipped off the Hanford Site. In 1955, the fuel production facilities were expanded to produce the fuel for the K Reactors. In January 1971, the manufacture of aluminum-clad fuel that was used in the first six reactors was terminated. After that time, only N Reactor fuel was manufactured (see Section 2.2.3.3).

2.2.3.2 314 Uranium Metal Extrusion Facility

The 314 Building was known as the Press Building, Metallurgical Engineering Laboratory, and later as the Uranium Metal Extrusion Facility. Operation of the 314 Press Building commenced in July 1944 with a mission to process raw uranium billets into extruded rods that were suitable for fabrication into fuel elements. In early 1945, a 1,000-ton large extrusion press was installed in the 314 Building. After installation, the 313 and 314 Buildings assumed all fuel manufacturing responsibilities for Hanford. An additional mission, uranium scrap recovery, was started in 1945. An oxide burner began operation on the north side of the 314 Building in spring 1946 and was moved to a separate building just north of the 314 Building in December 1947 (Gerber 1992, pp. 7, 8). Uranium scraps were mixed with UF₄ (green salt) and calcium chips. Conversion to metal took place in a reduction furnace (DOE2002). The high air contamination that occurred frequently involved a highly soluble class F material. The melt plant was shut down in 1954. The uranium billets were extruded into rods, outgassed, and straightened in the 314 Building. The extrusion process was terminated in 1948 when the fuel element process was changed to use rolled uranium rods. In 1950, a rolling mill was installed in the 314 Building to fabricate Hanford fuel elements on the site. The building was deactivated in 1971.

2.2.3.3 333 Fuel Cladding Facility

The N Reactor fuel was produced in the 333 Building starting in 1961. The N Reactor fuel elements used a tube-in-tube design that consisted of slightly enriched uranium inner and outer cores and thin Zircaloy-2 cladding. The production process included cleaning of the uranium billets and Zircaloy-2 cladding shells; billet assembly and preheating; extrusion; fuel element shaping and cleaning; and welding of the end cap. From 1965 to 1967, outer driver fuel elements were produced in support of tritium production at N Reactor. The lithium-aluminum inner target element was manufactured in the

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3722 Area Shop. The 333 Building was placed on standby in 1988 when the manufacture of N Reactor fuels ceased.

2.2.3.4 306 Reactor Fuel Manufacturing Pilot Plant

Construction on the 306 Building was completed in 1956, and it began operation in May 1957 as a pilot plant for reactor fuel production. The 306 Building had a complete fuel element canning line with the exception of autoclaving. The coextrusion process was developed in the 306-E Building in the early 1960s in support of N Reactor fuel needs. Since the 1980s the two halves of the building – 306-E and 306-W – have been operated by different companies. PNNL operated 306-W and shut it down in May 2004. Westinghouse and Fluor operated 306-E. The exact date for shutdown of 306-E is still being researched but would have been about the same time.

2.2.3.5 303 Facilities Fresh Metal Storage Facilities

Ten buildings (303-A, -B, -C, -D, -E, -F, -G, -J, -K, and -M) were built over a period from 1944 to 1983 to store fresh uranium, chemicals, uranium scraps, and plutonium. Uranium was shipped to Hanford in the form of metal billets. The metal billets were moved to the 303 Fresh Metal Storage Buildings where they were inspected, entered in accountability records, and held for processing. Building 303-L is another building not mentioned above; it was a small building that was constructed in 1961 to burn uranium metal scraps to an oxide form that would be suitable for shipment to the Feed Materials Production Center for recovery. Burning was stopped in 1971 due to the use of unconventional burning vessels (two cement mixers lined with concrete) and poor ventilation. These two factors combined to produce airborne contamination readings that frequently exceeded the then-current Hanford maximum permissible concentration of $2 \times 10^{-12} \mu$ Ci/mL for unrestricted areas and 6×10^{-11} µCi/mL for restricted areas (see Gerber 1992). Building 303L was shut down in 1971 and removed in 1976. In 1983, the new Oxide Burning Facility (303-M) was placed into operation at the same site. It operated from 1984 to 1987. In 1970*, the 303-C Building was used by Pacific Northwest National Laboratory (PNNL) for the storage of plutonium and americium. (PNNL was formerly the Pacific Northwest Laboratory; for convenience, this document uses PNNL throughout.) A pressurization of one of the sealed metal storage cans resulted in the spread of plutonium contamination and the subsequent shutdown of the building until cleanup. The 303-C Building is currently being used for other purposes.

2.2.4 Plutonium Finishing Facilities

Two plutonium finishing facilities, 231-Z (Plutonium Isolation Building) and 234-5Z (Plutonium Finishing Plant Complex), have operated at Hanford from 1945 to the present [9], (231-Z 1945-1977, 234-5Z 1949-present). The latter is still involved in plutonium stabilization efforts as a part of the Hanford cleanup program. Both of these complexes are in the 200-W Area. As noted in Section 2.2.4.3, the Plutonium Finishing Plant Complex consists of several buildings including 234-5Z that involve several processes [i.e., Rubber Glove (RG) Line, Remote Mechanical A (RMA) Line, Remote Mechanical C (RMC) Line, and Recovery of Uranium and Plutonium by Extraction (RECUPLEX)]; the 232-Z Incinerator; the 236-Z Plutonium Reclamation Facility; and the 242-Z Waste Storage Facility. External exposures related to radiation fields, primarily from plutonium low-energy X-rays and spontaneous fission neutrons. During the early years of plutonium finishing operations, essentially all photon radiation resulted from plutonium X-rays. Later, 60-keV gamma radiation from ²⁴¹Am contributed significantly to worker exposure. Neutron radiation was substantially elevated in part of the finishing operation where alpha radiation from plutonium-produced neutrons resulted from the (α,n) reaction with fluorine. Although relatively few Hanford personnel were involved in plutonium finishing. Internal

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exposures related to contamination incidents that involved americium and plutonium. These exposures occurred by inhalation, absorption, and injection (i.e., wound sites). The years of operation and the radionuclides of concern are listed in Tables A-15 and A-16, respectively, in Attachment A.

2.2.4.1 231-Z Plutonium Isolation Facility

The 231-Z Plutonium Isolation Facility began the plutonium finishing process in January 1945. The objective of the Plutonium Isolation Process was to separate the plutonium from the lanthanum carrier as received from T or B Plant and "to purify the product to a degree sufficient for further processing off-site or in the Purification and Fabrication Building (234-5Z Building)" (Lini 2008, p. 12). When the 234-5Z Building (see Table A-15) went into operation in July 1949, the refined product was seldom shipped off the site for plutonium finishing. Chemical processing was continued in the 231-Z Building until 1956, when all steps were transferred to the 234-5Z Building. In 1956, the plutonium metallurgy developmental laboratory was moved from the 234-5Z Building to the 231-Z Building. The 231-Z Building continued to be used for metallurgical research on plutonium and alloys until 1975, was cleaned up and stabilized by February 1977, and was turned over to Rockwell Hanford Operations in 1983 (Lini 2008, p. 1; Briggs 2001, p. 35). Studies on the use of ²³³U were also performed in this building. Small quantities of ²³⁵U and ²³⁷Np were also handled in the building (Lini 2008, p. 26).

2.2.4.2 234-5Z Plutonium Finishing Plant Complex

The 234-5Z Building was used starting in July 1949 for plutonium finishing operations. The Plutonium Finishing Plant (PFP) Complex consists of the 234-5Z Building, including several production areas and several other buildings discussed in subsequent sections. The building is currently undergoing D&D.

2.2.4.2.1 Rubber Glove Line

The RG Line in the 234-5Z Building was used starting in July 1949 for processing plutonium nitrate. All of the tasks Table 2-1 lists, with the exception of Task I, were performed in the RG Line.

Task	Name	Function
Ι	Purification, oxalate precipitation,	Precipitate the plutonium-nitrate feed solution with oxalic acid
	wet chemistry, or feed production	and other agents.
	Hydrofluorination or dry chemistry	Diffuse hydrogen fluoride gas through the precipitate.
	Reduction	Combine the plutonium tetrafluoride with calcium; fire at high
		temperature until fused into a metallic plutonium chunk called a
		button.
IV	Casting	Render plutonium buttons into pits.
V	Machining	Machine the pit to specified dimensions.
VI	Cleaning	Chemically clean the pit.
VII	Coating	Coat the pit for radiation safety.
VIII	Final inspection	Inspect the pit for correct specifications.

Table 2-1. Plutonium nitrate processing tasks [10].

In December 1949, production was temporarily halted to make repairs to solve operational and mechanical problems. It was restarted early in 1950. The RG Line was placed on standby at the end of March 1953, with the exception of the coatings units that were shut down in May 1953. In 1955, equipment from Tasks I and II was removed and buried. A majority of the remaining process equipment was removed from the RG Line in early 1957 in preparation for installation of the new RMC Line.

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2.2.4.2.2 Remote Mechanical A Line

The RMA Line was designed to reduce worker radiation exposure by automating the process described in Table 2-1. In March 1952, the RMA Line began to process the plutonium(IV) oxalate that was prepared in 231-Z. By March 1953, the RMA Line began handling all of the plutonium work. In July 1954 a fire in the RMA Line stopped fabrication of pits for 3 weeks. The RMA Line was taken completely out of service in 1964. The weapon fabrication portion of the RMA Line was removed (see Section 2.2.4.2.3) in March 1966. In September 1967, a glovebox in the RMA Line was reactivated to grind plutonium-aluminum scraps into turnings suitable for plutonium recovery. In 1968, Tasks I to III of the RMA Line were cleaned out and reactivated to participate in expanded programs for commercial (i.e., nondefense) nuclear development and experiments such as oxides for the FFTF. The last run in the RMA line occurred in December 1979 (Gerber 2005, p. 5-24). In 1984, it was decided to keep the RMA Line in standby status. It was never operated again.

2.2.4.2.3 **Remote Mechanical C Line**

The RMC Line began processing plutonium nitrate feed in October 1960. In 1962, a project was undertaken to install 10-in.-thick, water-filled shielding tanks to substantially reduce the neutron exposure to operators. Because of fire(s) in other facilities and the RECUPLEX criticality, other safety improvements including a new criticality alarm system, a new air sample counter, and a new paging system were installed in 1963 and 1964. By 1964, the RMC Line was being used for the bulk of the metal processing and the throughput was increased substantially. As a result of a decision to concentrate weapon fabrication work at the Rocky Flats Plant, the fabrication portion of the RMC Line was removed in March 1966. The line continued to operate on a mixed schedule for the next several years to produce weapons-grade plutonium and fuel-grade plutonium and oxides. As a result of the explosion in the americium recovery process in the 242-Z facility, the RMC Line was shut down in 1976. After preparing to restart production of weapons-grade components, the RMC Line restarted in July 1985. The RMC Line was permanently shut down in May 1989.

2.2.4.2.4 **Recovery of Uranium and Plutonium by Extraction Facility**

The RECUPLEX process facility began in July 1955; over the next few years, the facility was heavily used with increasing equipment failures and increasing radiation levels. In February 1960, RECUPLEX was temporarily shut down to permit an extensive cleanup of hoods and operating equipment. In April 1962, a criticality accident resulted in widespread contamination throughout the RECUPLEX room and in high external exposures to several workers. In May 1962, the U.S. Atomic Energy Commission (AEC) decided to deactivate that section of the 234-5Z facility.

2.2.4.2.5 232-Z Incinerator Facility

In January 1962, the 232-Z Incinerator began processing miscellaneous solid wastes to recover small quantities of plutonium. The process involved incineration of combustible materials, leaching of noncombustible materials in nitric acid, and wet leaching of ash. The recovered plutonium was transferred to RECUPLEX and later the Plutonium Reclamation Facility. In 1973, the 232-Z Incinerator was shut down.

2.2.4.2.6 236-Z Plutonium Reclamation Facility

The 236-Z Plutonium Reclamation Facility was placed in operation in May 1964 to recover plutonium from liquid waste that was generated in the PFP operations. Using a solution of tributyl phosphate diluted with carbon tetrachloride, metal nitrates and other wastes were stripped from a plutonium-

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bearing feed. Nitric acid-hydroxylamine solution was then used to separate the plutonium as a nitrate. The plutonium nitrate was then ready as a feed for the RMA and RMC Lines. In December 1975, 236-Z was shut down for maintenance upgrades and reviews of criticality safety specifications and procedures. In 1976, 236-Z operated for only a short time after restart until the explosion in 242-Z resulted in widespread high levels of ²⁴¹Am contamination in the work area. This accident resulted in the shutdown of the entire Plutonium Finishing Complex in April 1976.

2.2.4.2.7 242-Z Waste Treatment Facility

The 242-Z Waste Treatment Facility began operation in 1963 to recover plutonium from aqueous waste streams from the PFP. An ²⁴¹Am recovery process was installed in a glovebox in 242-Z and began operation in May 1965. The recovery process was converted from batch to continuous in 1969. In April 1976, the 242-Z facility was shut down as a result of a strike. In August 1976, during restart of the americium recovery process, an explosion occurred in a cation ion exchange column that contained approximately 100 g of ²⁴¹Am. This resulted in substantial americium internal exposure. As a result, the 242-Z facility was permanently closed. Doors into the operating area were welded shut and the facility is currently awaiting D&D efforts.

2.2.4.3 Plutonium Storage Facilities

Several plutonium storage facilities were constructed at the PFP (234-5Z) starting in 1949. These include 2736-Z, 2736-ZA, and 2736-ZB built in 1971, 1977, and 1982, respectively. There were 12 vault or vault-type rooms in the PFP. These vaults contained 93% of the Hanford Site's unirradiated plutonium items and 92% of the total Hanford unirradiated plutonium inventory in July 1994 at the time the Plutonium Vulnerability Study (DOE 1994) was performed. External exposure in the vault rooms from low-energy photons and neutrons occurred during periodic inventory and inspection activities. Internal exposure was minimal because there were no routine activities that involved opening the storage containers. Internal exposure would have been a concern in the event of an incident that voided the containment of the storage containers. Stabilization efforts were completed in August 2003.

2.2.5 <u>Research, Development, and Testing Facilities</u>

The 21 major research, development, and testing facilities at Hanford were in the 100, 200, and 300 Areas [11]. Construction of these started in 1944, with the last completed in 1977. These facilities were built to support the development and testing of new processes, materials, and equipment. In some instances, the facilities were used for several different purposes throughout their operating lives. Therefore, the potential exposures were dependent on the missions and processes at any given time. The years of operation and the radionuclides of concern are listed in Tables A-17 and A-18, respectively, in Attachment A.

2.2.5.1 108-F Biology Facility

The 108-F Building was remodeled in 1949 to provide laboratory and office space for the Hanford Site Biology Program [12]. The facility was used to perform radiation effects studies on plants, animals, and fish in support of Hanford operations. Studies involved many radionuclides including ⁹⁰Sr, ¹³¹I, ²³⁸U, and ²³⁹Pu. The building was expanded in 1953 and in 1962 to provide additional space for biological experiments. The facility was closed in 1977 when the Biology Program moved to the 331 Building (see Section 2.2.5.16).

2.2.5.2 120 Critical Mass Laboratory Facility

The Critical Mass Laboratory was originally operated in the 120 Building near the 100-F Area starting in April 1950 [13]. This laboratory was used for nuclear physics R&D studies of plutonium solutions and solids to avoid the accidental occurrence of a criticality in an operating facility. In November 1951, a substantial spread of contamination occurred as a result of an experiment with a plutonium solution. In December 1951, spontaneous ignition of rags that were soaked with nitric acid occurred during cleanup. While the metal building was not destroyed, the facility was permanently closed due to the extensive plutonium contamination. The site was subsequently decontaminated and decommissioned.

2.2.5.3 209-E Critical Mass Laboratory

The 209-E Critical Mass Laboratory was placed in operation in July 1961 [14]. The laboratory, in a room shielded by at least 3 ft of concrete, included a mixing room with a glovebox and mixing hood and a control room from which critical mass physics experiments were remotely conducted. Both ²³⁹Pu and ²⁴⁰Pu were used in the criticality studies. As a result, external exposures were minimized. Internal exposures were related to plutonium contamination incidents. This facility was shut down in 1986* after a series of occurrences that involved criticality safety specifications.

2.2.5.4 222-B Laboratory

The 222-B Laboratory was opened in the 222-B Building in 1945 to perform process or R&D radiochemical operations in support of irradiated fuel-reprocessing facilities [15]. One of the main functions was to test the solution samples from the 221-B and 224-B Buildings at various steps in the separations process. External and internal exposures were associated with the FPs and plutonium in the process samples. The laboratory was shut down in about 1975. This facility was converted to office space.

2.2.5.5 222-S Laboratory

Construction of the 222-S Laboratory was started in 1949. The facility contains laboratories and shielded cells that were used to provide analytical services in support of process control of REDOX, B-Plant, and 242-S and 242-A Evaporators [16]. It is currently used for analytical operations in support of waste management and environmental control processes. External exposures related primarily to FPs in process samples. Internal exposures would primarily be associated with the separations plants process samples (e.g., FPs and plutonium). In the early 1960s, some work was performed with ¹⁴⁷Pm. Internal exposures were related primarily to contamination incidents.

2.2.5.6 222-T Laboratory

The 222-T Laboratory was the first to operate commencing in 1944 in support of the separation process in 221-T and supporting work in 224-T [17]. External and internal exposure potential was similar to that in 222-B. It was decontaminated and used for office space in the 1970s. It was shut down in 1980*. Fire protection was shut off in 1986.

