

Review of Generation and Flow of Recycled Uranium at Hanford



Richland Operations Office

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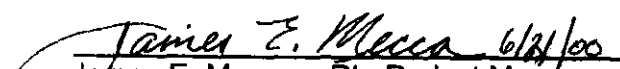
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EXECUTIVE SUMMARY

Hanford Recycled Uranium Project

On August 8, 1999, Energy Secretary Richardson announced a set of activities to address the extent of potential exposure to recycled uranium and its constituents, and the quantities of uranium involved at Department of Energy (DOE) operated sites. The DOE-HQ Recycled Uranium Project was to gather the pertinent information and provide a report of its findings to the Secretary by June 2000. On September 15, 1999, selected field offices were directed to support this Recycled Uranium Project.

The Richland Operations Office (RL) assembled a multi-contractor team of senior staff in response to the Headquarters directive to gather and consolidate the requested supporting information and to provide this information in a formal report in support of the June 2000 date. Based on the direction contained in the Secretary's letter, Hanford facilities and activities involving the production, treatment, and handling of recycled uranium and the trace impurities of plutonium (Pu), neptunium (^{237}Np), and technetium (^{99}Tc) isotopes were investigated.

Background

"Hanford Engineering Works" was established in secrecy during World War II to produce plutonium for an atomic bomb in support of the ongoing defense effort. Hanford produced the Pu used for the first atomic explosion test in New Mexico in July 1945. Since the first Pu production, Hanford continued to grow and support defense and other missions as directed. During its operating period, 1943 through 1993, Hanford built and operated 9 production reactors, five separations plants, several reactor fuel manufacturing facilities, a uranium trioxide (UO_3) production facility and several Pu processing facilities. Additional facilities were built to support the production of plutonium, recovery of uranium, waste treatment, and provide site infrastructure. Plutonium was Hanford's primary product and recovered uranium was a secondary product. Peak nuclear materials production was reached in the 1960s, when all nine production reactors were in operation. Altogether, Hanford supplied Pu for the United States nuclear weapons program for more than four decades. Weapons material production was halted in the late 1980s.

The Hanford Site presently consists of ~1,450 square kilometers (~560-square-miles), located just north of the city of Richland, in the south central part of Washington State. The Hanford Site is managed by the DOE Richland Operations and River Protection Offices.

Hanford's Role in Recycled Uranium

During Hanford's early years of operation, irradiated fuel was processed in T-Plant and B-Plant to recover Pu. Uranium from the irradiated fuel remained in the high level waste, which was sent to large underground storage tanks. In early 1952, operation of the Reduction-Oxidation Plant (REDOX) was initiated to process irradiated fuel,

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uranium went to the Paducah Gaseous Diffusion Plant (GDP) in KY, ~4,404 MT of recycled uranium went to the K-25 GDP and the Y-12 Plant at Oak Ridge, TN, and ~25,251 MT of recycled uranium was shipped to Fernald, OH. An additional ~5,646 MT of recycled uranium was sent to over 100 other destinations for research and miscellaneous non-defense and defense needs.

- Approximately ~4,006 MT of uranium remains at Hanford, excluding uranium in tanks and solid waste and uranium which was discharged to the cribs. This uranium is in various forms: metal received for making fuel, unused fuel, irradiated fuel (not in the scope of this study), uranium as oxide in mixed-oxide fuel (at the Fast Flux Test Facility and in vaults at the Plutonium Finishing Plant) and recovered uranium as UO_3 for recycle.
- In the ~50 years (1943 through the mid-1990s) Hanford was in operation, ~2,174 MTU waste was generated. (Approximately 958 MTU now reside in the waste tanks, ~1,054 MTU is in solid waste, and ~162 MTU of liquids present on the Hanford site were discharged to cribs.)
- With the exception of a few years in the 1940s, the metal turnings and scrap produced during fuel rod manufacture were shipped offsite for recovery. Most of this scrap metal was converted to uranium oxide before shipment. It has been estimated that, overall, ~ 10 percent (~11,927 MTU) of the uranium received might have gone to scrap during fuel fabrication and was shipped offsite for recovery. Shipping records for the ~112,287 MTU shipped do not show the distinction between the shipment of scrap and the shipment of recovered uranium as UO_3 but most of this ~11,927 MTU would have been recycled uranium. No Hanford process has been identified which would have changed the as-received ratio of Pu to uranium, or concentrated the constituents in the scrap prior to shipment.
- An estimated ~140 MTU was consumed during reactor operations and the generation of plutonium.
- A material difference of ~0.56 wt% (664.1 MTU) is indicated in the data reviewed between uranium received and uranium which was used up in the reactors, shipped, and uranium currently on site. The uranium currently on site includes the stored inventory of unirradiated uranium, uranium in irradiated fuel, uranium in tank waste and solid waste, and uranium in wastes in the cribs. This difference can be primarily attributed to limited available data from the early years of Hanford's operations, uncertainties in the quantities of uranium in tank waste and other waste forms, and uncertainties in the estimated amount of uranium that was consumed during reactor operation and the generation of Pu.
- Hanford first began receiving recycled uranium metal billets for reactor fuel rod manufacture starting about July 1952. The recycled uranium used to produce these billets had been processed through the GDPs and was reported to contain

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approximately 10 parts Pu per trillion parts uranium. After about 1963, as a result of a manufacturing change at Fernald, the metal billet Pu concentration rose to a level "not to exceed 10 parts plutonium per billion parts of uranium." Hanford did not routinely perform a Pu analysis of the incoming billets.

Information provided by Fernald indicates that the metal produced from recycled uranium contained ^{99}Tc ranging from < 0.01 to 6.1 ppm and ^{237}Np with an upper limit of ~500 ppb.

The prospect of an ingestion or inhalation dose in dealing with this metal is remote.

- In 1951, a tentative Pu specification for UO_3 was established at 100 parts per billion (ppb), based upon the expected composition of a blended UNH product mix from U-Plant and REDOX Plants. Preliminary specifications were identified by Oak Ridge in 1952 which required the Pu specification for UO_3 to be lowered to 10 ppb. This limit was subsequently firmed up in 1953 and remained in place from that time until UO_3 production was terminated in 1993.
- Shipments of recycled UO_3 powder from Hanford to the GDPs contained only trace amounts of Pu, usually less than 10 ppb. **The preponderance of Hanford recycle UO_3 powder shipments had Pu concentrations of 5 or less ppb in the recycled uranium.** Six shipments of UO_3 have been identified which contained Pu concentrations of 12, 13, 16, 19, 22, & 30 ppb. These shipments involved ~193 MTU containing an average plutonium impurity level of ~18 ppb (~3.4 g Pu). Documentation reviewed indicates that the receiver sites were typically notified of these conditions and accepted these limited quantities of recycled uranium prior to their shipment from Hanford.
- Analytical results on every lot of UO_3 powder shipped to the GDPs have not been located. Thus a quantitative assessment of the total Pu shipped with the recycled uranium was not possible. Based upon the findings that the predominant Pu concentration was in the range of 1 to 5 ppb, it is estimated that approximately 110 to 550 grams of Pu (with a mean of 330 grams) was included with the ~109,792 MTU shipped from Hanford.
- Hanford did not routinely analyze the UO_3 product produced before 1980 for ^{237}Np or ^{99}Tc , as it was not a specification requirement. Hanford did analyze the recycled UO_3 powder for total beta and gamma emissions and conformed to the required specification levels of less than a 100% increase in beta activity and less than a 300% increase in gamma activity above that of aged natural uranium. In some later campaigns where analyses were performed, the measured ^{237}Np concentrations typically ranged from 20 to 500 ppb, and ^{99}Tc concentrations ranged from 3-12 ppm. A rough estimate, based on limited analytical data, indicates that the ~109,792 MT of recycled uranium shipped offsite might have contained 2 to 55 Kg Np and 330 to 1,320 Kg Tc.

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The operation, maintenance, and waste handling operations associated with all of these facilities contributed to some personnel exposure and environmental releases. However, distinguishing exposures to trace quantities of transuranics and fission products in recycled uranium from those associated with normal Hanford operations (which involved the handling and processing of significant quantities of irradiated fuel, high-level waste, and Pu) is considered extremely difficult. Any such dose assessment is further complicated by the past practice of transferring personnel between facilities to meet work needs, necessitating the development of a historical worker profile in order to establish which portions of the dose can be attributed to the various facilities and process operations.

Plutonium was Hanford's principal product and all production processes were designed and operated to maximize plutonium recovery. The primary Hanford facilities which had the highest potential for uranium uptake by personnel were the UO_3 Plant, which handled large quantities of dry UO_3 powder, and the fuel fabrication facilities in the 300 Area of Hanford which handled uranium metal and uranium fabrication scrap.

An Occupational Potential Exposure (OPE) estimate suggests that ~1,128 staff at the Hanford site worked in areas directly involving the handling and processing of recycled uranium. Of these personnel, it is estimated that ~456 could have had moderate OPE (some potential or incidental exposure to recycled uranium but at levels not expected to have any measurable health effect). This is only a rough estimate. For a more thorough analysis of worker contact with recycled uranium, including an examination of Hanford's uranium bioassay records, a more detailed review would be required.

Section 1

Hanford Uranium Mass Balance Project

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Section 1.0 Hanford Uranium Mass Balance Project

1.1 Project Overview

On August 8, 1999, the Secretary of Energy announced a comprehensive set of actions to address issues at the Department of Energy's (DOE) Paducah Gaseous Diffusion Plant in Paducah, Kentucky, that may have the potential to affect the health of the workers. One of the issues with DOE complex-wide significance involved the need to determine whether radioactive fission products and plutonium in uranium feed and waste streams existed in concentrations that present potential health or environmental concerns. The fission products and transuranic contaminants are contained in uranium that has been irradiated in a nuclear reactor and then processed in a separations plant to recover the uranium for reuse.

A working group was established to address this issue from a DOE complex-wide perspective. The Hanford Uranium Mass Balance Project was established to provide Hanford-specific data to the working group for inclusion in a complex-wide report. The Project Team was established, staffed with present and former Hanford workers, many with direct experience related to the facilities, processes and materials involved in the recycling of uranium or with the identification and retrieval of historical data at Hanford.

Between October 1999 and April 2000, a systematic search of available historical data was conducted and technical members of the Project Team evaluated relevant information. This report represents the results and conclusions of their evaluation.

1.2 Purpose and Scope

The purpose of this Project was to develop and provide data to the DOE Working Group that is responsive to a letter from the Deputy Secretary of Energy [Glauthier 1999] to all DOE elements dated September 15, 1999, and two follow-up memorandums from EH-1 [Michaels 1999]. Specifically, the data was to be sufficient to permit the working group to:

- Identify the mass flow of DOE recycled uranium from early production to March 1999, including ultimate use and disposition, and create an inter-site flow sheet for public availability.
- Identify the characteristics and contaminants in the major uranium streams, specifically, the technetium, neptunium, plutonium or other isotopic content of concern to worker or public health and safety.
- Conduct site mass balance activities sufficiently thorough to identify any significant implications for personnel exposure or environmental contamination.

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- Conduct an occupational radiation exposure profile project at the Paducah, Portsmouth, and former Oak Ridge K-25 facilities.

The Hanford Uranium Mass Balance Project included an evaluation of shipments, receipts, and current inventories of recycled uranium to quantitatively estimate the flow and characteristics of recycled uranium to and from Hanford. It also included an evaluation of all Hanford facilities involved in processing recycled uranium, particularly with respect to the potential for concentrating plutonium or radioactive contaminants in the recycled uranium.

The uranium materials within the scope of this project are:

- All uranium (depleted, natural, or enriched, and in all physical and chemical forms) that has ever been irradiated in a reactor and separated from all or some of the fission products and transuranic products.

The uranium materials considered out-of-the-scope of this project are:

- Irradiated fuel and targets which have not been processed to separate all or some of the fission products and transuranic products from the uranium.
- Uranium in waste or under environmental management.
- Uranium in sources, samples, or under NRC agreement or State controls.