2.2.5.7 308 Plutonium Fuels Pilot Plant

The Plutonium Fuels Pilot Plant was constructed in the 308 Building in 1960 to perform R&D on fuel elements for the PRTR. In the mid-1960s, the PRTR fuel work was terminated. In the late 1960s, neptunium-aluminum alloy fuel target elements were produced for use in N Reactor for a ²³⁸Pu

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production run. A high bay area was added to the building in 1971. A ¹⁴⁷Pm contamination incident occurred in the 308 Building in 1971. From 1977 to 1990, the facility was used for the production of FFTF fuel elements. The 308-A annex was added to the Plutonium Fabrication Pilot Plant in 1979 to accommodate additional plutonium fuels work. A 250-kW Training, Research, Isotopes, General Atomics (TRIGA) reactor was installed in the annex in the late 1970s to perform neutron radiography, a feature of the QA testing program. During its operation, the building experienced many incidents that involved the loss of control of radioactive materials, primarily plutonium. Most of these were contained within usually one laboratory and in adjacent corridors. Glovebox fires, explosions, and ruptures were the most frequent cause of contamination. Improperly sealed irradiated sample containers were another common cause. These incidents occasionally resulted in internal exposures. Fuel fabrication activities in the 308 Building ceased on March 31, 1990; however, special nuclide material in the form of FFTF fuel elements continued to be stored there at least into 1992 (Neiderhiser 1992).

2.2.5.8 318 Radiological Calibration and Development Laboratory

The 318 Building was originally built in 1967 to house the HTLTR (see Section 2.2.1.9). In the early 1980s, the reactor was removed and the heavily shielded 10-m³ reactor cell was converted to a freein-air calibration facility that contains a large ²⁵²Cf neutron source (10⁹ n/s) and a large ¹³⁷Cs photon source (approximately 100 Ci) that were used for dosimeter calibration [18]. A shielded facility was built in the basement to house a high-level ⁶⁰Co source (approximately 1 kCi) to create accident dose rate levels and three 300-kVp X-ray machines. Three major additions were constructed in 1982, 1987, and 1989. The first contained four source wells that held two ¹³⁷Cs, one ⁶⁰Co, and one ²⁵²Cf sources for instrument calibration. The second addition housed instrument testing and repair laboratories, and the last addition provided office facilities for the staff. The calibration laboratory commenced operation in 1983. As a result of the shielding, isolation, remote operations, and operating procedures, external exposures were minimized. Internal exposures were negligible. The facility is still in operation.

2.2.5.9 320 Low-Level Radiochemistry Facility

The Low-Level Radiochemistry Laboratory was built in the 320 Building in 1966. The original missions were to house analytical chemistry services and to provide plant support that involved low-level and nonradioactive samples. The 320 Building currently provides a low-level radiochemistry facility in which very sensitive radiochemical analysis, sample preparation, and methods development can be performed. External and internal exposures were minimal. The facility is still in operation.

2.2.5.10 321 Separation Facility

The 321 Separation Facility (Cold Semiworks) was built in 1944 with a mission to be used as a cold (i.e., unirradiated material) pilot plant to test the bismuth phosphate chemical reprocessing methods and equipment and to study problems as the 221, 224, and 231 Buildings were being constructed. In late 1944, it was decided to add a laboratory for work with small amounts of solutions that contained several tenths to 1 Ci of radioactive materials (Gerber 1993a; DOE 2002). Soon after startup, the 321 Building assumed a mission to demonstrate the effects of proposed process changes on decontamination factors in the 221, 224, and 231 Buildings; conduct isotope separations; and perform experiments on small irradiated samples. In 1949, the 321 Building was used to develop the REDOX process. Tests using radioactive materials were discontinued after 1949. The 321 Building was used in 1953 to perform a cold test of the PUREX process. Pilot-scale tests of the U-Plant recovery process were conducted in 1955. Isotope campaigns were conducted in the 1960s using tracer levels

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of ⁹⁰Sr, ¹³⁷Cs, ¹⁴⁴Ce, ¹⁴⁷Pm, and ²³⁷Np. Primary contamination in the facility was due to thorium and uranium. External and internal exposures were minimal. The building was deactivated in 1988.

2.2.5.11 324 Chemical and Materials Engineering Laboratory

The Chemical and Materials Engineering Laboratory began operation in 1966 to provide R&D studies in support of PRTR operations. It was used first as a fuel recycling pilot plant by housing chemical reprocessing and metallurgical examination capabilities for PRTR fuel elements. It consisted of two groups of large shielded cells for radiochemical and metallurgical studies. The chemistry cells consisted of four cells that were connected to an airlock where studies of various processes for the solidification of high-level liquid waste were performed. The laboratory was used in conjunction with the 325-A hot cells to perform studies on solidification of high-level liquid waste (i.e., Nuclear Waste Vitrification Project) from processing irradiated fuel elements from commercial reactors. A special underground liquid waste pipeline connected the hot cells in 324 and 325-A Buildings. Most recently, the 324 Building was used as a Waste Technology-Engineering Laboratory. Because most of the work was performed in hot cells, external and internal exposures were minimized. The 324 Building is currently undergoing D&D activities.

2.2.5.12 325 Radiochemistry Laboratory

The 325 Building was completed in 1953 to permit multicurie-level chemical development work in support of production and process improvement (Marceau et al. 2002; DOE undated). A high-level radiochemistry wing, 325-A, was placed into operation on the east side of the building in 1959 and 1960. It included three large hot cells for isotope research activities. This included separation and concentration of ⁹⁰Sr, ⁹⁰Y, ¹³¹I, ¹³⁷Cs, ¹⁴⁴Ce, ¹⁴⁷Pm, ²¹⁰Po, ²³⁸Pu, ²³⁹Pu, and ²⁴⁴Cm for special programs. The ¹⁴⁷Pm was used in the manufacture of heat sources starting in 1966. The ²¹⁰Po work started in June 1972 and was terminated in 1975*. In 1990*, ⁹⁰Y was separated from ⁹⁰Sr in multicurie quantities for medical use. These cells were also used in support of the studies on solidification of high-level radioactive waste (i.e., Nuclear Waste Vitrification Project) from processing irradiated fuel elements from commercial reactors. A special underground liquid waste pipeline connected the hot cells in 324 and 325-A Buildings. The Nuclear Waste Vitrification Project that was conducted in the 324 and 325 Buildings in the 1970s involved high burnup commercial fuel elements that were characterized by nominally 26% ²⁴⁰Pu (PNNL 2003). High-level radioanalytical hot cells were added on the west side in 19UNK. The building became known as the Radiochemistry/Cerium Recovery Building. Limiting access to high-level dose rate or contamination areas controlled external and internal exposures. Most exposures related to maintenance in hot cells or accidents in laboratory areas where work was being performed with FPs, plutonium, and other radionuclides. A major ¹⁴⁷Pm contamination incident occurred in the late 1960s. The radiochemical laboratory work resulted in external exposure, and any internal exposure resulted from contamination incidents. The 325 Building still is in use in support of cleanup efforts at Hanford.

2.2.5.13 326 Pile Physics Technology and Metallurgy Facility

Operation of the 326 Building started in 1953 to continue exponential pile physics development work that was originally performed in the P-11 Facility and later in 189-D (Marceau et al. 2002; DOE undated). Polonium-beryllium and radium-beryllium neutron sources with neutron emissions up to 1×10^8 n/s were used to irradiate cobalt and copper test foils. The test foils exhibited radiation levels up to 10 rad/hr at 3 in. The facility was also used for metallurgical studies of reactor components and fuel elements to evaluate characteristics and performance. Some of this work was transferred to 209-E in 1961. A high-energy electron microscope was installed in the basement in 1971 to perform materials studies. During microscope operations, there was a major spread of plutonium and

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irradiated steel contamination. This and other contamination incidents resulted in some internal exposure. In 1983, the 326 Building became the Chemical Science Building. This facility is still in operation.

2.2.5.14 327 Radiometallurgy Laboratory Facility

The 327 Radiometallurgy Laboratory was built in 1953 to replace the 111-B Test Building (Gerber 1993b; Marceau et al. 2002; DOE undated) and first work with irradiated material occurred in July (GE 1953a, p. A-1). It was designed to provide post-irradiation testing and other studies on irradiated materials, particularly fuel elements and fuel cladding materials from and for the production reactors. These facilities originally consisted of eight freestanding shielded cells and two water-filled storage pools. Several modifications were undertaken to provide a shielded loadout facility to handle high-level radioactive waste, a large shielded cell to perform special environmental radiometallurgy studies, and office space. In 1967, it was used for the experimental production of ²³⁸Pu. It was used to study irradiated PRTR, FFTF, General Electric Test Reactor, Experimental Breeder Reactor II, and Materials Test Reactor fuel elements. In 1967, it was involved in a serious contamination event. The 327 Building was renamed the Post Irradiation Testing Laboratory in the 1980s. External and internal exposures related to irradiated materials and FPs that were undergoing metallurgical studies. Work was also performed with ²³³U and ²³⁸Pu. The facility was deactivated in 1987.

2.2.5.15 329 Biophysics Laboratory

The Biophysics Laboratory was opened in 1952 to pioneer development of water chemistry, environmental monitoring, and bioassay analysis programs. Missions for the laboratory include the analyses of low-level (i.e., near-background) quantities of radioactive materials in air, vegetation, soil, wildlife, and river and well-water samples. A neutron multiplier facility was built in 1974 as Section D of the Biophysics Laboratory. It commenced operation in 1977 with a large ²⁵²Cf source. External and internal exposures were minimized. The building is still in operation.

2.2.5.16 331 Life Sciences Laboratory Facility

The Life Sciences Laboratory opened in the 331 Building in 1972 as a replacement to the 108-F complex. The facility was used to perform radiation effects studies on plants, animals, and fish in support of Hanford operations. Studies involved many radionuclides including ⁹⁰Sr, ¹³¹I, ²³⁸U, and ²³⁹Pu. A hot cell that originally contained a 15-kCi ⁶⁰Co source was used for special irradiation studies. External and internal exposures were at a minimum. The facility is still in operation.

2.2.5.17 3706 Radiochemistry Laboratory

The 3706 Building, the original Radiochemistry Laboratory, was placed in operation in early 1945 (Gerber 1993c; Marceau et al. 2002; DOE undated). This building was also known as the Technical Building. The original mission was to perform small-scale experiments with both low- and high-activity radioactive materials in support of production activities with the primary focus on improving the bismuth phosphate process. The QA and quality control activities that were associated with fuel element fabrication also were performed in the 3706 Building. Other missions were the metallurgical examination of irradiated fuel elements, fuel development for the 313 Building, examination of graphite from the experimental levels of the 100 Area piles, and special sample analyses from 231Z and 200 Area separation facilities. In 1947, it was used in the development of the REDOX process. Pioneering radiochemistry work in the development of the REDOX, PUREX, and RECUPLEX processes was performed until 1953. The work involved many radionuclides including FPs, ¹⁴C, ¹³¹I, ²³⁵U, ²³⁸U, and ²³⁹Pu. External and internal exposures related to these radionuclides. Thyroid checks

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were initiated in May 1947. In 1954, many of the laboratories were decontaminated and converted to offices. Most work with radioactivity materials ended by 1964 (Gerber 1992, p. 84). During the 1970s and 1980s, laboratory work was phased out and most of the remaining building was converted to offices that still were in use in the early 1990s.

2.2.5.18 3730 Gamma Irradiation Facility

The 3730 Building was originally constructed in 1949 as a shop. In 1956, the work performed in the 3741 Building was transferred to this building. A 350-kCi ⁶⁰Co source was added to a pool in the 1960s for special irradiation studies. Irradiated samples measured from 25 to 50 rad/hr. Respiratory protection was required in the 1950s because of ¹⁴C and ³⁵S airborne contamination. Several facility improvements were made from 1974 to 1981.

2.2.5.19 3732 Process Equipment Development Laboratory

The 3732 Process Equipment Development Laboratory was constructed in 1949 as an engineering pilot plant for the triple-dip and lead-dip fuel canning processes (Gerber 1992). It is a one-story, metal frame structure on a concrete foundation with a concrete floor and a corrugated metal roof. Overall dimensions are 28.5 ft by 48.16 ft. Powdered thorium oxide fuel targets for ²³³U production were fabricated in the 3732 Building from 1965 to 1967. The process of handling these powder targets spread powdered and particulate thorium contamination throughout the building. After the thorium oxide program switched to pelletized targets in the 3722 Building in 1968, these new fuel targets were canned in the 3732 Building through 1970. As a result, the 3732 Building contains standard fuel fabrication chemical wastes as well as residual thorium oxide contamination in crevices and areas throughout and near the building. The facility is now empty.

2.2.5.20 3741 Graphite Hot Shop Facility

The Graphite Hot Shop was constructed in 1944 to perform metallurgical studies on irradiated graphite samples. Polonium-beryllium and radium-beryllium neutron sources were used in irradiation facilities in the basement to irradiate cobalt and copper foils. Internal exposures would have been primarily due to the large quantities of irradiated graphite (i.e., ¹⁴C) that were studied. The facility was torn down in 1956 and work was transferred to 3730 Building (DOE undated).

2.2.5.21 3745 Radiological Calibrations and Standards Laboratory

The Radiological Calibrations and Standards Laboratory opened in the 3745 Building in October 1944 [19]. The two-story wooden frame building contained a low scatter room in which dosimeters were calibrated using a ²²⁶Ra source and a 220-kVp X-ray machine. Other sources included large ⁶⁰Co and ¹³⁷Cs photon sources and ²⁵²Cf, ²²⁶Ra-Be, ⁸⁴Po-Be, ²³⁸Pu-Be, ²³⁹Pu-F, and ¹²⁴Sb-Be neutron sources. Instrument calibrations were performed in a different room that contained two vertical concrete-shielded wells in which the photon sources were housed and used. Instrument calibration sources included ⁶⁰Co, ¹³⁷Cs, and ²²⁶Ra sources. A supporting facility, 3745B, was constructed of concrete blocks to house a 4-MeV Van de Graaff positive ion accelerator. This was used starting in 1955 to perform fast neutron calibrations. A companion facility, 3745A, was built in 1950* to house a 2-MeV negative ion Van de Graaff accelerator. As a result of the shielding, isolation, remote operations, and operating procedures, external exposures were minimized. Internal exposures were negligible. The calibration operations were moved to the 318 Building in 1983. The building is still used for office space.

2.2.6 <u>Waste Handling Facilities</u>

2.2.6.1 High-Level Liquid Waste Tanks

High-level radioactive liquid wastes from reprocessing facilities at Hanford were stored in underground single- and double-shell tanks starting in 1944 [20]. The high-level radioactive waste is transported through underground process lines to the storage tanks in the 200-E and 200-W Area tank farms. The storage tanks are of two types, single-shell and double-shell. Both types are cylinder-shaped concrete structures with carbon-steel liners. The single-shell tank has a single liner, and the doubleshell tank has two liners with a space in between. The T and B Plants initially produced more than 10,000 gal of high-level radioactive waste for each ton of processed uranium. These wastes were transferred to the first 64 tanks, which were constructed in 1944 and 1945. Thirty new single-shell tanks were constructed in 1947 and 1948. Additional single-shell tanks were constructed in 1950 and 1952 (30), in 1953 to 1955 (21), and in 1963 and 1964 (4). From 1968 to 1988, 28 double-shell tanks were constructed. Most of the single- and double-shell tanks are still in use. A new waste vitrification facility is under construction to process the stored high-level liquid wastes. External exposure is due to high-energy beta and photon radiations from the FPs in the tanks. Under normal conditions shielding, distance, and time limits minimize personnel exposures. Most external exposures would relate to leaks of high-level wastes. Internal exposures to FPs were controlled through the use of respiratory protection.

2.2.6.2 216-Z-9 Trench Facility

The 216-Z-9 Facility, known as the Z-9 Crib (i.e., enclosed trench), is in the 200-W Area approximately 500 ft east of the Z Plant (i.e., PFP) exclusion area. The Z-9 trench was placed in operation in 1955 to serve the RECUPLEX Facility by receiving the acid waste stream of both aqueous and organic wastes as well as unwashed fabrication oil from the RMA Line from 1955 to 1962. The trench was an underground excavation within an active floor area of 30 by 60 ft, 21 ft beneath the top of the concrete slab cover. The excavation was covered by a 9- to 12-in.-thick reinforced concrete slab 90 ft wide and 120 ft long at ground level. In 1965, because of concern that the waste plutonium posed a potential criticality hazard, a decision was made to develop a soil mining and leaching operation to recover an estimated 45 kg of waste plutonium. An approximately 1,000-ft² containment structure was built to conduct the operation. It housed a soil packaging glovebox, personnel entry to the trench, and a personnel change room. The structure was on the east side of the trench with part of the structure over the top of the trench cover. The mining operator's cubicle, which contained the controls for operating the mining equipment, was in the west side of the trench cover through a reinforced opening in the cover. The mining operation commenced in August 1976 and was completed in July 1978. The principal internal exposure potential related to ²³⁹Pu contamination, most of which occurred in the soil packaging glovebox. Because the waste site was used only for plutonium, the external exposure potential during the mining operation was minimal. The facility is currently in a laid-away status.

2.2.6.3 241-Z Settling Tank

The 241-Z Settling Tank, inside the fence of the PFP complex in the 200 West Area, was used from 1949 to 1973 to store large quantities of plutonium-bearing wastes. In 1974, 80,000 L of supernate were removed from the tank, leaving an estimated 94-in.-deep layer of wet sludge that contains a few tens of kilograms of plutonium. Internal and external exposures from this operation were minimal. The facility is currently awaiting D&D.

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2.2.6.4 **242 Evaporator Facilities**

The evaporator facilities (242-A, 242-B, 242-S, and 242-T) were constructed near each of the separation facilities to reduce and concentrate the volume of high-level wastes. The 242-T and 242-B evaporators started processing high-level radioactive waste in 1951. Two new evaporator plants, 242-S and 242-A, commenced operation in November 1973 and 1977, respectively. External exposure would be due to high-energy beta and photon radiations from the FPs in the tanks. Personnel exposures normally are controlled using shielding, distance, and time limits. Most external exposures would relate to leaks of high-level wastes. Internal exposures to FPs were controlled through the use of respiratory protection. The 242-T Evaporator was shut down in 1976 (Briggs 2001, p. 33). The 242-A Evaporator is still in operation.

2.2.6.5 **Chemical Separations Exhaust Filtration Facilities**

Gaseous waste is grouped into three headings: dissolver off-gases, process vessel vent systems, and exhaust ventilation air. Dissolver off-gases contained the major portion of the radioactive iodine (primarily ¹³¹I), as well as krypton and xenon isotopes. Dissolver off-gases also carried a high concentration of nitrogen oxides from the reduction of nitric acid during the dissolution of the irradiated fuel elements. Table 2-2 lists the three exhaust filtration devices that were used starting in 1947 to reduce radioactive material releases from the B and T Separations Plants and later the REDOX Plant:

Table 2-2. Exhaust filtration facilities [21].					
Filtration devices	Date installed				
Caustic scrubbers	1947–1948				
Sand filters and fiberglass filters	1947–1948 1954 (REDOX)				
Silver reactors	1950				

Table 2-2.	Exhaust	filtration	facilities	[21]	
	Exhaust	mination	lacilities	2	

These filtration devices accumulated substantial quantities of radioactive materials from the separations and plutonium concentrations. The caustic scrubbers were used to remove radiochemical vapors from the process vessels. Sand filters were installed in the 202-S, 221-B, 221-T, 291-B, 291-S, 291-T, and 291-Z facilities. The filters were used in separations process exhaust streams to remove particulates and to reduce ruthenium emissions from the exhaust stream. Silver reactors were installed primarily to remove ¹³¹I from the exhaust stream. Radionuclides of interest in the filtration devices included FPs, ¹⁰³Ru, ¹⁰⁶Ru, ¹³¹I, and ²³⁹Pu. Internal and external exposures that are associated with these radionuclides would occur primarily during maintenance operations. These filtration devices became inactive as the separations facilities were shut down.

2.2.6.6 340, 340-A and 340-B Liquid Waste Handling Buildings

The 340 facility complex was operated starting in 1954 to receive radioactive liquid wastes from various 300 Area laboratories via the radioactive liquid waste sewer line [22]. The complex accumulates the liquid wastes in storage tanks. After sample analyses and pH adjustment, the liquid wastes are shipped via a shielded railcar tank to the 200-E Area Tank Farms for treatment and disposal. The radionuclides of concern include ⁶⁰Co, ⁹⁰Sr, ⁹⁰Y, ¹³⁷Cs, ¹⁴⁷Pm, ²³⁸Pu, ²³⁹Pu, and ¹⁴⁴Ce. Access to the tank area was administratively controlled. The external exposures to the operators relate to the high-energy betas and photons. Internal exposures were unplanned and related to leaks or an accident. The facility is still in use.

2.2.7 <u>Miscellaneous Areas</u>

2.2.7.1 700 Area

The 700 Area is in downtown Richland, Washington. It includes the Federal Building, where most of the AEC (and now DOE) staff were located, and other mostly office buildings. The *in vivo* counting facility is and always has been housed in the 747A Building; it contains sealed calibration sources and phantoms. The Dosimetry and Radiological Records Programs were housed in the 712 Building from the 1970s to 1985. No contamination areas or large radiological sources existed in the 700 Area.

2.2.7.2 3000 Area

The 3000 Area is near the Hanford Site in North Richland. Established during World War II, the 3000 Area was originally the site of a camp that housed construction personnel and military police. After the war, the 3000 Area became part of the North Richland Construction Camp. The establishment of the U.S. Army's Camp Hanford in North Richland in 1951 included the acquisition of most of the 3000 Area and the construction camp. Although the area presently contains mostly office buildings, laboratory work with limited radiological exposure has occurred.

2.2.7.3 1100 Area

The 1100 Area is a strip between the railroad tracks and Stevens Drive north of Snyder Street to the southern site boundary. The 1100 Area housed site support services, such as general stores, shipping, receiving, transportation, maintenance, and general warehouse facilities (Marceau et al. 2002, p. 2-1.7). Opportunities for radiological exposure were limited but cannot be ruled out.

2.3 WORK WITH RADIOACTIVE MATERIALS

2.3.1 <u>Thorium</u>

Thorium metal was also called myrnalloy, 10-66, and Q. Thorium oxide was referred to as thoria. Thorium fuel elements were used in the reactors for neutron flux-flattening and for the irradiation of ²³²Th to produce ²³³U.

2.3.1.1 1940s

Gydesen (1954) describes the first activities with thorium at Hanford. Small metallic pieces from Oak Ridge National Laboratory (ORNL) were used in October 1945 for experiments on physical characterization in Hanford laboratories (presumably in the 300 Area). This included machining two thorium slugs to standard size and performing trial canning (Smith 1945). Similar metallic pieces were used to determine thorium's effects on reactor reactivity in the 305 Test Pile.

In January 1946, Hanford received a shipment of 150 lb of thorium, which consisted of crooked 4- to 5-ft-long rods that required straightening before machining (Smith 1946; Gydesen 1954). To prevent the rods from being contaminated with uranium from the contaminated equipment in the 314 Building, these thorium rods were straightened manually in the 300 Area Maintenance Shop with a 75-ton press. This shop was possibly in the 3722 Building (known later as the Area Shop) or in the Tool Room Shop inside the 313 Building. After straightening, the rods were cut into 33 slugs with a power hacksaw, then machined on a small lathe in the 313 Tool Room (Kidder 1946). The slugs were then canned, welded, and tested for integrity by autoclaving in the 314 Building (Gydesen 1954; Gerber 1992). Airborne contamination readings in and near the 313 and 314 Buildings were frequently high.

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Health Instruments Division monitors often reported "metal dust" in building and vicinity air as being "over tolerance" for uranium with ventilation "not adequate" (Gerber 1992). Before charging them into a production reactor, some of these 33 slugs were placed in a test pile (assumed to be the 305 Test Reactor) to determine their effect on neutron flux distribution. On March 26, 1946, these thorium slugs were charged into the 105-D Reactor for testing of their effects on reactivity.

The first production batch of 30 thorium slugs was inserted into a Hanford reactor in 1946 (Wende 1946a,b).

In April 1947, 171.2 additional lb of thorium rods were received at Hanford and were machined into 44 slugs (Smith 1947; Kattner 1948; Gydesen 1954).

2.3.1.2 1950s

The first major tests of metallic thorium elements in the Hanford reactors began with element fabrication using standard Hanford techniques on metallic thorium slugs in the 300 Area facilities in late 1951. Production Test PT-105-516-A began in D Reactor in July 1952 (Brugge 1956). Production Test PT-105-551-A started about October 1953 in B, D, and H Reactors, while some of the earlier test slugs might have still been in place (Brugge 1956). It is likely that most of these fuel elements were actually shipped to ORNL for processing because no records of their dissolution in the Hanford facilities has been found and Gydesen (1954) indicates that they were to be visually examined "before shipment to Oak Ridge" [23].

Many problems connected with the rapid formation of a thick coat of oxide on the thorium metal targets led to experiments with a variety of bonding methods and coatings. These tests resulted in large-scale thorium contamination being introduced into the 313 Metal Fabrication Building and surrounding fuel warehouses (Gerber 1992).

By the end of 1953, thorium elements were in use in all Hanford reactors for use as flux-flattening poisons; they replaced lithium-aluminum and bismuth (DeNeal 1970).

Thorium work was conducted in the 306 Building (alloy and fabrication test and development) from 1955 through 1970 [first mention of canning and cold rolling found in Stevens (1955a), air concentration measurements in Stevens (1955b)]; the 3706 Building (radiochemistry in support of fuel fabrication) from 1954 through 1963; and the 3722 Building (machining of thorium rods in the early years, and later the fabrication of pelletized fuel targets) from 1946 through 1970. Chemical process development occurred in the 321 Building. Gerber (1992) indicates that the 3707-A and 3707-B change houses became contaminated from the nearby fuel fabrication and scrap storage buildings. Contamination (probably mostly uranium [24]) was spread into the change houses via powders and fines on the hands, clothing, shoes, and hair of personnel.

The chemical separation of various forms of thorium target fuel elements after irradiation took place in the 321 Building using the thorium extraction (THOREX) process (Gerber 1992; EPA 2001). Activities in the 321 Building basically involved small-scale prototypes of processes in the large processing facilities using nonradioactive chemicals or tracer amounts of radioactivity materials, although activities up to curie amounts were used for some tests. It is not known how much thorium was used in the building, but equipment was designed and installed in the Cold Semiworks (321 Building) and approximately 1,300 lb of ThO₂ was fired to about 1,000°C in March 1964 (GE 1964a, p. C.2). Training for radiation safety with thorium was given to the 321 Building staff in 1964 (GE 1964a).

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The 3706 Building was involved with development activities including all components of the bismuth phosphate, REDOX, PUREX, and RECUPLEX processes; bioassay and environmental sample analyses; and machining and grinding of metallurgical test samples. Work with thorium is not known but probable. Contamination resulted from inadequate containment systems, spills, overflows, vaporization, spreads of radioactive dusts and fines, and other incidents that involved the loss of control of radioactive materials. In 1954, the building underwent a major decontamination and remodeling effort and many of the laboratories were converted to offices. Sampling laboratories for fuel fabrication operations continued until they were transferred to the 3720 Building in the mid-1960s (EPA 2001).

2.3.1.3 1960s

A small test of thorium irradiation and ²³³U extraction was conducted in 1963 (Matsumoto ,Weiler, and Schneider 1963a,b). The test was conducted in two series, each involving irradiation of 4 (or maybe 8) ampoules of about 1 g each of thorium oxide. After irradiation, the ²³³U, ²³²U, and ²³⁰Th were extracted and measured. The operations were conducted in the Shielded Analytical Laboratory, which was a wing of hot cells in the 325 Building.

There were two major campaigns and an earlier smaller test campaign intended to produce ²³³U at Hanford (Isochem 1967a; Walser 1978) (see Section 2.3.5.) The campaigns involved manufacture of thoria target elements (called wafers), irradiation, element dissolution in PUREX, shipping of the ²³³U product, and recovery and recycling or storage of the unactivated thorium.

After a canning technique was determined, powdered thorium oxide fuel targets for these campaigns were fabricated in the 3732 Building from 1965 to 1967. Sintering of the wafers was part of the process in the 3732 Building. A new technique involving pelletized targets replaced the 3732 Building operations. The pelletized targets were fabricated in the 3722 Building from 1968 through 1970. In addition, that building housed a furnace for the "recycling" (reduction) of depleted thorium oxide after it was processed in PUREX.

Thoria wafer irradiation occurred in B, C, D, KE, and KW reactors. Details are provided in Bowles and Schmidt (1966) and DeNeal (1970, pp. 46–48.).

Processing of the irradiated thorium targets was performed in three distinct campaigns with extensive cleaning of the PUREX lines and tanks before and after introduction of the irradiated thorium wafers. The campaigns included a small test campaign in 1965 (Briggs 2001) and two production campaigns in 1966 and 1970. For the 1965 campaign, the original dissolution was not conducted at PUREX but additional purification of the nitrate solution was (Isochem 1967a; Walser 1978). The PUREX thorium work recovered approximately 565 tons of thorium in the nitrate form from both campaigns (Gerber 1992; Walser 1978; ARHCO 1971).

When not recycled to the 3732 Building, the thorium nitrate was stored in the WR Vault in the 200 West Area (Isochem 1967a). This was a standalone storage area slightly northeast of U Plant. The date the thorium was removed has not been discovered, but the building had residual contamination up to recent years. Entries into the building were rare. Entries required respiratory protection because of the potential for resuspended contamination [25].

There were several storage buildings in the vicinity of the 313 Building that might have stored thorium in some manner. Those were 303 A, B, C, D, E, F, G, and K (Gerber 1992). Until recently, thorium scrap materials were stored in the 303-K Facility; removal was completed September 30, 2001 (FH 2001, Section C.2).

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During 1965 and 1966, an experimental processing of commercial thorium nitrate into thorium oxide powder was carried out in the UO_3 Plant using the old electric pots. The goal of this work was to produce thorium oxide powder suitable for fabrication into reactor target elements for ²³³U production (Gerber 1993a). The task involved a few hundred pounds of thorium.

2.3.1.4 Thorium-Related Fuel Element Failures in Hanford Reactors

All known fuel-element failures in the single-pass Hanford reactors are summarized in Gydesen (1993). Just over 2,000 leaks in the fuel cladding of various degrees of severity are reported. The vast majority of the fuel-element failures involved uranium elements. A history of the thoria program and descriptions of the first 25 failures are provided by Bowles and Schmidt (1966). Table 2-3 lists the thorium element failures. Except perhaps for brief periods in 1965 and 1966 in KE and KW Reactors, dosimetric contributions from thorium, either from external irradiation or inhalation, would have been greatly overwhelmed by the activation products, FPs, and actinides in the uranium elements from other fuel failures [26].

2.3.2 <u>Americium-241</u>

2.3.2.1 1940s and 1950s

The Hanford monthly report for July 1949 (GE 1949a) notes work on a method to separate americium and curium from lanthanides. This appears to be benchtop-scale chemistry. No building was mentioned, but the same report had a separate report for activities in the 234-5Z Building, which had first come on line that same month and was having some startup difficulties (Gerber 2005), with no mention of americium separation activities. This work is mentioned again in the September report, so apparently it was ongoing at that point (GE 1949b). It is most probable that this work was being done in the 231-Z Concentration Building.

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Year	Date	Facility	Activity	Number
	Duit	. aonty	First Th slug failure in a reactor ("Grossly	
1954	21 Jan	н	mis-machined, should not have been canned.")	3884-H
1954	27 Apr	В	Pushed out tube and metal column	10-66
1954	20 Sep	В	PT-105-551	3(Q)
	•			
1965	19 Jan	KW	4653-KW	T1
1965	19 Jan	KW	4653-KW	
1965	19 Jan	KW	Unknown KW	
1965	18 Feb	KE	5881-KE	T2
1965	22 Feb	KW	1865-KW	Т3
1965	22 Feb	KW	3083-KW	T4
1965	22 Feb	KW	1374-KW	
1965	1 Mar	KE	Caused by a spline left in the tube; tube removed	T5
1965	25 Mar	С	0172-C	T6
1965	29 Mar	KW	1853-KW	T7
1965	29 Mar	KW	4265-KW	T8
1965	31 Mar	KE	5885-KE	Т9
1965	3 May	KW	4971-KW	T10
1965	3 May	KW	3553-KW	T11
1965	3 May	KW	3559-KW	
1965	12 May	С	1196-C	T12
1965	12 May	С	2396-C	T13
1965	14 May	С	3496-C	T14
1965	23 Jun	С	0358-C	T15
1965	26 Jun	KW	4168-KW	T16
1965	27 Jul	С	3853-C	T17
1965	13 Sep	KE	2898-KE; end cap completely removed	T20
1965	21 Oct	KE	5852-KE	T21
1965	30 Dec	KE	4274-KE	T22
1966	13 Apr	KE	3298-KE	T23
1966	7 Jul	KE	5791-KE	T24
1966	17 Oct	KW	5546-KW	T25
1968	25 Nov	KW	5791-KW	T26
1968	18 Dec	KW	4898-KW	T27
1970	2 Jun	KE	0174-KE	

Table 2-3. List of thorium fuel element failures in Hanford reactors.^a

a. From Gydesen (1954, 1993), Bowles and Schmidt (1966).

The June 1950 report (GE 1950a) indicates that a 1-day special irradiation on a 0.01-g sample of americium was conducted in the reactor and the sample shipped to the University of California Radiation Laboratory. There is also mention of work on a procedure to separate americium and curium from plutonium in irradiated uranium solutions and that several samples had been successfully obtained. There is no mention of sample sizes or where the work was being conducted (GE 1950a).

The July 1950 report (GE 1950b) states that a request for >50 mg of americium was received from the AEC for an offsite customer, so 25 sample cans from the 234-5Z Building were set aside to allow ingrowth of the ²⁴¹Am. In September, possible extraction chemistry was presented as Production Test 321-12. The letter report indicated that the process could be done by one to three operators (Szulinski and Parker 1950). In November 1950, 70 mg of ²⁴¹Am had been extracted from those samples in the 231-Z Building. The report says that the americium-bearing supernatants were to be concentrated at the 234-5Z Building in one of the recovery units (GE 1950c; Gerber 2005). However, this task was not mentioned again in subsequent monthly reports.

In March 1951, about 24 mg of americium were recovered in a special run in the 231-Z Building (GE 1951).

Recovery of plutonium from scrap metals and powders began in the Skull Recovery Facility (Hoods 40 and 41 in the 234-5Z Building) in November 1952. The process also allowed for extraction of americium (Gerber 2005). However, the February 1953 Hanford Works monthly report indicates that the actual separation of americium from the dissolved solutions occurred at the 231-Z Building (GE 1953b) as does the final report on the production test (Packer 1954, p. 3).

Improved scrap recovery processes including RECUPLEX in 1955 and ion exchange after the RECUPLEX criticality accident in 1962 provided the capability to skew the plutonium-americium ratio substantially in some liquid waste streams by selectively removing the plutonium. Therefore, waste lines and tanks would have been enriched with americium [27].

2.3.2.2 1960s and 1970s

J. S. Buckingham proposed a method to separate ²⁴¹Am from high-level waste from REDOX or PUREX using the I-F cubicle in 222-S in 1964 (Buckingham 1964a). The cubicle was a hot-cell-like facility that was designed to handle samples of high-level waste remotely. It was used to separate samples of ¹⁴⁷Pm and ²³⁷Np from PUREX and REDOX high-level wastes. However, Mr. Buckingham stated in an interview that the work was not funded and major work involving separation of ²⁴¹Am was not done at 222-S while he was there. He said that he believed a short project was conducted after the ¹⁴⁷Pm work was concluded (Bihl 2008a). This agrees with a series of ²⁴¹Am bioassays that were obtained from several construction workers in response to a contamination spread that occurred due to cutting into some process lines in the 1-F cubical in 1967. Contamination smears showed that the contamination consisted of ²⁴¹Am and ¹⁴⁷Pm.

The Plutonium Reclamation Facility, 236-Z, recovered plutonium from various types of production scrap. The Facility initiated a chemical run on March 3, 1964, even though construction was not complete until June 1964 (Briggs 2001). In early years, most americium in the liquid waste from the 236-Z Facility went to soil disposal cribs. Later, the waste stream was sent to the 241-Z storage tanks and ultimately to the Tank Farms. The Waste Treatment Facility, 242-Z, was built to capture traces of plutonium in the liquid waste stream from the 236-Z Facility and to selectively remove and concentrate the americium from its waste stream. Briggs (2001) indicates that Phase I started in August 1964, and the first americium was recovered in September 1964. The Waste Treatment Facility was shut down and closed off after an ion exchange column explosion on August 30, 1976.