This information is intended to enable DOE to assess the historical potential for worker exposure from recycled uranium and its trace transuranic and fission product impurities.

1.3 Project Implementation Strategy

DOE Richland Operations Office (RL) management determined that the Project Team would be staffed with present and former Hanford chemists, engineers and other workers with knowledge of the Hanford facilities, processes and materials related to the recovery and reuse of recycled uranium. The Team also included personnel with experience in DOE nuclear material control and accountability, and document classification and declassification requirements and processes.

Hanford currently has approximately 75,000 boxes of historical records in archives, both on site and off. A discussion of the document research process used on this study is provided in Appendix E. The Project organization and functional activities are depicted in Figure 1-1.

In addition to the database searches, the RL Project Team made extensive use of the first hand knowledge of Team members to identify and search out specific

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- Radioactive contaminants in the uranium trioxide shipped from Hanford for which no radiochemical data has been found are assumed to be in the same range as those for which analytical data was located.

References

Glauthier 1999

Memorandum, T. J. Glauthier, Deputy Secretary of Energy, to All Department Elements, *Paducah Gaseous Diffusion Plant -- Follow-Up Activities*, U. S. Department of Energy, Washington, DC, September 15, 1999

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2.0 Hanford Site Historical Overview

2.1 Site Description

The Hanford site was established by the Manhattan Project as the plutonium production site for the U. S. World War II defense effort. The site initially consisted of ~1,735 square kilometers (~670 square miles) of primarily remote, semiarid land. Over time, this has been reduced to ~1,450 square kilometers (~560 square miles) as parcels of land no longer needed by the site were made available for other uses. The site has access to significant water and power supplies and nearby barge shipping, road, air, and rail facilities. Construction was initiated in March 1943 to build facilities to produce plutonium for the first atomic bombs. The principal early facilities included B-, D-, and F-Reactors, the 313 Fuel Fabrication Facility, the T-Plant and B-Plant fuel processing facilities for plutonium separation, and the 231 Isolation Process facility for final plutonium purification and concentration. Waste storage tanks, laboratory facilities, warehousing, roads, power plants, housing, and a myriad of other support facilities were simultaneously built during the construction period. Initial plutonium production commenced in September 1944 when the first reactor was brought on line. The first batch of refined plutonium was available for the war effort in February 1945.

Buildup of the Hanford site continued for many years in support of nuclear weapons program needs. Additional facilities were eventually needed to satisfy planned plutonium production requirements and to improve process efficiencies. An additional six reactors and four processing facilities were constructed and operated, including: the Reduction Oxidation (REDOX) Plant, U-Plant, UO₃ Plant, and the Plutonium-Uranium Extraction (PUREX) Plant. During the plutonium production years at Hanford, uranium was received at the site, fabricated into nuclear fuel, irradiated for plutonium production, and processed to recover the plutonium. Until about 1952, the waste from plutonium separation, which included fission products and unused uranium, was stored in large, underground waste tanks. After 1952, uranium was mined from these tanks and recovered for reuse. All subsequent fuel processing operations included uranium recovery and recycle. Most of this uranium was shipped offsite for recycle and contained residual trace quantities of transuranics and fission products. By 1994, all plutonium production and uranium recovery activities were shutdown and a cleanup mission for the Hanford site was initiated. A brief historical timeline of Hanford's history is provided in Appendix H.

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support functions were located nearby in facilities in the 700, 1100, and 3000 Areas. Completed fuel was shipped to the reactors at the 100 Areas (B, C, D, DR, F, H, KE, KW, and N) located along the south shore of the Columbia River in the northern portion of the Site for use in the reactors. After irradiation, the fuel was stored in pools at the reactors for a cooling period and then transported in special rail cars to the separation processing facilities located in both the 200 W and 200 E Areas. Discarded high level waste from the separation plants were transferred to storage in underground storage tanks located in both the 200 W and 200 E Areas. All recovered solutions of uranium were transported to the UO_3 Plant located in the 200 W Area for conversion to oxide.

2.2 Key Uranium Processing Facilities

As part of the Manhattan Project and its successors, the U. S. Atomic Energy Commission (AEC), Energy Research and Development Administration (ERDA), and the U. S. Department of Energy (DOE), a number of specialized facilities were constructed at the Hanford site and operated between 1943 and 1986 to produce fuel for the nine plutonium production reactors and to recover plutonium and uranium from the irradiated fuel. A historical overview of all the major plants, process flows, and facility interfaces at Hanford is shown in Figure 2-2. All of the uranium received at Hanford for non-research reactor operation was fabricated into fuel in the 300 Area throughout Hanford's production mission. This fuel was used in nine production reactors which operated over various time periods. Essentially all of the irradiated fuel went to one of four separation processing facilities; T-Plant, B-Plant, REDOX, and PUREX. In addition, the U-Plant was operated for a short time period to recover uranium from high level waste from the early T- and B-Plants, which did not have the capability to recover uranium during irradiated fuel processing. All of the high level waste from the processing of irradiated fuel was transferred to underground storage tanks. Recovered plutonium was refined and converted to a suitable chemical form in either the 231-Z Isolation Building or the Plutonium Finishing Plant. Recovered uranium (as recycle uranium) was converted to a stable oxide for shipment in the UO_3 Plant. The remainder of this section provides a brief description of the facilities, processes, and operating history for the major Hanford uranium processing plants.

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process was developed by Glenn T. Seaborg in the early 1940s and applied at Hanford to achieve separation of the plutonium produced from irradiated uranium. Uranium was not recovered, and both the fission products and uranium were sent to large underground waste storage tanks.