Laboratory-scale R&D of flowsheets (separations procedures) for the processes in the Plutonium Reclamation Facility and the Waste Treatment Facility was done in the Plutonium Chemistry Laboratory (in the 234-5Z Building), apparently starting in 1963 and continuing until at least 1966 (Hopkins 1965, p. 68; Szulinski and Curtis 1963; Kingsley 1964a,b, 1965; Isochem 1966a). Similar chemistry research, development, and support was conducted in the Chemical Technology Laboratory (also in the 234-5Z Building) on a number of things most likely including ²⁴¹Am, according to an interview of a PFP chemist (Bihl 2008b). Dates for the latter work were not determined precisely, but the chemist suggested the early to mid-1960s (Bihl 2008b).

Other americium flowsheet development was conducted in a modified fume hood in the 325 Building in 1964 (Wheelwright 1964; Bihl and Napier 2008). This work involved some tracer-level work that was followed by a trial with 1.6 Ci of ²⁴¹Am in 7 L of nitrate with trace quantities of plutonium, thorium, and other metals. The chemist who conducted the work was interviewed by phone (Bihl 2008c) and produced a set of overheads for a presentation he had given a few times about the subject of these

separations at 325 Building (no hard copy available). According to the presentation, 3 kg of pure ²⁴¹Am were separated in A Cell of the 325 Building in four campaigns in 1973 through 1976. The material was converted to oxide form, placed in shipping containers, and shipped to ORNL. The final steps were done in a glovebox.

There was considerable interest in separating various radionuclides from high-level waste in the 1960s and 1970s, including ²⁴¹Am. The feed material was generally PUREX 1WW (first cycle waste) or waste from the commercial nuclear power plant at Shippingport, Pennsylvania (Gerber 1992, p. 138). In 1966, Isochem and Battelle Memorial Institute were gearing up for extraction of a number of isotopes from dissolution of Shippingport fuel in REDOX (Roberts and Bray 1966; Beard 1967). The isotopes being investigated were ¹⁴⁷Pm, ²⁴¹Am, ²⁴⁴Cm, and ¹⁴⁴Ce (Gerber 1992, p. 138). The plan was to do initial cleanup of the FP waste at 201-C (Hot Semiworks, Strontium Semiworks) with final purification of the various radionuclides at the 325 Building. This work was done in A Cell (a hot cell), which was also used to purify the ¹⁴⁷Pm (Gerber 1992, p. 139). To test the developed flowsheets, the chemists ran test samples in 1966 using actual REDOX and Shippingport fuel wastes at 201-C and in the hot cells in 325 Building (Roberts and Bray 1966; Beard 1967). Some of this work might have been done in 1965. The May 1965 PNNL monthly report (PNL 1965a, p. 152) indicates that attempts at extraction of americium were being conducted at "Semiworks." The 200 Area monthly report for March 1966, for instance, stated, "The promethium feed concentrate, recently sent to Battelle Northwest, and containing 1.5 to 2 kg of well-aged Pm-147, is estimated to contain 40 grams of Am-241; it is planned to recover this material" (Christy 1966a, p.16). Larger scale work was done in 1967 and 1968. Bray (1967) indicates that a campaign using the waste from Shippingport fuels was conducted at REDOX from December 1966 through February 1967. As planned, the crudely separately material was sent to "Strontium Semiworks" for additional cleanup, which was completed by September 1967. The material was then sent to the hot cells in the 325 Building to await final separation of americium, curium, and promethium. In a July 2008 interview, one of the chemists indicated that the separation of the americium and curium was conducted during 1968 and into the 1970s. He showed a picture of 8 g of curium that was purified in the 325 Building hot cells in 1968 (Bihl and Napier 2008). The February 1969 monthly report stated that the remains of the Shippingport americium and curium separation was shipped to the Savannah River Site (SRS) at that time (Albaugh 1969, p. 57).

Some special flowsheet development work was done in the 324 Building from August through November 1967, specifically applicable to the possibility of separating ²⁴¹Am and rare earths from B-Plant stock (PNL 1967a, p. 42; 1967b; p. 50; 1967c, p. 53). The August runs used cold reagents with some ²⁴¹Am tracer. Later runs in October and November might have used higher level activity (PNL 1967b, p. 50; 1967c, p. 53). Slightly different americium flowsheet development was also done in the 324 Building by the same researcher in November and December 1967 (PNL 1968, pp. 64, 65.)

Gerber (1992, p. 41) states that the north end of the 3708 Building was used in the early 1970s for the experimental canning of americium and curium oxide fuel blends for irradiation in the single-pass reactors.

2.3.3 <u>Neptunium</u>

Neptunium-237 was irradiated to produce ²³⁸Pu, which was a heat source for thermal generators.

At Hanford, neptunium had several pseudonyms: *birch, palm*, and *brandy*. One of the programs to extract and irradiate ²³⁷Np to create ²³⁸Pu was called *Palmolive*, with the ²³⁸Pu being called *olive*. *Brandywine* was also used to refer to ²³⁷Np nitrate being shipped from Hanford.

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2.3.3.1 Before 1958

Possibly the first handling of ²³⁷Np at Hanford occurred in 1945. In a letter to file, W. E. Jordan discusses plans to use a Hanford reactor, probably B Reactor, to irradiate 1 mg of ²³⁷Np. However, the target was to be fabricated at the Chicago Metallurgical Laboratory and returned there after irradiation (Jordan 1945). Whether the irradiation took place was not found, but small-scale special irradiations in the Hanford reactors were common throughout their lifetimes.

The May 1948 200 Area monthly report listed as activities for the month "preparation for and crude separation of Neptunium from metal wastes of 385 g/ton material" (Christy 1948, p. 50). The June 1948 monthly report states, "170 liters of neptunium solution were shipped to Argonne National Laboratory" (Christy 1948, p. 71). It is surmised that this crude separation occurred at either T Plant or B Plant and, because purification of neptunium had not been developed at Hanford yet (see Section 2.3.2.2), it contained high activities of plutonium and FPs.

In 1957, Hanford was investigating the amount of ²³⁷Np in and potentially extractable from weaponsgrade spent fuel and investigating chemical procedures for extracting the neptunium at the fuel separations facilities (Sheppard and Becker 1957; Judson 1958). The Sheppard and Becker report indicates that small laboratory-scale samples were grabbed from the PUREX dissolution stream and analyzed for the ²³⁷Np content as early as November 1956 (Sheppard and Becker 1957, p. 3). Three small runs that simulated the PUREX chemistry conditions were run in February 1957 (GE 1957, p. 62).

2.3.3.2 Work with Significant Quantities

2.3.3.2.1 Separation of Neptunium from Normal Fuel

Extraction of neptunium-enriched nitrate solution from PUREX began on August 7, 1958, with two campaigns in 1958 (August and November) that resulted in 485 g of neptunium (Briggs 2001, p. 25; Travis 1958; Geier and Rathvon 1961, p. 1). Purification of neptunium was performed in a special room at the 222-S Laboratory called the 1-F Cubicle (Bihl 2008a; Buckingham 1964a, pp. 2, 3). The operation was described as mostly remotely handled in a hot-cell-like room, which required some hands-on work at the loading and unloading ends of the process (Bihl 2008a). Similar extraction started at REDOX in November 1959. This material was also purified at 222-S.

During the period of neptunium solution purification at 222-S, the incoming material from PUREX and REDOX was highly contaminated. For instance, impurity data on run numbers 12 through 58 (in 1958) and 40, 44, 46, 52, and 53 (circa 1961) show that the activity of plutonium, mostly $^{239+240}$ Pu, ranged from 3 to 81 times greater than the activity of 237 Np (Buckingham 1959, 1961). [Note: the plutonium impurities are recorded as "g Pu/g Np"; these were converted to activity using the specific activity for weapons-grade plutonium of 0.08 Ci/g and specific activity of 237 Np of 7.05 × 10⁻⁴ Ci/g.] [28]. However, the product of the purification work had low levels of contaminants; the activity of plutonium ranged from 0.3% to 4% of the neptunium activity for the six runs (Buckingham 1959, 1961) [29]. Therefore, during the part of the process that involved conversion of neptunium nitrate to oxide (done remotely) and especially loading of the neptunium product into shipping containers [more hands-on but done while wearing respirators (Bihl 2008a)], an intake of the neptunium would not have been readily detected by plutonium bioassay. According to Buckingham, all of this work was done by himself and a laboratory assistant with one other chemist helping on a few occasions. Also according to Buckingham, there were no incidents or high air concentrations that required special bioassay or that interrupted the processing (Bihl 2008a).

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In addition to the purification work at 222-S, laboratory-scale fume-hood-level research was conducted at the 325 Building in the 300 Area in late 1958 or early 1959 (Ryan 1959). This research on purification methods was eventually used in Q Cell at PUREX.

Starting in 1963, the purification of the neptunium was transferred to PUREX in J Cell and Q Cell, and purification at 222-S was stopped (Buckingham 1963, p. 3; Briggs 2001, p. 27; Duckworth 1963, Part 1, p. 2, Part 2, p. 1;CC 1963). Specifications for most of the shipments of neptunium from PUREX from August 1963 to May 1972 were found, but no data were found for the shipments in 1965 (GE 1965; Isochem 1967a; Isochem and ARHCO 1972; Malody 1971a,b,c, 1972a,b). The final product that was shipped from PUREX to the SRS was generally of similar purity to the earlier material that was produced at 222-S. Of the 84 shipment records that were found, the median plutonium activity in relation to neptunium activity greater than the neptunium activity [30]. For samples drawn from early stages of the separation and purification process the plutonium activity probably dominated the total alpha activity of the mixture, but for exposure to the final product plutonium would not have been a sensitive indicator of intake.

There is evidence that separation of neptunium at REDOX continued after the work at 222-S stopped, with concentration being done at the 233-S Building followed by shipment to PUREX for purification. This work was done intermittently throughout 1966 (Christy 1966a, p. 5; 1966b, p. 4). The December monthly report indicates that REDOX was gearing up for the final neptunium campaign (Christy 1967a, p. 5).

Some small-scale experimentation on neptunium separation techniques continued at 222-S in 1970 and 1971 (e.g., Schultz 1971 in ARHCO 1972b, p. 783).

The known records for PUREX mention that the neptunium nitrate was being shipped to the SRS. However, some of the nitrate was converted to oxide at the Plutonium Chemistry Laboratory in the PFP. The first mention was in a brief history of work in the Plutonium Chemistry Laboratory that covered 1952 to 1965. The entry is for August 1958 and states, "Neptunium oxide (<200 ppm metallic impurities) produced by oxalate precipitation and calcinations..." (Hopkins 1965, p. 26). Therefore, this work must have involved the first batch that was separated at 222-S. It appears that this material was then shipped to the University of California Radiation Laboratory (Hopkins 1958). Neptunium is not mentioned again in the Hopkins history through 1965, which implies no other work on neptunium was done in the Plutonium Chemistry Laboratory. Neptunium work started up again in 1966 (Dickeman 1966, pp. 8.9; Kingsley 1967a, title page; Kingsley 1967b, pp. 6–9; Christy 1966a p. 3; 1966c, p. 4). This material appears to be part of the effort to create a complete Palmolive project at Hanford (Section 2.3.2.3). The March 1967 monthly report by Kingsley (1967b, pp. 6–9) clearly shows that the neptunium nitrate came from PNNL and it had high activities of FPs (e.g., 1,000 µCi ¹⁰⁶Ru/g Np, 225 µCi ⁹⁵Nb-Zr/g Np). It has not been discovered yet when this work ceased. A Radiation Survey Card for the Development Laboratory (also known as the Plutonium Chemistry Laboratory) in 1969 mentions survey of neptunium samples in a glovebox, but it is not known if this was part of the oxidation project or just some leftover samples.

PUREX shut down in 1972, and neptunium separation was not done when PUREX restarted in the 1980s. Therefore, 1972 is the endpoint of work with neptunium nitrate in the 200 Areas.

2.3.3.2.2 Fabrication of Neptunium Target Elements

Neptunium-237 was irradiated to produce ²³⁸Pu. Fabrication of neptunium target elements would have involved handling the purified neptunium mixture from 222-S or PUREX Q Cell with little plutonium impurity.

Fabrication of neptunium target elements appears to have started in early 1959 at the 231-Z Plutonium Metallurgical Laboratory. Buckingham mentioned that either the nitrate or oxide was shipped to ORNL and SRS, but some of the oxide was mixed with aluminum powder and sent to the 231-Z Building for fabrication into neptunium-aluminum alloy target elements (Bihl 2008a,d). A February 1959 letter from A. T. Gifford, AEC Hanford Operations Office, seems to indicate that fabrication was occurring at 231-Z, although the legibility of the letter is poor (Gifford 1959a). Other Gifford letters authorize fabrication of the third and fourth batches of target elements, which implies that the first and second batches had been done or were in process (Gifford 1959b,c). Manufacturing of target elements is also mentioned by Judson and Beard (1959) but not by Judson and Kusler (1958), which narrows the start date to early 1959. A series of Radiation Monitoring Operations monthly reports sporadically mention work with ²³⁷Np fuel rods from January 1960 through April 1961 (Stevens 1960a,b,c, p. 9; 1961a,b, p. 10; 1961c, p. 9).

The next mention of fabrication of target elements is associated with another effort to get Hanford involved with the complete Palmolive cycle starting about 1966. This project started in late 1965 with a request to AEC Headquarters to hold back 5,000 g of neptunium from the regular shipments to SRS for irradiation in N Reactor (Williams 1965). The N Reactor irradiation program is explained in detail by Dickeman (1966). It involved obtaining a total of 5,000 g of neptunium in two batches (January and March 1966), target element fabrication as a joint activity by PNNL and General Electric, irradiation in N Reactor, dissolution in PUREX, separation and purification of the neptunium and the ²³⁸Pu by PNNL, conversion of the neptunium to oxide and shipment to SRS by Isochem (which was the operating contractor for the 200 Åreas at that time), and conversion of the ²³⁸Pu to oxide and shipment to Mound by PNNL. Gerber (1992, p. 186) states that the fabrication was done in the 308 Plutonium Fuels Pilot Plant and that neptunium oxide fuel targets were manufactured and canned in the 3708 Building in 1968 (p. 41). Subsequent documents indicate that the purification work by PNNL was done in C Cell in the 325 Building and the conversion of the neptunium to oxide was done at the Plutonium Chemistry Laboratory at 234-5Z, with the first elements being dissolved in October 1966 (Kingsley 1967a,b; Soine and Benedict 1968; Soine 1968; Roberts and Lerch 1968; Lerch 1968). Forty-four elements were fabricated and irradiated in N Reactor (Constable 1967). Constable's report of July 1967 is called the final report on the irradiation phase of the project, and the PNNL documents were all issued on October 1968, so it is concluded that the work on this project ended in 1968.

According to Wheelwright (1970), another round of irradiation and neptunium and plutonium separation occurred in 1969 and 1970 using a new dissolver in C Cell. The article does not say when the target elements were fabricated, but the semiannual report on process development in the Atlantic Richfield Hanford Company (ARHCO) for May 1 through October 31, 1969, states that 1 kg of neptunium was converted from nitrate to oxide during the reporting period for use by Douglas United Nuclear for target fabrication and irradiation studies (Isaacson and Tomlinson 1969, p. 46). The decanning occurred at the Radiometallurgy Building (327 Building) (p. 8), and the remaining work occurred in the A, B, and C Hot Cells at the 325 Building. The article shows that the leftover neptunium was oxidized at the 325 Building (p. 17) and returned to ARHCO (p. 19).

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2.3.3.2.3 Separation of ²³⁷Np from ²³⁸Pu After Irradiation of the Target Elements

The facilities and times of the two campaigns for dissolution of the irradiated neptunium targets are discussed above. The removal of ²³⁸Pu from the neptunium product was not highly effective and, because of the large difference in specific activities, the product had more (by activity) ²³⁸Pu than ²³⁷Np. For instance, Soine and Benedict (1968, p. 1) states that the ²³⁸Pu impurity in the ²³⁷Np product was 0.14% by weight. This converts to nearly 35 times more ²³⁸Pu by activity than ²³⁷Np [31]. Similarly, the ²³⁷Np product from the dissolution and purification work by Wheelwright was predominately ²³⁸Pu by activity with an activity ratio of nearly 40 (Wheelwright 1970, p. 19) [32].

2.3.4 Promethium-147

In the 1960s, Hanford was involved in extracting radionuclides from waste products, usually to produce heat sources. The pattern of development was similar in the cases of all isotopes.

Initial laboratory scouting studies were performed and flowsheet variables optimized with "cold" or tracer-level synthetic solution. This was followed with full-level proof-ofprinciple "brick pile" or "hot cell" tests on actual plant feed materials. Not infrequently, the intense radioactivity of these solutions or the presence of unsuspected impurities would require extensive modifications to achieve a workable process. Following hot cell testing and cold engineering development ..., plant tests were conducted by the Chemical Process Department. Further laboratory and hot cell work was often required at this stage, with very close day-to-day collaboration between the laboratory and plant personnel (Moore 1964, p. 3).

Promethium-147 was one of the most significant of these extraction and purification operations. Hanford was the first location in the world to produce significant quantities of pure ¹⁴⁷Pm.

2.3.4.1 Research, Development, and Early Tests

The earliest Hanford work found on ¹⁴⁷Pm separation was in 1960 by Wheelwright, presumably in the 325 Building because that is where he did most or all of his work (Wheelwright 1960). However, this work appears to be with simulated waste rather than actual FPs. It was common when developing new chemical flowsheets to start with cold simulated materials first, then work with trace or intermediate activities of the target material before trying actual waste products (Moore 1964, p. 3). The early Wheelwright work appears to be follow-on work to earlier development of a method to separate radioactive isotopes of cerium, promethium, lanthanum, yttrium, praseodymium, samarium, and niobium as a group from the PUREX 1WW waste (Wheelwright and Swift 1961) and separation of pure ¹⁴⁴Ce from the same material (Wheelwright and Howard 1960).

Similar work was mentioned in the July and August 1961 monthly reports of the Chemical Research and Development Organization (Reas 1961a, pp. C-4, C-5; 1961b, p. C-7). This work appears to be development of a flowsheet for use at Hot Semiworks (201-C Building, 200 Area) for recovery of a rare earth fraction free of strontium and cerium, which could be further purified to separate pure elements (e.g., ¹⁴⁷Pm). This work was done in the 325 B Cell with simulated PUREX 1WW waste.

Work with actual PUREX 1WW waste and purification of ¹⁴⁷Pm started in September and October 1961 and continued intermittently over the next several years. For instance, the September report of the Chemical Research and Development Organization mentions two B-Cell runs (Reas 1961c, p. C-7). The October report states that the second extraction of rare earths at PUREX was shipped to "the hot cells," and that B Cell was used to reduce the ¹⁴⁴Ce activity, the material then being moved to

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A Cell for promethium purification (Reas 1961d, p. C-4). About 40 kCi of ¹⁴⁷Pm were expected to be purified during this campaign. The November report adds that work at Hot Semiworks toward separating ¹⁴⁷Pm was abandoned because of poor yield; however, the run at A Cell successfully purified 26.6 kCi (Reas 1961e, p. C-6). Although the original intent appears to have been to purify the promethium at Hot Semiworks, it is not clear that any pure ¹⁴⁷Pm was extracted there. Purification at the 325 Building hot cells continued with emphasis on improving the chemistry. For instance, the February 1963 report (Reas 1963, pp. C-3–C-5, C-9) stated that laboratory work on the Cesium Strontium Rare Earth Extraction CSREX) process included studies that were aimed at improving the kinetics of cesium and promethium extraction with hot cell experiments with full-level PUREX 1WW waste. The report added that "analytical results verified last month's conclusion that the 'heart cut' of over 10,000 Curies of promethium from the recent C-Cell ion exchange purification run is indeed chemically and radiochemically pure."

Associated research on promethium was being conducted in 1963, such as determining the decay energy and magnetic susceptibility (Wheelwright, Fleming, and Roberts 1963; Sheppard, Wheelwright and Roberts 1963). All this work appears to have been conducted in the 325 Building.

Wheelwright and Roberts (1963) summarized the various activities to that date. The earliest runs were "cold." Runs in 1961 were hot with about 85 Ci/L¹⁴⁷Pm and several times that of ¹⁴⁴Ce and ⁹⁰Y. The product of this run was apparently not sent anywhere, and in January 1963 was repurified in C Cell "as a demonstration of the capability of the process to yield high purity promethium and to highly purify 10,000 Ci for transmission to Oak Ridge at the request of the Atomic Energy Commission" (Wheelwright and Roberts 1963, p. 26).

In February 1963, there was a major contamination spread of ¹⁴⁷Pm during work on the 10,000 Ci mentioned above (AEC 1963).

In the 329 Building in 1962 and 1963, research was done to determine the specific power and half-life of ¹⁴⁷Pm (Fleming 1964, pp. 2.40–2.41). According to the author, this work was done with a clean, encapsulated, sealed source having about 1 kCi of activity. A sealed high-activity ¹⁴⁷Pm source was subjected to neutron irradiation in a Van de Graaff accelerator in the 3745-B Building in February 1963. Contamination up to 400 cpm was found in the tube where the source was housed for the irradiations. There was no mention in the radiation monitoring report of personal contamination or high airborne contamination (Stevens 1963a). Gerber states that chemists in the 321 Building were involved in prototype separation chemistry, but only tracer levels were used for the prototype experiments (Gerber 1992, p. 75). No timeframe for this work was mentioned. Gerber also mentions that work was done in the 327 Radiometallurgy Building in support of the ¹⁴⁷Pm campaigns (next section) (Gerber 1992, p. 154).

2.3.4.2 Major Production

In early 1964, a feasibility study was conducted for installing equipment in Strontium Semiworks (Hot Semiworks or 201-C) for large-scale purification of ¹⁴⁷Pm from either PUREX 1WW or waste from the separation of ¹³⁷Cs and ⁹⁰Sr operation at B Plant (Beard, Boldt, and Kendall 1964). The same report indicated that pilot-scale test equipment was "now being installed in the 222-S 1-F cubical" (p. 1). The 1964 summary report on isotope separation projects stated that "to date" 20,000 Ci of ¹⁴⁷Pm had been purified at the High Level Radiochemistry Facility (the 325 Building hot cells) and equipment had recently been installed in the 1-F cubical at 222-S for expected purification of about 1 MCi of ¹⁴⁷Pm. Plans were being scoped for changes to Strontium Semiworks for routine processing of ¹⁴⁷Pm on a production scale (Moore 1964, pp. 13–14). The hazards review for the work at 222-S on July 15, 1964, indicates that the work had not started (Buckingham 1964b,c). J. S. Buckingham's personal

letterbook indicates that the first ¹⁴⁷Pm run at 222-S started in October 1964 and ended in November, with a second run starting in November (Buckingham 1964d). A letter from the 222-S radiation monitoring manager from November 1964 indicates that purification of ¹⁴⁷Pm is scheduled for "one run to be processed for each month for the next four to six months…," and provides a list of seven workers to be given ¹⁴⁷Pm bioassay (Glover 1964). The end date for the work at 222-S was not precisely determined, although Buckingham left 222-S to work on B Plant and Waste Management issues in (approximately) September 1965 and said he was pretty sure the promethium work did not carry on much longer after he left (Bihl 2009a).

In 1965, the Pacific Northwest Laboratory (now PNNL) was created to include most of the R&D scope from the old General Electric contract, and the work in the 325 Building became part of the laboratory. Work continued toward major production of ¹⁴⁷Pm sources in the 325 Building. Moore provided a summary of promethium isolation and purification steps as of June 1965 with an indication of future plans (Moore 1965, pp. 3–4). These included (1) separation of a crude concentrate at PUREX or B Plant, (2) aging, (3) chemical processing to remove some contaminates (e.g., iron, sodium, and lead) and concentrating the product before shipping to the 300 Area, and (4) final purification of promethium in the 325 Building hot cells. Moore indicated that steps 1 and 2 were underway, with the material being stored pending completion of upgrades to A Cell in the 325 Building (Moore 1965, pp. 3_5). Moore also indicated that steps in encapsulation of sources were to be done in 325, 308, and possibly 309 Buildings (Moore 1965, pp. 7–8). The Monthly Activities Report for May 1965 indicated that the installation of ¹⁴⁷Pm purification equipment "has been essentially completed in A Cell of the High Level Radiochemistry Facility" (PNL 1965a, p. 21). This apparently was the upgrade Moore mentioned in his summary.

The January and February 1966 reports for the 200 Area show about 1.7 and 3.0 kCi, respectively, of ¹⁴⁷Pm being extracted from PUREX and B Plant (Christy 1966c, p. 8; 1966d, p.10). The March report stated, "At the Strontium Semiworks, promethium processing was started. By month end, two runs had been completed with 730 kCi Pm-147 recovered in the first run and 1,250 kCi Pm-147 recovered in the second run." These were the first-step separations that included other desirable FPs as well: about equal activity of ¹⁴⁴Ce and "greater than 95% of the americium-241 was also recovered" (Christy 1966c, p. 11). The purification and fabrication work in the 300 Area appears to have started in March or April 1966 (Vogel 1968; Fawcett, Albaugh, and Paul 1966, p. 53), although Howell and King stated that it started in the summer of 1966 (Howell and King 1968, p. 6). The Howell and King document described the process equipment and radiation control practices in detail. In addition to the hot cells, gloveboxes were used in the final steps of building of the heat sources, including the conversion of the nitrate solution to oxide, firing, and compaction of the oxide (p. 2). The document also stated that final encapsulation as a heat source was "accomplished in another glovebox of similar construction in a different building" (p. 2). It also mentioned that development work with low-concentration solutions of promethium was being done in fume hoods as well as some development of encapsulation methods. "No significant contamination control problems have resulted from any of the work in the fume hoods" (p. 2).

Some promethium work was done in the 308 Building in 1969, and these workers had bioassay (Howell 1969; Jech 1969). The 308 Building is probably the "other building" mentioned in Howell and King (1968).

2.3.5 <u>Highly Enriched Uranium</u>

The first known mention of work with ²³⁵U was in the 313 Building in July 1948 (GE 1948, p.139). The report states, "Special process material containing U-235 was received and canned in special pieces. Special containers were used to keep the number of slugs stored in one place to less than thirty. No

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contamination was encountered and exposure-rates were less than for natural uranium." The slugs were irradiated in D and F Reactors, and shipped back to ORNL (Gross 1948).

Hanford canned and irradiated fuel elements that were referred to as J slugs and C slugs between 1949 and 1956 (Burley 1956, p. 722; Gifford 1956). J slugs were a uranium-aluminum alloy with about 7.5% uranium by weight, of which about 93% was ²³⁵U. C slugs were a uranium-aluminum alloy with about 4% uranium by weight, of which about 93% was ²³⁵U (Devlin, Palmer, and Upson 1958, pp. 4, 9; Burley 1956, p. 722). These slugs were manufactured at ORNL, canned at Hanford, irradiated in several Hanford single-pass reactors, and shipped to the Idaho Chemical Processing Plant at the Idaho National Laboratory (INL; then called the National Reactor Testing Station) for dissolution of the slugs and recovery of the uranium (Burley 1956, pp. 722–725, Figure VII-22). Canning of C slugs stopped in December 1953 (GE 1954), but a considerable number of C slugs were still in reactors in 1957 and were scheduled for discharge through 1958 (Priode 1957). Priode (1957) also indicates that the slugs would be cooled in the fuel storage basin for about 4 months before shipping, which would put the expected date for removal from the basins as late in 1958.

For the C and J slug program, exposure to HEU would have occurred to workers in the 300 Area canning facility in 313 Building (Gerber 1992, p. 9). It is reasonable to expect that testing of canned slugs would have occurred in the 314 Building before shipment to the reactors, as was done for other slugs [33]. Burley (1956, p. 1006) states that samples of new slugs that were received on the 313 Building dock were sent to the 305 Building for testing.

Outside of the C and J slug program, there was no major work with HEU, but there were quite a number of small or short-term usages.

The August 1958 Radiation Monitoring Operations report mentions work with 8% enriched uranium in the 325 Building (Stevens 1958, p. 9).

The February 1961 Radiation Monitoring Operations report briefly discusses an experiment in the 326 Building with "93% ²³⁵U per gram" of UO_2F_2 solution (Stevens 1961d, p. 14). This appears to be a single experiment that was not carried over to other periods. The work was carried out in an exhausted greenhouse, and leaks of the solution did occur during the work.

Under a Metallic Fuel Development Program, two small zirconium-clad extrusions with thorium and a small 93%-enriched uranium core (2.5 wt% of the core) were produced in January 1963 and examined metallurgically in February (GE 1963a, p. A-48). The buildings where the manufacturing and examining took place were not mentioned. Follow-up work with thorium-uranium cores used normal uranium (GE 1963b, p. 56).

Extrusion of a 50-lb thorium ingot with 1.5 wt% of 93%-enriched uranium was performed in June 1963 (GE 1963c, p. A-9). The building was not given, but extrusions like this were generally done in the 314, 306, or 333 Building and the documentation seems to be associated with work on N Reactor fuel, which was done in the 333 Building.

Extrusion of a small (~1–in.-diameter) rod with a thorium annulus that contained 1.5 wt% "fully enriched" ²³⁵U was conducted in July 1963 (GE 1963d, p. A-6).

A fuel capsule that contained 50%-enriched uranium was fabricated in August 1963 for experimental tests in the INL Engineering Test Reactor (GE 1963e, p. A-24).

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A tungsten-uranium cermet (ceramic-metal alloy) fuel plate using 93%-enriched uranium was irradiated and examined in September and October 1963 (GE 1963f, p. A-25; 1963g, p. A-31). The September 1963 radiation monitoring report mentions a slight spread of contamination in the Ceramic Fuels section of the 325 Building basement that involved ²³⁵U oxide (Stevens 1963b, p. 9), probably associated with the cermet work.

A small amount of ²³⁵U was irradiated in DR Reactor in October (GE 1963g, p. C-23).

Work on a long-life, constant heat source for a crud probe was started in February 1964. This involved creating a thorium-uranium-zirconium alloy with 1.5 wt% fully enriched uranium. The building where this work was done was not mentioned (GE 1964a, p. A-4).

Experiments in the PRCF (309 Building), which began operation in 1962, sometimes involved test fuels with HEU. For instance, the Hanford Laboratories report for May 1964 mentions planning for an experiment with a "plastic-moderated U-235 lattice" using uranium-aluminum disks with 93.5% enrichment (GE 1964b, p. B-12). The July 1964 report discusses irradiation of a UO₂-niobium cermet in which the uranium was 85% enriched (GE 1964c, p. A-20). The Hanford Laboratories report for August 1964 mentions creation of an experimental UO₂-PuO₂ fuel in which the uranium was 93% enriched. The work involved milling and compacting of 1 kg of the material (GE 1964d, p. A-21). The building where this work took place was not mentioned, but the 308 Building is likely because its mission was to support R&D on fuels for the 309 Building reactors. Examination of the test fuels might have occurred in the 309 Building Hot Fuel Examination Facility (Gerber 1992, p. 191).

A series of experiments were conducted in the Physics Constants Testing Reactor (305-B Building) on polyethylene moderation using ²³⁵U-Al fuel in about late 1964. The amount of ²³⁵U in these experiments was not mentioned (GE 1964e, p. B-16). The report does not state where the fuel was made. The nature of the work would have been consistent with the missions of either the 306 or 333 Building (Gerber 1992, pp. 15, 19, 20).

Blending of various oxides for experimental work for a non-Hanford client was conducted in or about September 1965. A total of 850 g of PuO_2 - UO_2 powders were crushed, screened, and blended. Some of the uranium in this work was nominally 93% enriched and some was nominally 40% enriched. The building where the work was performed was not given (PNL 1965b, p. H-5).

Also in 1968, there was experimental work with HEU fuel in the Physics Constants Testing Reactor (305-B) from June through September (Albaugh 1968a, pp. 7, 17–18; 1968b, pp. 6–7, 20–22; 1968c, pp. 19–20.]). The reports do not state where the fuel was made, but the May report does mention machining by the Battelle Engineering Services Department (Albaugh 1968d, pp. 9, 23).

A radiation work permit was issued for work in the 306 Building for 37%-enriched uranium (PNL 1970; Jech 1970) that was scheduled to last several years.

There is indication that some work with ²³⁵U was performed in the 231-Z Building sometime during its history, but details have not been found (Powers 1969; Lini 2008, p. 23).

2.3.6 <u>Uranium-233</u>

Uranium-233 was the intended product of the program to manufacture and irradiate thorium target elements.

2.3.6.1 Early Irradiation of Thorium, Probably No Dissolution at Hanford

Thorium rods were used in the reactors starting in 1946 for various purposes (Wende 1946a,b). As of September 1947, an estimated 500 mg of ²³³U had been created at Hanford and had been separated in laboratory-scale operations (Author unknown ca. 1947, p. 1). The location of the separation was not given, although no evidence has been found that the separation was conducted at Hanford. Thorium continued to be used in the reactors, such that by the end of 1953 thorium elements were in use in all Hanford reactors as flux-flattening poisons that replaced lithium-aluminum and bismuth (Sturges 1951; DeNeal 1970). Extraction of the ²³³U took place at another site (probably ORNL) (Sturges 1951; Gydesen 1954, pp.11, 17). The Briggs history stated that thorium was being irradiated for production of ²³³U in August 1954 (Briggs 2001, p. 21). It is unlikely that any exposure to ²³³U occurred to Hanford employees from these activities.

There is evidence of work with ²³³U at 231-Z circa 1957 or 1958 that involved fabrication of fuel rods for the INL Materials Test Reactor (Baumgartner ca. 1957; Powers 1969, p. 21; Lini 2008, p. 23.)

2.3.6.2 Major Production of ²³³U

Hanford conducted major-scale, full-cycle ²³³U generation and extraction from thorium in two campaigns at PUREX in May through July 1966 (producing 220 kg ²³³U) and August through December 1970 (producing 320 kg ²³³U), with a smaller scale experimental run in January 1965 (Isochem 1967a, p. II-1; Jackson and Walser 1977, pp. 7, 8). The ²³³U product was loaded into approximate 3-L bottles in N Cell in PUREX, transported to 221-U for temporary storage, and shipped off the site (principally but not exclusively to ORNL). Some rebottling of product occurred in a specially constructed metal enclosure at the northeast end of the pipe gallery; this was ventilated with a blower-filter arrangement (Isochem 1967a, p. III-54; Jackson and Walser 1977, pp. 52–54; ARHCO 1971).

Some or all product from the 1965 experiment was sent to the Plutonium Chemistry Laboratory in Z Plant (Fullam 1965, pp. 3–4; Hopkins 1965, p. 68). The purpose was:

- 1. Experimental evaluation of the various steps of the proposed Z Plant flow sheet for converting U^{233} nitrate solution to metal.
- 2. Purification of 1 kilogram of U²³³ produced in the first thorium run at the PUREX Plant.

According to Fullam (1965), approximately 1,250 g of ²³³U were purified and converted to oxide. It is likely that the experiment was conducted between February 1965 (the end of the first thoria/²³³U campaign in PUREX) and September 1965 (the date of Fullam's article). A letter from J. T. Christy to A. T. Gifford in April 1965 provided a long list of customers for the ²³³U shipments and indicated that 102 g of oxide had already been shipped; however, there was trouble getting sufficient ²³³U to fulfill requests in the next term (Christy 1965). Hopkins (1965), which is a history of work conducted at the Plutonium Chemistry Laboratory, did not mention any other work on ²³³U other than the Fullam work in 1965. There is no evidence that the process moved beyond this single evaluation; that is, that Z Plant was actually used to convert ²³³U nitrate to oxide for the other PUREX campaigns. For instance, the 200 Area report for August 1966 said that the ²³³U from the 1966 campaign was being shipped to ORNL (Christy 1966b, p. 3). In addition, a January 1966 report mentioned the earlier work and implied that flowsheet development was still an "objective," but only minor work was being done at that time because neptunium work was a higher priority (Curtis 1966, pp. 1–2).