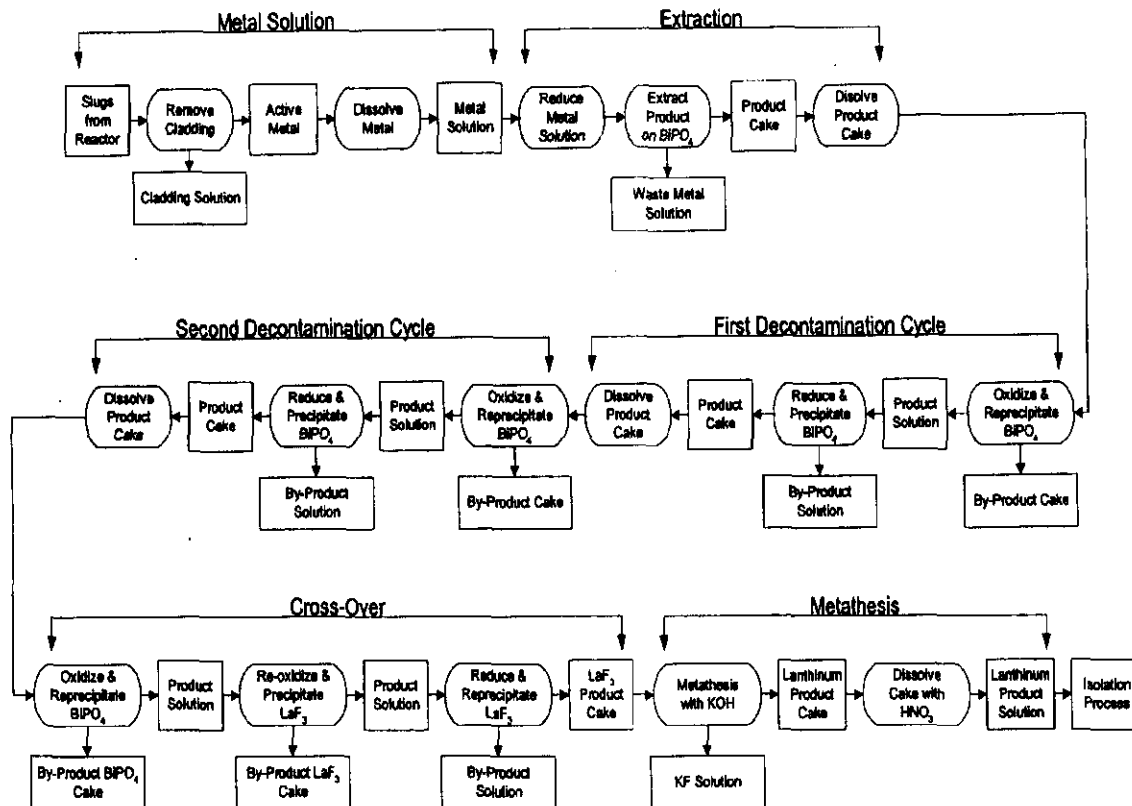


Figure 2-3 T-Plant Process Flow Diagram

The process consisted of removing the aluminum cladding from the irradiated fuel rods (also known as "slugs") in dissolver tanks using hot sodium hydroxide. The cladding waste was drained from the dissolver tanks and transferred to the underground waste tanks. The declad slugs were washed with water to remove any residual hydroxide. Nitric acid was then added to dissolve the bare uranium slugs and form a uranium nitrate solution. The uranium nitrate solution was chemically adjusted with sulfuric acid and sodium nitrite to assure extractability of the plutonium from the uranium solution. Bismuth nitrate and phosphoric acid were added to co-precipitate plutonium and bismuth as insoluble phosphates. The solution was centrifuged from the bismuth phosphate co-precipitate. Waste solutions containing uranium and fission products were treated and sent to the underground waste tanks. The solid cake was water washed and dissolved in concentrated nitric acid. The nitrate solution was chemically adjusted to permit precipitation of metal contaminants upon addition of bismuth nitrate while maintaining the plutonium in solution. The solution was again chemically treated and

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bismuth nitrate and phosphoric acid added to co-precipitate the plutonium, further reducing the metal contaminants. This precipitation, re-solution and re-precipitation process was repeated two more times to further reduce the metal contaminants and reduce volume. Final purification of the plutonium was accomplished by repeated co-precipitation using lanthanum nitrate and hydrofluoric acid to form the lanthanum fluoride which served as the carrier. Upon completion of the purification steps, the plutonium was dissolved in nitric acid and transferred to the Isolation Process Facility (231-Z building) for final treatment and concentration.

2.2.1.3 Feed Specifications

Feed for the extraction process was irradiated natural uranium and the expected plutonium content was low. During the 10 years of T-Plant operation, the irradiated fuel feedstock varied in plutonium and fission products content as the reactor operations were improved and power levels increased.

2.2.1.4 Product Specifications

Plutonium nitrate solution was the principal product of T-Plant and the nitrate solution was further purified at the 231-Z Facility. The uranium and fission products were discharged to the underground waste tanks. Waste transferred to the underground tanks was chemically adjusted to minimize corrosion of the tanks using specifications based on maintaining alkalinity and nitrate/nitrite concentrations. Since uranium did not play an important role in corrosion control, no purity requirements were established for the waste uranium [HW-10475-ABC 1944].

2.2.1.5 Operating History

Processing of irradiated uranium fuel commenced on December 26, 1944, in the T-Plant. A number of operational improvements and installation of a third fuel dissolver in 1954 increased plutonium production rates. T-Plant was shut down in March 1956 following startup of the PUREX plant in January 1956.

2.2.1.6 Current Status

T-Plant is used on an irregular basis as an equipment decontamination and repair facility supporting Tank Farms operations. Although recently the 224-T Building was used for TRU waste storage, the TRU has now been removed and 224-T is now being deactivated.

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2.2.2.3 Feed Specifications

The feed specifications for B-Plant were the same as those for T-Plant, described earlier.

2.2.2.4 Product Specifications

Like T-Plant, B-Plant's principal product was plutonium nitrate solution. The uranium and fission product wastes delivered to the underground tanks were chemically adjusted to assure adequate control of expected tank corrosion using specifications based on maintaining alkalinity and nitrate/nitrite concentrations [HW-10475-ABC 1944].