2.3.6.3 Research and Development

Walling (1964a, p. 4) reported on chemistry development work on ²³³U in February 1964. This work involved millicurie activities. PNNL Chemistry Department monthly reports in 1965 indicated studies on ²³³U separation techniques were occurring (probably at the 325 Building). The work consisted of tracer-level flowsheet development studies (Walling 1965a, p. 7-C; 1965b, p. C-5; 1965c p. C-3; PNL 1965c, p. A-30). Similarly, the activities reports for April, May, June, and July 1966 indicated that work on a flowsheet for removing thoria from the ²³³U stream had been experimentally validated and other studies were occurring (PNL 1966a, p. 42; 1966b, pp. 50, 51; 1966c, p. 47; 1966d, pp. 37, 40, 41). The June report indicated that these were tracer-level studies: "... simulated feeds spiked with appropriate tracers for analytical purposes were used" (PNL 1966c, p. 47). These studies do not appear to continue into the second half of 1966 or in subsequent years [34]. An exception to the tracer-level work was a criticality mass experiment in July 1966 that involved subcritical neutron multiplication experiments with bottles that contained approximately 960 g of ²³³U each (9.3 Ci) (PNL 1966d, pp. 40, 41). During the experiments the bottles were undoubtedly sealed, but information has not been found about where the bottles were filled. If the material came from the May to July 1966 PUREX campaign, the bottles would have been filled either in PUREX N Cell or at U Plant before the product was sent to ORNL. It is also possible the bottles came directly from ORNL (that is, they could have been independent of the recent PUREX campaign).

The June 1967 PNNL report discussed a criticality experiment with bottles of uranyl nitrate that contained both ²³⁵U and ²³³U. However, it does not appear from the brief description that the bottles were opened or any chemical analysis was done (PNL 1967d, p. 71).

2.3.7 <u>Polonium-210</u>

Polonium-210 (also called "chemical 14-92" or "postum") was generated by the activation of bismuth metal (sometimes called "chemical 37-77," "soda pulp," or "B Metal") in the Hanford reactors.

2.3.7.1 Production Chronology

Hanford started irradiating bismuth to produce polonium in 1945.

Bismuth rod suitable for canning will be delivered by the Government to Hanford where the material will be placed in cans suitable for exposure in the pile.... After completion of irradiation, du Pont will place the irradiated pieces in containers furnished by du Pont and turn the packages over to the Area Engineer for shipment.

The material was to be sent to Monsanto in Dayton, Ohio (Tilley 1945).

To get advance information six bismuth slugs were inserted in a P-column in the B-pile during the shutdown of February 22, which will be removed on March 7. We will keep three of these to follow their decay, so as to be sure that the shielding provided on the shipping containers is adequate. Two of the remaining three slugs will have their jackets stripped off, using the stripper for unbonded cans in the 300 Area. They will then be shipped with the remaining jacketed slug in one of our regular bismuth containers, W-76138, to Lum [Ed: at Monsanto] who will assay them for their polonium content and use the regular jacketed one in his program of learning how to deal with jackets when the regular shipments come in. Lum expressed a preference for unjacketed slugs but has agreed to accept jacketed ones for the main shipment. The

results of the assay should be available three days after arrival of the metal in Dayton (Worthington 1945a; also discussed in HEW 1945a, p. 32).

Equipment was assembled for stripping the aluminum cans from some of these slugs before shipping for an assay of the polonium content (HEW 1945b, p. 31; HEW 1947, p. 3; Worthington 1945b, p. 9). This work was also mentioned in the Health Instruments report for March 1945, "The survey program ... covered the handling of special bismuth slugs. Uncanned slugs were a potent beta source ... in addition to the hazard of polonium contamination in the air. The canned slugs presented no handling problems" (Parker 1945, p. 16). This is the only example of jacket removal and manipulation of Hanford bismuth slugs that has been discovered; all others seem to have been shipped in intact condition [35]. The opened slugs were subsequently shipped to Monsanto as well, "in order to protect from the alpha active vapors which are known to be discharged from uncovered activated bismuth metal, the shipping container was hermetically sealed..." (Grane 1945, p. 3; Wheeler 1945; HEW 1945c, pp. 26–27).

After these initial measurements, shipments of irradiated bismuth became routine.

On 5 May 1945, the first relatively large-scale shipment of soda pulp was made from the 100-B Area substantially in accordance with the program outlined in the conference held in Richland on 20 February 1945. The shipment consisted of seventy units of soda pulp which had been charged in the 100-B pile on 9 March and discharged on 26 April. As a result of the neutronic irradiation, a quantity of postum was produced which was estimated as equivalent to 210 cases (HEW 1947, p. 8). ("Case" is code for curie [36].)

Canning of bismuth metal was done in the 313 Building. Once canned, the bismuth was guarded. "No change occurs in the physical description of the material other than irradiation." It was placed in containers after discharge, put in a box weighing about 950 lb, and shipped by military air transport from Pasco to Monsanto and Mound Laboratory (George 1947). To minimize material requirements, the idea of recycling irradiated bismuth was investigated. It was determined that there would be too many problems with contamination in the production and canning of slugs, so recycled bismuth was not used (Lee 1949).

Bismuth irradiation for polonium production continued as a routine part of Hanford operations through the 1950s and 1960s. Many documents illustrate the ongoing shipment of irradiated bismuth from Hanford to Mound (e.g., Priode 1953, 1954a,b, 1958; Gifford 1958). Bismuth was being irradiated in B, D, and F Reactors, and it was planned for H Reactor by 1949 (Lee 1949). Discussions for bismuth irradiations in N Reactor occurred as early as 1963 (Dieterich 1963). It can be assumed that polonium was produced in all Hanford reactors.

2.3.7.2 Separations and Use of Polonium

Demand for ²¹⁰Po for the Space Nuclear Auxiliary Power generator led to the desire to have a complete polonium supply capability at Hanford. Many feasibility studies and proposals were conducted (e.g., Fry 1956; Johnson 1963a,b; Richardson, Sheppard, and Guay 1965; Hill 1966; Hasty and Sheppard 1965). However, none of the proposals was funded and routine production of ²¹⁰Po did not take place at Hanford. Small-scale studies or special-order production of ²¹⁰Po did occur.

Some efforts were undertaken in the Hanford Laboratories Biology Department in 1955 to investigate the relative radiotoxicity of polonium to yeast cells (GE 1956, pp. 93–102). Survey reports from January and May 1955 indicate that work was occurring in a glovebox in the 108-F Building, Rooms

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105, 106, and 107, and generating small amounts of waste (GE 1955, survey numbers 550132 and 550887).

A series of experiments of irradiated "B Metal" was undertaken by the Chemical Laboratory in 1963 in which canned and uncanned bismuth slugs were heated to 1,300° for 1 to 2 hours. It was found that polonium vapors would be released from liquid bismuth metal, and that the bismuth metal would damage the aluminum cans and cause leaks (GE 1963a, p. C-3; 1963b, p. C-2; 1963c, p. C-1; 1963h, p. C-1; 1963i, p. C-2; 1963j, p. C-2).

A great deal of laboratory work was undertaken in the early to mid-1960s to develop mass production methods for polonium separation from bismuth. Numerous approaches were evaluated, beginning with an examination of requirements for handling of recycled bismuth in open hoods without gloveboxes (GE 1964f, p. C-3; Walling 1964b, p. 4):

- Pyrometallurgical processes that involved molten bismuth with cerium scavenging (GE 1963k, p. C-5, ; Walling 1964c, p. 4; Walling 1964d, p. 4; GE 1964g, p. C-3),
- Volatilization of mercury polonide (GE 1964h, p. C-4),
- Molten salt separations (Reas 1964a, p.4; Sheppard 1964; PNL 1965a; Sheppard 1966),
- Volatilization of polonium from nitric acid solutions (Walling 1964e, p. 6; 1964c, p. C-3),
- Solvent extraction of polonium with various solvents (GE 1964a, p. C-5; 1964c, p. C-3; 1964e, p. C-3),
- Distillation (GE 1964e, p. C-3),
- Electrochemistry (GE 1965, p. C-2, C-3), and
- Pyrochemical separation in fused NaOH from molten bismuth (Fawcett 1967a, p. 58; Albaugh 1968a, p. 50-51; 1968b, p. 29, 43-44).

The patent application in HWIR-1793 reports that "polonium volatility from the oxide has been examined…" in the 325 Building (Hasty 1964). A lot of this work was done by J. C. Sheppard, who had offices in the 325 Building (Sheppard 1964; Sheppard and Warnock 1962). Other key investigators were R. H. Moore, G. F. Shieffelbein, and R. A. Hasty. These investigations are supported by the interview with L. Bray (Bihl and Napier 2008), who recalled that ²¹⁰Po was purified in the 325 Building under a contract with the 3M Company. The polonium work was all done in gloveboxes in the center hall of 325 Building.

During the mid-1960s, complementary studies were being undertaken in the 200 Areas – small-scale engineering experiments on solvent extraction in the 200 Area Chemical Research and Development organization (Tomlinson 1965, p. 8; Christy 1966c, p. 12). It is assumed that this Chemical Research and Development work occurred in the 222-S Building [37].

PNNL did attempt one major program for the investigation and development of polonium microspheres in 1968 and 1969. A series of monthly reports by J. E. Hansen (1968a,b,c,d; 1969a,b,c,d) and Drumheller (1968a,b) describes the installation of a glovebox line to make ²¹⁰Po microspheres, ²¹⁰Po radiation measurements reported by H. A. Van Tuyl and W. Howell, vacuum

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deposition and fueling studies, and other preparations. However, the program terminated prematurely around April 1969. The Laboratory received only about 1 g (Hansen 1969b).

2.3.8 <u>Curium</u>

There was interest in extracting both ²⁴²Cm and ²⁴⁴Cm from spent fuel waste as heat sources or for other uses. Interest in the americium and curium content of irradiated Hanford fuel dates to at least 1950 (Albaugh 1950), but no evidence was found that anything was done in the 1950s other than estimates of how much curium was in the spent fuel.

2.3.8.1 Research, Development, and Proposals

Interest picked up again in 1963 as evidenced by letters between the AEC Richland Operations Office and AEC Headquarters that discuss quantities of ²⁴¹Am and ²⁴²Cm in waste streams from PUREX and REDOX (Travis 1963a,b). Travis states that at that time all of the americium and curium in spent fuel was going to the waste tanks.

In 1966, AEC Headquarters took the next step by asking for a proposal for major production. "Division of Production [AEC] has requested that study proposal be submitted by both the Savannah River Plant and Hanford for production of large quantities (up to 70 kg by 1974) of curium-244" (McKenzie and Watrous 1966). Hanford proposed a five-step plan to create and irradiate ²⁴²Pu to produce ²⁴³Am, then irradiate ²⁴³Am to create ²⁴⁴Cm. The plan involved separation of americium and curium at B Plant using the D2EHPA method (which would also contain rare earths, e.g. promethium), conversion of the nitrate to oxide at the PFP, and final extraction of ²⁴³Am and ²⁴⁴Cm at a new facility to be built at Z Plant (236-Z). Fabrication of interim target elements of ²⁴²Pu and ²⁴³Am was to be done by Douglas United Nuclear (presumably at the 300 Area fuel fabrication facilities) (McKenzie and Watrous 1966; Beard 1967, p. 97). [Note: The extraction facility at Z Plant was not built and the crude extraction was not done at B Plant, but rather at 201-C Hot Semiworks as discussed in the next section.]

Flowsheet development also started in 1966. For instance, the July report of the Fission Products Processing Engineering Group indicated that flowsheet work was being done at PNNL looking at methods to separate curium from americium using the D2EHPA method available at Semiworks (Beard 1967, p. 114). The August and September reports (Beard 1967, p. 128; 1967, p. 148) discussed upcoming dissolution of the Shippingport fuel at REDOX and possible extraction of the ²⁴⁴Cm from the resultant solutions. The plans for this work were still in development in December (Beard 1967, p. 198). Roberts and Bray (1966) published the flowsheet in December. In that article, they stated that they started with simulated waste with tracers (no curium was used in the tracer experiments), then progressed to experiments with actual Shippingport waste solutions in hot cells in the 325 Building (Roberts and Bray 1966, p. 5). The incoming solution had plenty of gamma-emitting FPs, but the final extractions had ²⁴¹Am, ²⁴⁴Cm, and ²⁴²Cm in about 2:3:1 ratios and only small activities of FPs. This was still small-scale flowsheet development and testing work (Roberts and Bray 1966, pp. 1, 7, 8).

PNNL involvement in R&D was also documented in the series of monthly reports for the Division of Production and Hanford Plant Assistance Programs during this period. These reports discussed flowsheet development and other assistance to Isochem starting in July 1966 and running through September 1967 (Fawcett 1966a, p. 45; 1966b, p. 36; 1967a, pp. 64–65; 1967b, pp. 35–36; 1967c, pp. 39–40; 1967d, pp. 50–51; 1967e, p. 47). Most of the development work during this period involved tracer levels of americium and curium (Fawcett 1967d, pp. 64–65), but some hot cell work

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was done as well in preparation for using the hot cells for final large-scale extractions (Fawcett 1967f, p. 72). All of the work appears to have been done in the 325 Building.

2.3.8.2 Major Production of Curium

Although there were plans to irradiate target elements to create curium, no evidence was found that this approach was ever taken. Rather, FP waste from dissolution of Shippingport fuel was used as the source for curium. Dissolution of the Shippingport fuel occurred in early 1967. For instance, the February 200 Area report stated that an americium-curium solution from REDOX had been loaded into a "bowling ball" cast and "shipped to PUREX for storage until Hot Semiworks is ready to process the material" (Christy 1967b, p. 4). The Shippingport waste solution was then processed at Hot Semiworks and delivered to PNNL on September 18. This material still contained rare earths, americium, and curium (Fawcett 1967e, pp. 47–48). Meanwhile, development work was still going on to refine the flowsheet to be used on the material that was delivered to the 325 Building hot cells (Fawcett 1967e, p. 47). Full-scale, hot-cell processing of the Shippingport rare earth-americium-curium solution started in December 1967 (Albaugh 1968e, p. 67; 1968f, pp. 69–70). The pure curium nitrate product was converted to oxide and placed in storage in April and May 1968 (Albaugh 1968g, p. 66; 1968a, p. 67). A total of 41.6 g of curium was prepared for shipment. Shipping began in June 1968; subsequent monthly reports did not mention when shipping concluded but presumably it was within a few months.

Hot Semiworks shut down in 1967 (Ballinger and Hall 1991, pdfp. 62), apparently soon after processing of the Shippingport solution. No evidence was found that the plan to use B Plant to extract americium and curium either as a crude solution for further processing at the 325 Building or to create pure products was ever carried out. A 1974 document indicates that the Shippingport curium purification campaign was the only one carried out at that time (Wheelwright et al. 1974).

2.3.8.3 Other Research

PNNL's Space Nuclear Systems Division reported on laboratory work in March 1969 on phase transformations in curium oxide in which a small sample of curium oxide was heated in a vacuum in a differential thermal analysis apparatus (Hansen 1969c, p. 4). The building where the work took place was not mentioned. One of the authors was interviewed briefly by phone. He said that he and a few others worked with milligram quantities for special studies in the 308 Building between about 1969 and 1973. All the work was done in gloveboxes. Each experiment was of short duration (a month or so), but there were multiple experiments. The curium was not from Hanford. Because quantities of curium and other higher order transplutonium elements were scarce, who, when, and how much of these special isotopes was made available to researchers was controlled by the AEC-wide Transplutonium Research Coordinating Committee. That committee arranged for delivery of the samples, which were used then shipped to the next researcher on the priority list. The researcher said there were no contamination spreads with the curium samples (Bihl 2009b).

2.3.9 <u>Technetium</u>

Technetium-99 was extracted from PUREX waste in kilogram quantities. The interest in technetium was not based on its radiation characteristics because ⁹⁹Tc has a small specific activity and emits only a very soft beta (average energy 0.1013 MeV). Interest centered on the fact that technetium does not exist in nature and it was thought that it might have useful chemical and physical properties sufficiently unique to make its recovery attractive. In particular, it was evaluated at Hanford Laboratories (for the National Aeronautics and Space Administration) for use as an alloying agent to impart ductility to tungsten (Moore 1964, pp. 15–16).

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The earliest evidence of ⁹⁹Tc extraction from Hanford waste was an invention report by F. P. Roberts and F. M. Smith (1960). The stated first date of entry into a record was June 20, 1960. The report indicated that 100 mL of 3.5-yr-cooled, neutralized PUREX waste supernate was used as feed. The work was done in the 325 Building. Although about 0.2 mg of technetium was isolated, it was still heavily contaminated with ¹⁰⁶Ru (0.5% wt% or about 3 mCi of ruthenium versus 3 µCi of technetium) (Roberts and Smith 1960, p. 3).

Work continued at an R&D level in 1961, but the early work did not produce pure ⁹⁹Tc. For instance, the August 1961 report of the Radiation Monitoring Operation stated that the pilot recovery program for ⁹⁹Tc had begun in the 325 Building (Stevens 1961e, p. 7). The report stated that handling of quantities of pure technetium would not be encountered, however, thereby decreasing the radiation monitoring difficulties that are normally expected with low-energy beta emitters. The reason the technetium was not pure was not given, but from a radiation monitoring perspective the product apparently had enough gamma-emitting contaminants to allow normal monitoring methods for contamination control. The December 1961 report of the Chemical Research and Development Operation (Reas 1962, p. C-5) stated that the technetium recovered by anion exchange from the PUREX alkaline waste supernate required additional decontamination before it could be handled without shielding. The report also stated that further flowsheet development had been done and a large-scale run was in progress in A Cell of the 325 Building.

Work continued at both an R&D level and a full-scale level from 1962 through 1964. The November 1963 report of the Hanford Laboratories reported that successful absorption of about 1 kg of ⁹⁹Tc from PUREX alkaline supernate on an anion exchange bed had been accomplished at the Hot Semiworks (201-C) in October, and in November the material had been evaporated to a volume of 100 L and shipped to Chemical Research for final processing (GE 1963k, p. C-10 pdf p. 143). Complications included high concentrations of ⁹⁵Zr/Nb and ¹⁴⁴Ce. By March 1964, methods to remove the FP contaminants had been worked out in laboratory studies and work had started in the 325 Building to purify the shipment from Hot Semiworks. By May 1964, the purification of the technetium had been completed in the 325 Building (GE 1964f, p. C-6pdf p. 130). It consisted of 1,540 g of ammonium pertechnetate with a radiochemical purity of greater than 99.99% with metallic impurities of less than 50 ppm based on spectrographic analysis (Reas 1964b, p. 1pdf p. 5). By July this material had been converted to metal and 800 g had been transferred to the Metallurgy Development Operation (presumably 327 Building) for tungsten alloying studies, including arc melting (Reas 1964c, p. 1). (There was no mention of where the remaining 200 g of the 1-kg quantity was sent). The product was extremely clean from FPs with only one count of ruthenium present per 10⁴ counts of technetium (Moore 1964, p. 16).

A second 1-kg campaign started at Hot Semiworks in September 1964 (GE 1964b, p. C-6pdf p. 120). However, the second campaign was mentioned only peripherally in subsequent monthly reports, so when or if this material was purified and turned into metal is not well-established (GE 1964i, p. C-5pdf p. 127). Research on technetium pertechnetate continued through December 1964 (GE 1965, p. C-4pdp p. 127).

Fifty grams (~850 mCi) of ⁹⁹Tc were shipped to Brigham Young University on June 5, 1967. This represents the first offsite shipment of gram quantities of technetium from Hanford (Isochem 1967b, p. A-2). That this shipment was mentioned in the Isochem monthly report along with shipments of neptunium, plutonium, and crude-separated strontium and promethium implies that the technetium was being stored at a 200 Area facility, although which facility was not mentioned.

Another 45-g shipment was noted in the January and February 1970 reports of the ARHCO Separations Chemistry Laboratory. This material was evidently in solution because it was expressed

as 1.7 g/L and was loaded on a resin. The origin of the material was not stated other than it came from "low-level feed material," mixed with material from "a previous run," and "three gallons from raw feed from a barrel with higher concentrations." The resin was loaded at REDOX (ARHCO 1971, pp. 29, 95).

2.4 ATTRIBUTIONS AND ANNOTATIONS

Where appropriate in this document, bracketed callouts have been inserted to indicate information, conclusions, and recommendations provided to assist in the process of worker dose reconstruction. These callouts are listed here in the Attributions and Annotations section, with information to identify the source and justification for each associated item. Conventional References, which are provided in the next section of this document, link data, quotations, and other information to documents available for review on the Project's Site Research Database (SRDB).

Donald Bihl served as the initial Subject Expert for this document. Mr. Bihl was previously employed at Hanford and his work involved management, direction or implementation of radiation protection and/or health physics program policies, procedures or practices related to atomic weapons activities at the site. This revision (and earlier revisions) have been overseen by a Document Owner who is fully responsible for the content, including all findings and conclusions. Mr. Bihl continues to serve as a Site Expert for this document because he possesses or is aware of information relevant for reconstructing radiation doses experienced by claimants who worked at the site. In all cases where such information or prior studies or writings are included or relied upon by Mr. Bihl, those materials are fully attributed to the source. Mr. Bihl's Disclosure Statement is available at www.oraucoc.org.

John (Jack) Selby served as the initial Subject Expert for this document. Mr. Selby was previously employed at Hanford and his work involved management, direction, or implementation of radiation protection and/or health physics program policies, procedures, or practices related to atomic weapons activities at the site. This revision has been overseen by a Document Owner, who is fully responsible for the content of this document, including all findings and conclusions. In all cases where such information or prior studies or writings are included or relied on by the Document Owner, those materials are fully attributed to the source.

Fred Duncan assumed responsibility as Document Owner for this document in September, 2008. Mr. Duncan replaced Edward Scalsky when Mr. Scalsky's employer declared a new corporate conflict of interest for the Hanford Site. Mr. Scalsky continues to participate on this document team in the appropriate role of Subject Expert in compliance with the NIOSH Conflict or Bias policy.

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GLOSSARY

10-66

An AEC pseudonym for thorium.

14-92

An AEC pseudonym for polonium.

37-77

An AEC pseudonym for bismuth.

B metal

Bismuth metal.

birch

An AEC pseudonym for neptunium.

brandy

An AEC pseudonym for neptunium.

brandywine

An AEC pseudonym for neptunium nitrate.

burnup

Depletion of fissionable material in nuclear fuel.

case

A code name for the unit curie used in the 1940s.

cermet

Ceramic metal alloy.

chemical 14-92

An AEC pseudonym for polonium.

chemical 37-77

An AEC pseudonym for bismuth.

cold

Not containing radioactive material.

E metal

Uranium enriched to 1.75% ²³⁵U. Also called E-1 metal, Eisenhower metal, and Ike metal.

flowsheet

A procedure for chemical analysis or separation process.

heart cut

Material obtained from the temporal center of a chromatographic column; i.e., eliminating the first and last material to separated in the column.

mint

A Hanford pseudonym for tritium.

myrnalloy

Another name for thorium.

olive

An AEC pseudonym for ²³⁸Pu.

palm

An AEC pseudonym for neptunium.

P-10

An AEC pseudonym for tritium.

Palmolive

An AEC pseudonym for the process to irradiate ²³⁷Np to produce ²³⁸Pu.

pile

Early term for a reactor. This term comes from the pile of graphite blocks forming the moderator.

pit

Package at the center of an implosion weapon that contains the machined fissile material that begins the fission chain reaction. Also called primary.

postum

An AEC pseudonym for polonium.

Q

An AEC pseudonym for thorium.

soda pulp

A pseudonym for bismuth.

slug

Uranium target in the form of short cylinders encased in corrosion-resistant metals, placed in a nuclear reactor to produce plutonium.

weapons grade

Plutonium mixture with approximately 4 to 6% ²⁴⁰Pu.

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A.1 REACTORS

External exposures at the production reactors are primarily due to the presence of activation and FPs [38]. During operation, entry was restricted or prevented in areas where high direct dose rates from photons and neutrons could occur.

Internal exposures at the production reactors relate primarily to the inhalation or absorption of activation products and, to a lesser degree, FPs during refueling and maintenance operations. Studies in later years at N Reactor to characterize worker exposure indicated the primary internal exposure in the production reactors was due to ⁶⁰Co, a corrosion product. Sula, Carbaugh, and Bihl (1989) stated, "In general, a major characteristic of these corrosion product radionuclides, regardless of origin, is the presence of several radionuclides within a matrix of oxidized metal with ⁶⁰Co the predominant radionuclide." Radionuclides in the metal oxide corrosion product mixture include ⁵⁸Co, ⁶⁰Co, ⁵⁴Mn, and ⁵⁹Fe, with ⁶⁰Co being the predominant contributor to dose and activity. These radionuclides might be present in trace amounts, but they generally are of minimal internal dosimetry significance. Tables A-1 and A-2 list the years of operation and radionuclides of concern, respectively, for power reactors.

	Reactor								
В	С	D	DR	F	Н	KE	KW	N	
9/1944-	11/1952-	12/1944-	10/1950-	2/1945-	10/1949-	2/1955-	12/1954-	12/1963-	
2/1968	4/1969	6/1967	12/1964	6/1965	4/1965	1/1971	1970	1987	

Table A-1. Production reactor years of operation [40].

Table A-2. Radionuclides of concern for all production reactors^a (Marceau et al. 2002)

2002).		
As-76	I-135	Sr-91
Ca-41	Mn-54	Sr-92
Co-58	Mn-56	U-238
Co-60	Na-24	Y-90
Cr-51	Nb-97	Y-92
Cu-64	Np-239	Y-93
Fe-59	P-32	Zn-65
Ga-72	Si-31	Zn-69
I-133		

 a. The composition and relative abundance are not known. These radionuclides represent 98% of radioactive materials in effluents. It should be noted that several of these are short-lived activation products (Si-31, As-76, etc.) that would result in only limited, if any, exposure to workers.

There were seven small research and test reactors that were operated at Hanford starting in 1944; only FFTF is still in existence [39]. External and internal exposures at these reactors were low (i.e., minimal in most cases) and unique to the specific reactor. With the exception of routine tritium exposures when working in the containment vessel at PRTR, external and internal exposures relate

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primarily to minor accidents. Tables A-3 and A-4 list the years of operation and radionuclides of concern, respectively, for research and test reactors.

Table A-3. Research and test reactors years of operation [42].

Reactor								
HTR	PCTR	TTR	PRTR	PRCF	HTLTR	FFTF		
1943–1972	1954–1970	1954–1978	1960–1969	1962–1976	1968–1971	1979–1993		

Table A-4. Radionuclides of concern for all research and test reactors (Marceau et al. 2002). Ba/La-140 Np-237 Ru-103 Ce-141 Po-210 S-35 Ce/Pm-144 Pu-238 U-233 Cf-252 Pu-239 U-234 Cm-244 Pu-240 U-235 Co-60 Ra-226 U-238 H-3 Rh/Ru-106 Zr/Nb-95

External exposure at the PRTR was minimized as a result of the remote operation and the shielding. Maintenance operations resulted in external exposure from activation and FPs. A high extremity exposure occurred when a worker picked up an irradiated steel pin. Tritium oxide was the principal internal exposure contaminant during normal operation. This was added to the whole-body dose (see ORAUT 2010a,b). The reactor was shut down in 1969 after an accident resulted in contamination in the reactor. A sample of the contamination showed the major contaminant was ⁹⁵Zr/Nb. Other contaminants included ⁶⁰Co, ¹⁰³Ru, ¹⁰⁶Ru, ¹⁰⁶Rh, ¹⁴⁰Ba/La, ¹⁴¹Ce, ¹⁴⁴Ce, and ¹⁴⁴Pr.

A.2 FUEL REPROCESSING FACILITIES

Tables A-5 and A-6 list the years of operation and radionuclides of concern, respectively, for Hanford fuel reprocessing facilities.

Separation Facility										
В	B T REDOX PUREX U UQ ₃ 225-B C Tritium									
4/1945-	12/1944-	1/1952-	1/1956-	3/1952-	1/1952-	10/1974 ^t -	1949–	8/1949-		
10/1952 ^a	present ^b	12/1967 ^c	121992 ^d	1/1958	1993 ^e	present	1967 ⁹	1955		

Table A-5. Fuel reprocessing facilities years of operation [43].

a. In 1968, B Plant started to remove Sr-90 and Cs-137 from high-level wastes.

b. The T Plant discontinued reprocessing operation in March 1956* and was used as a decontamination facility; the 2706-T Annex was added in 1959.

c. The 233-S facility at REDOX started operation in 1957 and shut down in July 1967.

d. Closed in June 1972 and reopened in November 1983. The plant was started up and shut down several times between 1983 and 1992.

e. Shut down in 1972 and opened in 1983 (there were 17 startups and shutdowns between 1984 and 1992)

f. Startup of operations involving radioactive materials (Briggs 2001, p. 32).

g. Started operations in 1952* as REDOX Pilot Plant; 1954, started operations as PUREX Pilot Plant; 1962, started operation as Pilot Plant for recovery of cesium and strontium waste tanks.

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Table A-6. Radionuclides of concern for all reprocessing facilities (Marceau et al.

2002).	
Am-241	Ru-103
Ba/La-140	Ru-106
Ce-141	Pu-239
Ce-144	Sr-90
Co-60	Th-232/228 ^b
Cs-137	U-233 ^b
H-3	U-234
I-131	U-235
Np-237 ^a	U-238

a. PUREX only 1958-72.

b. Special campaigns in PUREX only 1965, 1966, 1970.

External exposures were primarily from high-energy betas and photons in the separations facilities associated with FPs [41]. Internal exposures, primarily associated with contamination incidents, could be due to FPs or plutonium. The following mix of radionuclides – ²³⁸PU, ²³⁹PU, ²⁴⁰Pu, and ²⁴¹Pu – comprises most plutonium mixtures at Hanford. The Hanford plutonium mixtures in Tables A-7, A-8, and A-9 were categorized by their weight percent of ²⁴⁰Pu (PNNL 2003). When the production reactors were operating, the target mixture was ~ 6 wt% ²⁴⁰Pu (i.e., weapons grade).

Mixture designation:	Fresh	5-yr	10-yr	15-yr	20-yr	25-yr	30-yr
Years of aging ^a :	0	5	10	15	20	25	30
Specific activity in							
mixture (Ci/g)							
Pu-238	8.56E-03	8.23E-03	7.91E-03	7.60E-03	7.31E-03	7.03E-03	6.75E-03
Pu-239	5.77E-02						
Pu-240	1.36E-02						
Pu-241	8.24E-01	6.48E-01	5.09E-01	4.00E-01	3.15E-01	2.48E-01	1.95E-01
Pu-242	1.97E-06						
Am-241	0	5.83E-03	1.04E-02	1.39E-02	1.66E-02	1.87E-02	2.03E-02
Pu-239+240	7.13E-02	7.13E-02	7.13E-02	7.13E-02	7.12E-02	7.12E-02	7.12E-02
Pu-alpha	7.99E-02	7.95E-02	7.92E-02	7.89E-02	7.85E-02	7.83E-02	7.80E-02
Total alpha	7.99E-02	8.53E-02	8.96E-02	9.28E-02	9.52E-02	9.70E-02	9.83E-02
Activity ratios							
Pu-239+240: Am-241	NA	12.2	6.87	5.13	4.28	3.80	3.50
Pu-239+240: Pu-238	8.33	8.67	9.01	9.38	9.74	10.1	10.5
Pu-241: Pu-239+240	11.6	9.09	7.15	5.62	4.42	3.48	2.73
Pu alpha:Pu-239+240	1.12	1.20	1.26	1.30	1.34	1.36	1.38
Pu alpha: Pu-238	9.33	9.66	10.0	10.4	10.7	11.1	11.6
Pu alpha: Am-241	NA	14.6	8.63	6.67	5.72	5.18	4.84
Pu-241: Pu alpha	10.3	8.15	6.43	5.07	4.01	3.17	2.50

Table A-7. Activity composition of Hanford reference weapons-grade plutonium mixture (PNNL 2003).

a. Time since separation of Am-241 from the plutonium mix.

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Table A-8. Activity com Mixture designation:	Fresh	5-year	10-year	15-year	20-year	25-year	30-year
Years of aging ^a :	0	5	10	15	20	25	30
Specific activity in							
mixture (Ci/g)							
Pu-238	1.71E-02	1.64E-02	1.58E-02	1.52E-02	1.46E-02	1.40E-02	1.35E-02
Pu-239	5.26E-02	5.26E-02	5.26E-02	5.26E-02	5.26E-02	5.26E-02	5.25E-02
Pu-240	2.72E-02	2.72E-02	2.72E-02	2.72E-02	2.72E-02	2.71E-02	2.71E-02
Pu-241	3.09E+00	2.43E+00	1.91E+00	1.50E+00	1.18E+00	9.29E-01	7.30E-01
Pu-242	3.93E-06						
Am-241	0	2.19E-02	3.89E-02	5.22E-02	6.24E-02	7.03E-02	7.63E-02
Pu-239+240	7.98E-02	7.98E-02	7.98E-02	7.97E-02	7.97E-02	7.97E-02	7.97E-02
Pu-alpha	9.69E-02	9.62E-02	9.56E-02	9.49E-02	9.43E-02	9.37E-02	9.32E-02
Total alpha	9.69E-02	1.18E-01	1.35E-01	1.47E-01	1.57E-01	1.64E-01	1.69E-01
Activity ratios							
Pu-239+240: Am-241	NA	3.64	2.05	1.53	1.28	1.13	1.04
Pu-239+240: Pu-238	4.67	4.86	5.05	5.24	5.46	5.69	5.90
Pu-241: Pu-239+240	3.87	3.05	2.40	1.88	1.48	1.17	9.16
Pu alpha:Pu-239+240	1.21	1.21	1.20	1.19	1.18	1.18	1.17
Pu alpha: Pu-238	5.67	5.87	6.05	6.24	6.46	6.69	6.90
Pu alpha: Am-241	NA	4.39	2.46	1.82	1.51	1.33	1.22
Pu-241: Pu alpha	31.9	25.3	20.0	15.8	12.5	9.91	7.83

Table A-8. Activity composition of Hanford reference fuel-grade plutonium mixture (PNNL 2003).

a. Time since separation of the Am-241 from the plutonium mix.

Table A-9. Activity composition of Hanford reference commercial power fuel-grade plutonium mixture (PNNL 2003).

(1 ININE 2003).							
Mixture designation:	Fresh	5-year	10-year	15-year	20-year	25-year	30-year
Years of aging ^a :	0	5	10	15	20	25	30
Specific activity in							
mixture (Ci/g)							
Pu-238	1.71E-01	1.64E-01	1.58E-01	1.52E-01	1.46E-01	1.40E-01	1.35E-01
Pu-239	3.41E-02						
Pu-240	5.90E-02	5.89E-02	5.89E-02	5.89E-02	5.89E-02	5.88E-02	5.88E-02
Pu-241	1.34E+01	1.05E+01	8.28E+00	6.51E+00	5.12E+00	4.03E+00	3.17E+00
Pu-242	1.97E-04						
Am-241	0	9.49E-02	1.69E-01	2.26E-01	2.79E-01	3.04E-01	3.31E-01
Pu-239+240	9.31E-02	9.31E-02	9.30E-02	9.30E-02	9.29E-02	9.29E-02	9.29E-02
Pu-alpha	2.65E-01	2.58E-01	2.52E-01	2.45E-01	2.39E-01	2.34E-01	2.28E-01
Total alpha	2.65E-01	3.53E-01	4.20E-01	4.71E-01	5.10E-01	5.38E-01	5.59E-01
Activity Ratios							
Pu-239+240: Am-241	NA	0.981	0.551	0.411	0.344	0.305	0.281
Pu-239+240: Pu-238	0.544	0.568	0.589	0.612	0.636	0.664	0.688
Pu-241: Pu-239+240	144	113	89.1	70.0	55.1	43.3	34.1
Pu alpha:Pu-239+240	2.85	2.77	2.71	2.63	2.57	2.52	2.45
Pu alpha: Pu-238	1.55	1.57	1.59	1.61	1.64	1.67	1.69
Pu alpha: Am-241	NA	2.72	1.49	1.08	0.857	0.770	0.689
Pu-241: Pu alpha	50.6	40.7	32.9	26.6	21.4	17.2	13.9

a. Time since separation of the Am-241 from the plutonium mix.