2.2.2.5 Operating History

B-Plant began processing irradiated uranium fuel in April 1945 using the bismuth phosphate process. Plutonium recovery continued until 1952 when the plant capacity became unnecessary once the combined output of T-Plant and REDOX was sufficient to satisfy production needs. In 1968, the plant was converted to a waste fractionation plant. Cesium and strontium were removed from the high level tank wastes and encapsulated [Gerber 1996].

2.2.2.6 Current Status

The cesium and strontium capsules are currently being stored in water filled basins in an addition on the west end of the plant. Present plans call for vitrification of these capsules as high level waste. The remainder of the plant has been shutdown.

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tributyl phosphate (TBP). The TBP was found to complex very well with both uranium and plutonium to allow their separation from fission products and from each other after proper chemical adjustments. The process consisted basically of contacting the clarified and concentrated acidic uranium solution that had been sluiced from the waste storage tanks with TBP carried in a hydrocarbon solvent to complex the uranium with the TBP. This process was done as a one-pass counter-current solvent extraction process using packed columns. The uranium complexed with the TBP and thus transferred to an organic phase, while the fission products remained in the aqueous phase. The acidic aqueous waste from the column was neutralized and returned to the underground waste tanks for storage. The uranium was then stripped from the TBP-uranium complex in the organic phase in another packed column with acid of a specific molarity. The resultant uranium solution was then concentrated in an evaporator to a consistent concentration for feed to the UO_3 Plant. The concentrated uranium nitrate solution (uranyl nitrate hexahydrate (UNH)) was analyzed to assure that it met feed specifications for the UO_3 Plant and then transferred to that facility for conversion to shippable powder. Product solutions not meeting the stringent specification of the UO_3 Plant were recycled back through the extraction process. In order to accommodate the required throughput rate, there were two extraction trains in U-Plant.

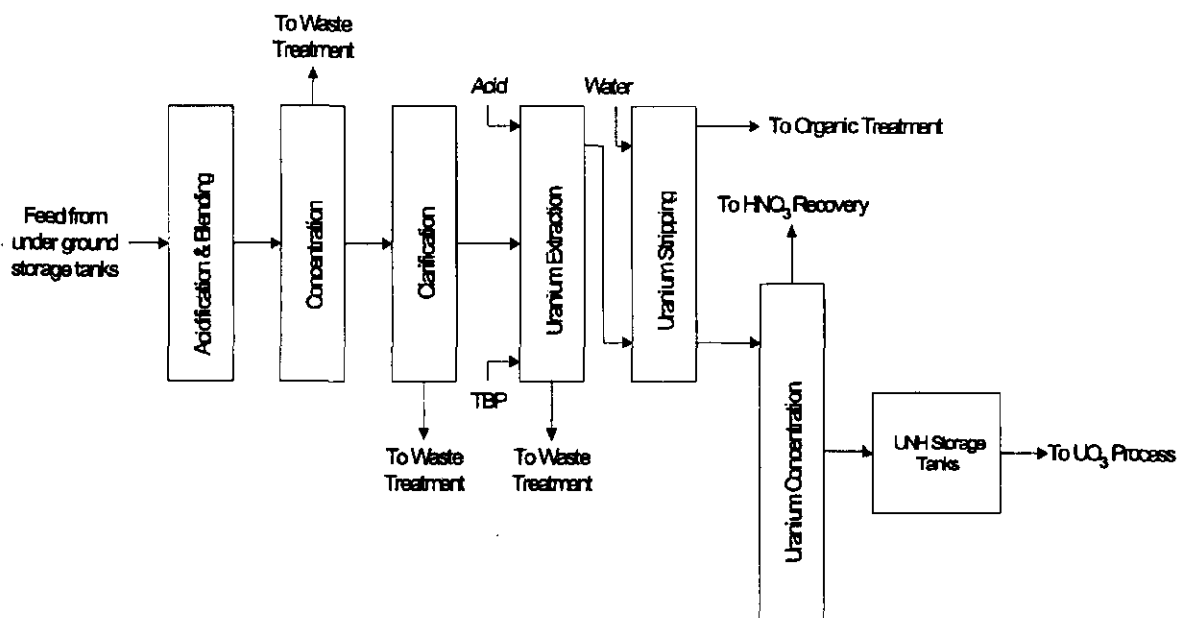


Figure 2-4 Simplified U-Plant Tributyl Phosphate (TBP) Process Flow Diagram

2.2.3.3 Feed Specifications

Feed for U-Plant came from the underground storage tanks which contained high level waste from the B and T separation plants. The uranium-bearing tank wastes consisted

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of sludge and supernatant liquor (sludge washings and solutions). The sludge contained approximately 75 percent of the uranium. The plutonium content of the uranium waste was about two to four grams per metric ton (MT) of uranium (2-4 ppm Pu). Approximately 20 MT of slurried tank waste was required to produce one ton of recovered uranium for conversion to UO_3 . A description of the chemical constituents of the "mined" uranium feed can be found in the Uranium Recovery Technical Manual [HW-19140 1951].

2.2.3.4 Product Specifications

The U-Plant product specifications were dictated by the specifications for the UO_3 powder produced in the UO_3 Plant. The initial specification in the 1951 technical manual [HW-19140 1951] indicated the UNH product from U-Plant should contain less than 80 ppb Pu. The UO_3 Plant did not further refine the UNH product from the U-Plant process. Blending of the U-Plant product with product from REDOX was expected to help assure the concentration of transuranics and fission product impurities were at acceptable levels. U-Plant did have the capability to concentrate the UNH prior to calcination, however this did not alter the ratio of any impurities to uranium.

2.2.3.5 Operating History

Uranium was scarce in the late 1940s. Escalating demand for weapons materials led to the decision to reclaim uranium from the underground storage tanks. This was first discussed in 1947 when a decision was made to develop a process for extracting uranium from the Hanford waste tanks to supplement the scarce uranium supplies [Peterson 1947]. Over the next few years, a uranium recovery process based upon TPB was developed at Hanford and served as the design basis for both the U-Plant conversion and the PUREX separations operations. U-Plant began recovering uranium from tank wastes in 1952 and completed its mission in 1958. UNH product from U-Plant was routinely blended with UNH product from REDOX. During U-Plant's operating period, ~7,200 MTU was recovered from high-level waste for conversion to UO_3 and recycle.

Worker exposure to uranium was normally low, as processing occurred remotely in heavily shielded and ventilated canyon cells. The uranium solutions were of low concentration until the solution was evaporated to create the UNH product. Transfer of concentrated UNH to the UO_3 plant was by pipeline. Liquid wastes containing fission products and plutonium were transferred to underground waste storage tanks. Solid wastes were packaged and sent to the Hanford burial sites for disposal.

2.2.3.6 Current Status

U-Plant is presently undergoing decontamination and decommissioning. It's final state is the subject of an ongoing Canyon Disposition Initiative at Hanford.

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2.2.4.2 Material Flowsheet

The initial UO_3 process operations utilized a batch calcination process involving 18 stirred kettles, as shown in Figure 2-5. The concentrated UNH was initially heated in the stirred kettles until it was dry and then further heated until the UNH decomposed into UO_3 . The nitric acid resulting from the drying and calcining processes was recovered and concentrated. The concentrated nitric acid was returned to the separation plants for reuse. The UO_3 powder was removed from the kettles and pneumatically transferred into storage bins. The air used to transport the UO_3 was filtered before discharge to the atmosphere. The condensate stream was sampled to verify compliance with existing regulations and discharged to the cribs. The UO_3

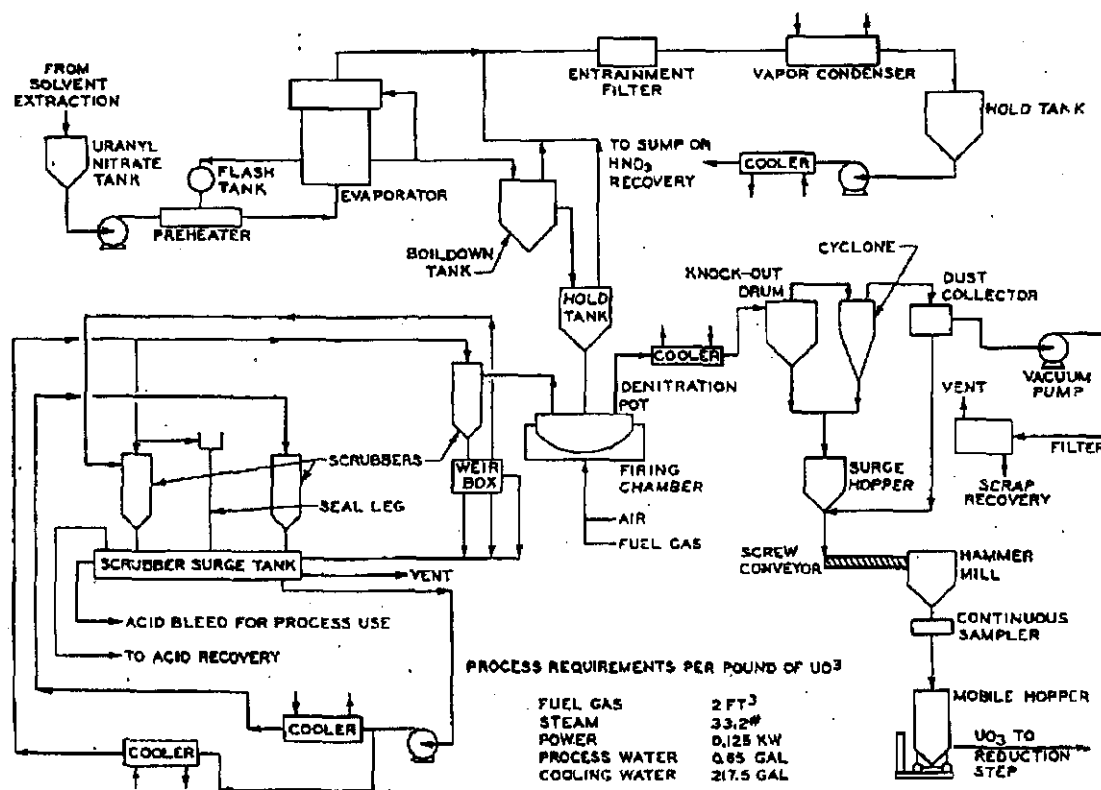


Figure 2-5 UO_3 Plant Process Diagram Involving Batch Pot Calciners

product was sampled and tested to assure compliance with product specifications. At times, the powder was ground to a specific particle size to meet chemical reactivity specifications. Sulfuric acid was added to the UNH to control the hydrofluorination reactivity of the UO_3 powder at the gaseous diffusion plants. Off-specification UO_3

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powder was either blended with acceptable UO_3 product to bring the impurities to within specification requirements, or waivers were obtained before shipment.

In 1956, the 18 kettle calciners were replaced with 6 stirred continuous calciners as shown in Figure 2-6 to improve the product quality and increase the production rate. In addition, an evaporator further concentrated the UNH feed solution from the separation plants to allow more effective calcination. The concentrated UNH was sprayed on a heated UO_3 powder bed in the continuous calciners. The UNH quickly dried, decomposed and was calcined to UO_3 powder. The UO_3 product overflowed the calciner and was pneumatically transferred to the holding bins pending acceptance testing and subsequent packaging into T-hoppers or drums. Nitric acid driven off in the calcination process was recovered and returned to the separation plants for reuse.