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A special study in the 1980s indicated frequent, intermittent releases of plutonium in the workplaces at levels below detectability for normal air sampling. This resulted in <10 mrem committed effective dose equivalent (Bihl 1993; Lyon et al. 1988, 1989).

The new lung model for internal dosimetry uses absorption types. The new types are F, M, and S for fast, moderate, and slow, respectively. A highly insoluble form, Super S, should also be considered (ORAUT 2008). Plutonium in nitrate form is type M, which would apply to the separations plants up to the point when N Cell began operations at PUREX. Plutonium oxide is classed as type S. This would apply to material calcined at N Cell at PUREX. Type S at PUREX would apply only to N Cell, not the PR Room. Contamination remaining in old gloveboxes, hood exhaust ducts, high-efficiency particulate air (HEPA) and sand filters, housings, etc., probably would be classed between types M and S. This is because plutonium slowly oxidizes. Plutonium at 233-S was type M until 1967 when a fire resulted in the facility shutdown. The plutonium would have been type S by the time D&D started at 233-S in the 1980s. It should be assumed that after 1990 all plutonium at Hanford would be type S or Super S because of natural oxidation. In general, plutonium absorption types M, S, and highly insoluble Super S should be considered for dose reconstruction (ORAUT 2008). Note: The default solubility classes should be used only when bioassay data are inconclusive.

The B Plant facilities were modified in 1968 to remove ⁹⁰Sr and ¹³⁷Cs from high-level wastes from the single- and double-shell waste tanks. The purified ⁹⁰Sr and ¹³⁷Cs solutions were transferred to the 225-B Waste Encapsulation Storage Facility (see Section 2.2.2.6). Internal exposures from the operation were minimal due to the remote operation. As a result of remote operation, external exposures to high-energy photons and beta particles that resulted from the processing of large quantities of strontium and cesium were minimized. The 108-B Facility was used for the extraction of ³H for several years. Internal exposure was related to ³H, which was added to whole-body exposure (see ORAUT 2010a,b).

Internal exposures at the U and UO₃ Plants were due to the large quantities of uranium that were processed. The contaminant could be found ranging from soluble uranyl nitrate and UO₃ to insoluble UO₂ and U₃O₈.

The inhalation class for Hanford uranium compounds, based on International Commission for Radiological Protection (ICRP) Publication 66 (ICRP 1994), is listed in Table A-14 (see Section A.3).

A.3 FUEL FABRICATION FACILITIES

External exposures related to the beta and photon radiation that is associated with uranium. Due to the pyrophoric nature of uranium turnings and chips, internal exposure related primarily to inhalation of uranium. A pressurization of one of the sealed metal storage cans resulted in the spread of plutonium contamination and the subsequent shutdown of the building until cleanup. The years of operation and radionuclides of concern are listed in Tables A-10 and A-11, respectively. The radiological characteristics of Hanford uranium mixtures are listed in Table A-12, and the impurities in recycled Hanford uranium are listed in Table A-13. The inhalation class for Hanford uranium compounds is listed in Table A-14. Note: The default solubility classes should be used only when bioassay data are inconclusive [44].

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Table A-10. Fuel fabrication facilities years of operation [45].

	Fuel fabrication							
313	314	333	303 Facilities	306				
3/1944–1/1971	7/1944–1971	1961–1988	1944-present	5/1957-2004*				

Table A-11. Radionuclides of concern for all fuel fabrication and storage facilities [46].Am-241Np-237Pu-238Pu-239Th-232U-235

Table A-12. Radiological characteristics of Hanford uranium mixtures (PNNL 2003).

U-238

	Uranium mixture										
Weight percentage ^{a,b}	Natural (NU)	Depleted (DU)	Recycled (RU)	Commercial fuel (CF)							
U-234	0.0057	0.0005	0.0082	0.0300							
U-235	0.7204	0.2500	0.9700	2.9600							
U-236	Negligible	Negligible	0.0680	Negligible							
U-238	99.2739	99.7500	98.9500	97.0100							
Specific constitue	ent activity in m	ixture (µCi/g, nC	i/mg, or pCi/µg	0							
U-234	0.3563	0.0313	0.5125	1.8750							
U-235	0.0156	0.0054	0.0210	0.0639							
U-236	Negligible	Negligible	0.0440	Negligible							
U-238	0.3336	0.3352	0.3325	0.3260							
Total	0.7054	0.3718	0.9099	2.2649							
Specific constitue	ent activity in m	ixture (dpm/µg) ^c									
U-234	0.7909	0.0694	1.1378	4.1625							
U-235	0.0345	0.0120	0.0465	0.1419							
U-236	Negligible	Negligible	0.0977	Negligible							
U-238	0.7405	0.7441	0.7381	0.7236							
Total	1.5659	0.8254	2.0200	5.0281							
Constituent fracti	on of total uran	ium activity in m	nixture								
U-234	0.5051	0.0840	0.5632	0.8279							
U-235	0.0221	0.0145	0.0230	0.0282							
U-236	Negligible	Negligible	0.0484	Negligible							
U-238	0.4729	0.9014	0.3654	0.1439							
Total	1.0000	1.0000	1.0000	1.0000							

a. NU, DU, and CF data from Rich et al. (1988).

b. RU data based on average of data presented by Sula, Carbaugh, and Bihl (1991).

c. Can be used to represent specific alpha activity in the mixture as well.

A.4 PLUTONIUM FINISHING FACILITIES

Although relatively few Hanford personnel were involved in plutonium finishing, these facilities provided the majority of personnel external exposures to neutron radiation. As production increased, 10-in.-thick water-filled walls were installed to reduce worker exposure to neutrons. Special lead-filled gloves were installed to reduce extremity exposure. Worker external exposure in the vault rooms was due to low-energy photons and neutrons that occurred during periodic inventory and inspection activities [47].

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Table A-13. Impurities in recycled uranium at Hanford (PNNL 2003).

Constituent	Maximum allowed ^a	Observed range ^b	Reference level ^c
Plutonium	10 ppb U	<1–2 ppb U	0.4 nCi Pu-alpha/g U
Neptunium	Not established	0.04-0.16 ppm U	0.4 nCi Np-237/g U
Thorium	750 ppm U	8–10 ppm U	5 pCi Th-232/g U
Tc-99	Not established	3–4 ppm U	0.2 μCi Tc-99/g U
Ru-103, -106	<20 µCi/lb U	<6 µCi/lb U	40 nCi Ru-106/g U
Zr/Nb-95	<10 µCi/lb U	<4 µCi/lb U	20 nCi ZrNb-95/g U
Other gamma emitters	<2 µCi/lb U	0.09 0.75 µCi/lb U	Negligible
excluding Tc-99			

a. From UO₃ Plant operating specifications (Sula, Carbaugh, and Bihl 1991).

b. From analysis of uranium lots 88-1, 88-2, 88-3 that were processed in 1988, and lots 93-01, 93-02, 93-03, 93-04, and 93-05, processed in 1993.

c. A reference level is chosen for determining bioassay monitoring needs and for use as an initial assumption in evaluation of intakes. The use of the reference levels is expected to result in a slight overestimate of dose compared to levels actually observed in 1988.

ICRP Publication 30 ^a inhalation class from lung fluid studies	Compound and location	Recommended ICRP Publication 66 ^b lung absorption type ^c
80% D	Hanford UO ₃ Plant smear sample dissolution study in	F
20% W	1984, ^d (UO ₃ powder)	
10% D	Hanford 303-M Building air sample dissolution study ^e (300	S
90% Y	Area Uranium Fuel Production Facilities)	
29% D	Hanford 333 Building air sample dissolution study ^e (300	S
71% Y	Area Uranium Fuel Production Facilities)	
20% D	Hanford 306-W Building Machine Shop air sample	S
80% Y	dissolution study ^e	
	Uranyl nitrate at PUREX or UO ₃ Plant	F
	UCl ₄ or U carbonate (assumed form after discharge to the	M ^t
	soil)	

Table A-14. Inhalation class for Hanford uranium compounds (PNNL 2003).

a. ICRP (1979).

b. ICRP (1994).

c. The default solubility classes should be used only when bioassay data is inconclusive.

d. Sula, Bihl, and Carbaugh (1989).

e. Fisher (1986).

f. Cooke and Holt (1974).

Internal exposures in the operating areas related primarily to contamination incidents that involved americium and plutonium because airflow patterns were designed to control the flow from areas of lower air contamination potential to higher. Maintenance and cleanup operations were conducted in protective clothing that included respiratory protection. Exposures occurred by inhalation, absorption, and injection (i.e., wound sites). Internal exposure in the vault areas was minimal because there were no routine activities that involved opening the storage containers. Internal exposure would have been a concern in the event of an incident that breached the containment [48].

In February 1960, RECUPLEX was temporarily shut down to permit an extensive cleanup of hoods and operating equipment. A criticality accident in the RECUPLEX facility in April 1962 resulted in high external radiation exposures and in widespread contamination throughout the RECUPLEX room [49].

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Plutonium in nitrate form is absorption type M, which would apply to the plutonium finishing plants where plutonium nitrate has been involved (RECUPLEX, 231-Z, etc.). Plutonium oxide that is classed as type S would apply to material that was calcined at the RMC Line at PFP. Contamination remaining in old gloveboxes, hood exhaust ducts, HEPA and sand filters, and housings, etc., probably would be classed between types M and S, but the default would be S. This is because plutonium slowly oxidizes. It should be assumed that after 1990 all plutonium at Hanford would be class S or Super S because of natural oxidation. In a study in the early 1990s of the isotopic distribution of plutonium at PFP, it was stated there were approximately 3.7 million g of plutonium with the ²⁴⁰Pu content ranging from 4% to >19% (Crowe and Szempruch 1994). In general, plutonium absorption types M, S, and highly insoluble Super S should be considered for dose reconstruction (ORAUT 2008). Note: The default solubility classes should be used only when bioassay data is inclusive.

The years of operation and the radionuclides of concern are listed in Tables A-15 and A-16. The activity composition for Hanford reference plutonium mixtures is listed in Tables A-7, A-8, and A-9.

	Facility/process							
	234-5Z 234-5Z 234-5Z 234-5Z 234-5Z							
231-Z	232-Z	RG	RMA	RMC	RECUPLEX	Storage vault	236-Z	242-Z
1/1945-	1/1962-	7/1949–	3/1952-	10/1960-	7/1955–	1949–	5/1964-	1963–
2/1977 ^a	1973	1957	1984 ^b	5/1989 ^c	5/1962	present	8/1976 ^d	6/1977* ^e

Table A-15.	Plutonium	finishing	facilities	vears of	operation	[50]	

a. In 1957, the mission changed to plutonium metallurgy development. From February 1975 to February 1977 the principal mission was cleanup and stabilization (Lini 2009, p. 1)

b. In 1964, line shutdown; in 1967, a glovebox was reactivated; in 1968, Tasks I-III were cleaned out and reactivated; final cessation of operations occurred in 1979; in 1984, it was decided to keep the RMA Line on standby but it never operated again.

c. In 1962, 10-in.-thick water-filled shielding tanks were added to substantially reduce operator exposure to neutrons; several safety improvements were made in 1963 and 1964; fabrication of pits was removed from line in 1966. Shut down in 1976 as a result of explosion in 242-Z; restarted in 1985; shut down in 1989; restarted briefly in 1997 to process material from cleanout of 242-Z;

d. In December 1975, it was shut down for upgrades; restarted in April 1976; shut down in August 1976 as a result of explosion in 242-Z.

e. In 1969, changed from batch to continuous process; in April 1976, shut down because of strike; restarted in April 1976; shut down in August 1976 as a result of explosion in americium recovery area. It was restarted briefly in early 1977 to process material left over from the shutdown the year before.

Tab	ole A-16.	Radionuclide	es of concern	for plutoniun	n finishing fac	cilities [51].		
	U-233	Np-237	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241

A.5 RESEARCH AND DEVELOPMENT FACILITIES

The 21 major research, development, and testing facilities at Hanford were in the 100, 200, and 300 Areas. Construction of these started in 1944, and the last was completed in 1977. These facilities were built to support the development and testing of new processes, materials, and equipment. In some instances the facilities were used for several different purposes throughout their operating lives. Therefore, the potential exposures are dependent on the missions and processes at any given time. See Section 2.2.5 for discussion of internal and external exposure potential at specific facilities. The years of operation are listed in Tables A-17 and A-18; the radionuclides of concern are listed in Table A-19.

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Table A-17. 100 and 200 Area R&D facilities years of operation [52].

Facility/process					
108-F	108-F 120 209-E 222-B 222-S 222-T				
1949–1977	4/1950–11/1951	7/1961–1986* ^a	1945–1975	1949-present	1944–1986 ^b

Only decade date found. a.

More than one date found. b.

Table A-18. 300 Area R&D facilities years of operation [53].

	Facility/process						
308	308 318 320 321 324 325 326						
1960–1990* ^a	1960–1990* ^a 1983–present ^b 1966–present 1944–1988 ^c 1966–present 1953–present ^d 1953–present						

	Facility/process						
327	329	331	3706	3730	3732	3741	3745
7/1953–	1952-present	1972-present	1945–	1949–1981	1949–1970	1944–1956	10/1944-
1987			late1960s ^e				1983 [†]

a. In the mid-1960s, PRTR fuel work was terminated; neptunium-aluminum fuel elements produced in late 1960s; high bay area added in 1971; from 1977 to March 1990, FFTF fuel elements were produced; the 308A annex was added in 1979; TRIGA reactor was installed in the late 1970s.

b. The HTLTR commenced operation in the 318 Building in 1967; the reactor was shut down in 1971; the reactor was removed in the early 1980s; there were major additions in 1982, 1987, and 1989; the calibration facility commenced operation in 1983.

The 321 Separation Building started in 1944; in late 1944, work with small amounts of irradiated materials (<1 Ci) to c. conduct separations experiments and to study decontamination factors; in 1949, a cold test of REDOX process was performed; a cold test of the PUREX process occurred in 1953.

d. The 325-A hot cell wing was added in 1959*; routine manufacture of Pm-147 heat sources began in 1966; Po-210 work started in June 1972 and ended in 1975* for use with the demonstration of the Waste Vitrification Process for commercial wastes.

e. Most of the radiation laboratories were removed in 1954; building converted to offices in the 1970s and still in use in the early 1990s.

The 3745-B annex that housed a 4-MV Positive Ion Accelerator started operation in 1955; the 3745-A annex that f. housed a 2-MV Negative Ion Accelerator started operation in 1950*; the calibrations operations were transferred to the 318 Building in 1983; the building is being used as offices.

[54].	
Am-241	Pu-240
C-14	Ru-103
Ce-144	Ru-106
Co-60	S-35
Cm-244	Sr-90
Cs-137	Th-232/228
Cu-64	U-233
I-131	U-234
Np-237	U-235
Pm-147	U-238
Po-210	Y-90
Pu-238	Zr/Nb-95
Pu-239	

Table A-19. Radionuclides of concern for all 100, 200, and 300 Area R&D facilities

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A.6 WASTE FACILITIES

The years of operation and radionuclides of concern for Hanford Site waste facilities are listed in Tables A-20 and A-21, respectively.

Table A-20.	Waste facilities	vears of ope	ration [55].
		youro or opo	10001

Facility/process					
	Z-9 Trench		242 Evaporator	Exhaust Filtration	340 Waste
Waste tanks	Facility	241-Z	Facilities	Facilities	Complex
1944-present	1955–7/1978	1949–1973	1951-present	1947–19UNK	1954-present

Table A-21. Radionuclides of concern for all waste facilities [56].

Am-241	Pu-238
Ce-141	Pu-239
Ce-144	Pu-240
Co-60	Ru-103
Cs-137	Ru-106
I-131	Sr/Y-90
Pm-147	Th-232/228

External exposure related to high-energy beta and photon radiations from the FPs in the tanks. Under normal conditions, shielding, distance, and time helped keep personnel external exposures at a low level. Leaks of high-level wastes that could result in unusual or unplanned external exposures were documented in incident or accident reports. In 1994, data were included in Boomer et al. (1994) on tank waste radionuclide inventory. The data from that report are included in Table A-22. Internal exposures to FPs were controlled through the use of respiratory protection.

Mega curies of	Liquid components		Solid components	
radioactive specie	Double-shell tanks	Single-shell tanks	Double-shell tanks	Single-shell tanks
Cs-137 and Ba-137	6.40 E+01	2.53 E+01	2.33 E+00	8.45 E+00
Sr-90 and Y-90	3.93 E+00	1.14 E+01	2.17 E+01	1.13 E+02
Tc-99	2.28 E-02	7.82 E-03	1.35 E-03	7.82 E-03
Am-241	1.45 E-02	4.70 E-03	7.97 E-02	4.23 E-02
Np-237	1.64 E-05	3.30 E-06	1.92 E-05	2.97 E-05
Pu	3.84 E-03	5.50 E-04	6.80 E-03	2.69 E-02
Transuranic elements	1.93 E-02	5.25 E-03	9.10 E-02	6.92 E-02

Table A-22. Tank waste radionuclide inventory [57].

A mining operation to remove an estimated 45 kg of plutonium from the Z-9 trench was initiated in August 1976 and was completed in July 1978. The principal internal exposure potential related to ²³⁹Pu contamination, most of which occurred in the soil packaging glovebox. Because the waste site was used only for plutonium, the external exposure potential during the mining operation was minimal.