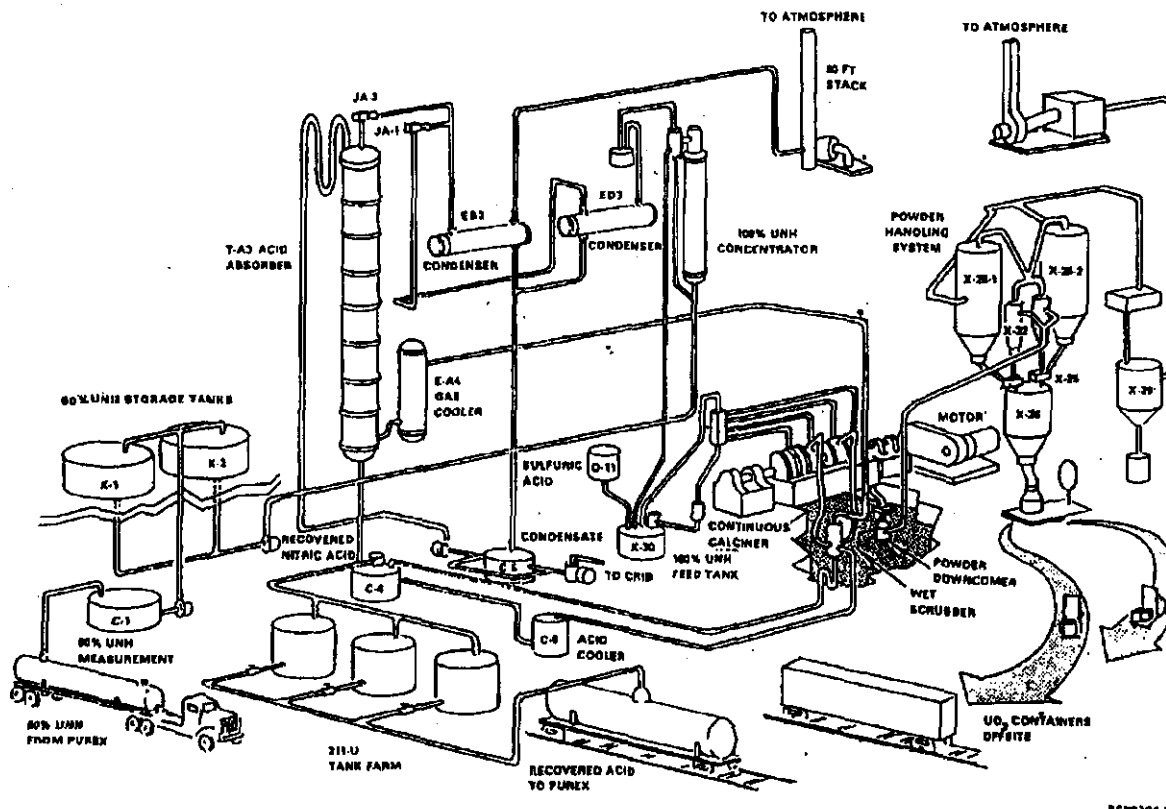


Figure 2-6 UO_3 Plant Process Diagram Involving Continuous Calciners

During the time when UO_3 Plant operated with continuous calciners, the product loadout system was configured approximately as shown in Figure 2-7. Powder in the pickup bin was fluidized and transported to cyclone separators on the fourth floor of the 224-UA tower. The heavy powder dropped out of the cyclone into the storage hopper. The remaining air and powder fines were filtered on primary bag filters, secondary filter

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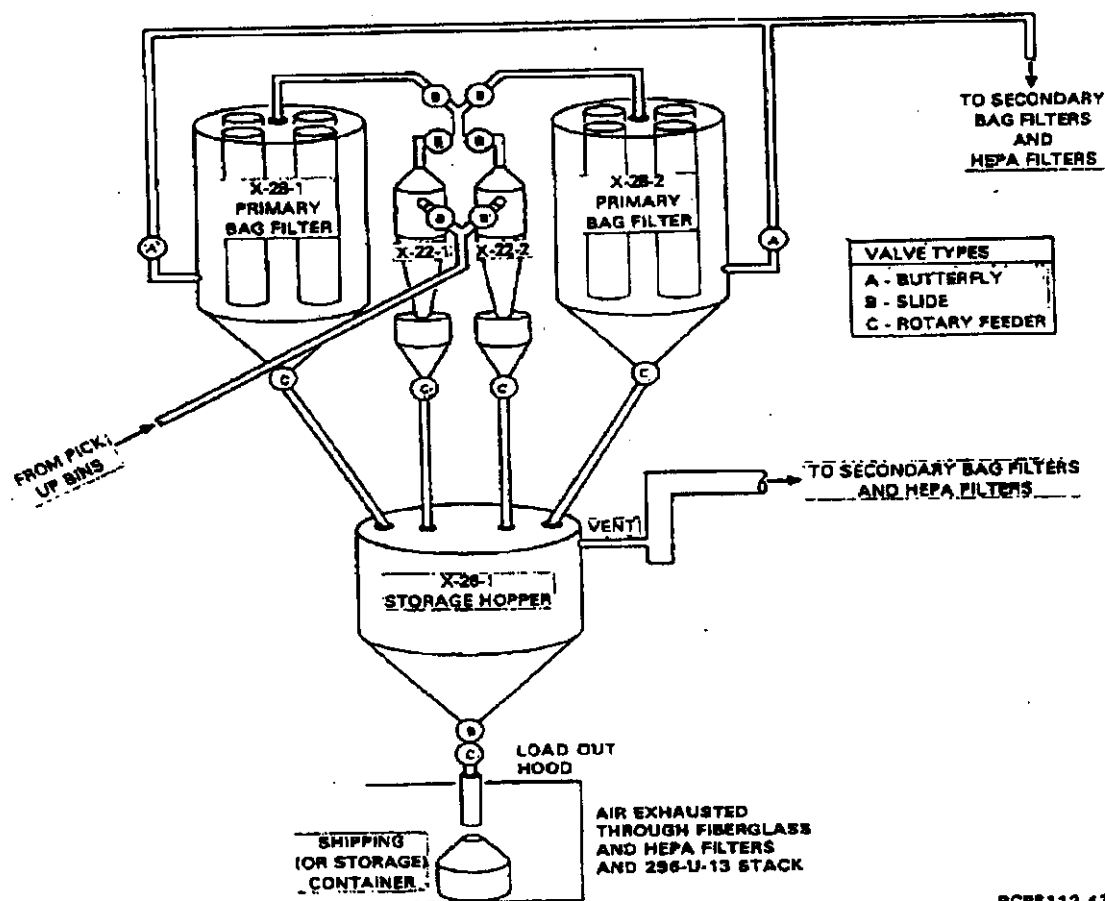


Figure 2-7 UO₃ Product Loadout System

bags, and then on high efficiency particulate air (HEPA) filters, which were added during the 1972 to 1984 shutdown time period. The powder was sampled for acceptance testing [Gustavson 1950].

Acceptable powder in the storage hopper was loaded into approved containers in the 224-UA loadout room. A forklift transported these containers to the 2714-U storage area. From this area, the UO₃ powder was shipped offsite by railcar or truck.

Of all Hanford uranium recycle activities, operation of the UO₃ Plant presented one of the greatest potentials for worker inhalation exposure to uranium and its contaminants because the product was a powder. In this facility, liquid uranium solutions were converted into dry UO₃ powder in ventilated batch kettles or continuous calciners. The calciners were ventilated to recover the nitric acid produced during calcination for reuse. The dry powder was handled pneumatically to minimize worker exposure. Packaging of the UO₃ into drums and T-Hoppers and maintenance on calciners and off-gas filters

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offered the highest potential for exposure to airborne powder. The transfer operation was performed within a ventilated enclosure to assist in dust control.

More detailed information on the UO_3 Plant is provided in Appendix G.

2.2.4.3 Feed Specifications

Feed specifications for the initial operation of the UO_3 Plant are provided in the "Uranium Recovery Technical Manual" [HW-19140 1951]. This specification indicates that the product material from REDOX and U-Plant was tightly controlled, as the UO_3 process provided no further purification of the uranium. The need for process control was recognized in the late 1940s and early 1950s before the plant came on line. The need to maintain the plutonium contamination to very low concentrations was recognized and included in the technical manual. Decisions were made in the late 1940s [Peterson 1947] to enrich the depleted uranium back to normal concentrations. Discussions followed regarding "firm specifications" for the final uranium product to be delivered [Greninger 1950].

The separation plants were originally designed on the basis that the recovered uranium would be sufficiently decontaminated with respect to Pu and gross beta and gamma radiation to permit essentially direct physical handling of the final product in its last form at Hanford. It was also recognized at this early stage that subsequent processing at other plants might result in fractionation or concentration of either fission products or of plutonium and cause a need for more highly specific or greater decontamination than would be required at Hanford. In a 1951 letter [Gamertsfelder 1951], it is stated that "...reclaimed uranium should contain no more than one part plutonium in 7.8×10^6 parts uranium." in order that the hazard due to breathing air contaminated with reclaimed uranium should be no more than 10% greater than for ordinary uranium.

2.2.4.4 Product Specifications:

A "tentative" UO_3 product specification was initially defined in the "REDOX Technical Manual" [HW-18700 1951]. This tentative specification established the following constituent limits:

- | | |
|--|---|
| • Beta Activity from Fission Products | Not more than 30% of beta activity of natural uranium |
| • Gamma Activity from Fission Products | Not more than 300% of gamma activity of natural uranium |
| • UO_3 Purity | 97% minimum |
| • Plutonium | 100 parts per billion parts of uranium |

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Negotiations continued on UO_3 specifications in 1952, when preliminary specifications were considered which indicated a desired maximum plutonium concentration of 10 ppbp uranium [Hurd 1952]. In 1953, based on operating experience, representatives from Hanford and Oak Ridge agreed upon the properties of the Hanford UO_3 to be included in a firm specification [Smith 1953]. These properties included fission product activity, plutonium content, purity (UO_3 content), particle size, and volatile impurity content, and specified that "The maximum acceptable plutonium concentration shall be ten parts of plutonium per billion parts of uranium. Plutonium shall be determined on each carload composite." The 1953 revised specification is given in [Smith 1953]. The new constituent levels were established as follows:

Firm Specifications Properties:

- Gamma Activity - the average gamma activity for any ten consecutive carload shipments shall be 100% the gamma activity of aged natural uranium. Defined as follows: for the purpose of establishing the average acceptable gamma activity specification, shipments of uranium oxide to K-25 will be considered in designated blocks of ten consecutive carloads. The average acceptable gamma activity due to fission products for a block shall be 100% the gamma activity of aged natural uranium. For the purpose of establishing the maximum gamma activity specification, one carload of oxide shall be considered a unit. The maximum acceptable gamma activity, due to fission products, for a unit shall be 300% the gamma activity of aged natural uranium.
- Beta Activity - the maximum acceptable beta activity, due to fission products, shall be 100% the beta activity of aged natural uranium. Beta activity shall be determined on each carload composite.
- Plutonium Content - the maximum acceptable plutonium concentration shall be ten parts of plutonium per billion parts of uranium. Plutonium shall be determined on each carload composite.
- UO_3 Content - the minimum acceptable UO_3 content shall be 97%. The UO_3 content shall be determined on each lot of 8 drums.

During the operating history of UO_3 production, numerous changes were made to the product specification to better control specific isotopes contributing to beta-gamma exposure of workers. Figure 2-6 shows a copy from one of the early Hanford UO_3 product specifications, indicating an allowed maximum plutonium concentration of 10 ppbp uranium [Smith 1959]. A more complete discussion on specification development and compliance is provided in Section 4.

Product specifications were also discussed, but not adopted, for ^{233}U , ^{232}U , ^{237}Np , and ^{99}Tc . In 1962 [Judson 1962] it was proposed that the maximum concentration of ^{233}U be set at 90 ppm on a ^{235}U basis, and that ^{232}U be set at 110×10^{-4} ppm on a ^{235}U

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	HM-59136 Page -5-
Analysis	Method Ref.
Sodium	1000.1, 769
Calcium	1000.1
Aluminum	1000.1
Iron	1000.1, 515.25
Chromium	1000.1, 341.1
Nickel	1000.1
Density, bulk	847.2
Density, packed	847.5
3.22 Properties with Established Limits	
3.22.1 Gamma Activity	
Gamma activity due to fission products shall be determined on each earload composite. For the purpose of setting the average acceptable gamma activity, shipments of uranium oxide will be considered in designated blocks of ten consecutive earloads. The average acceptable gamma activity due to fission products for a block shall not exceed 100 percent of the gamma activity for aged normal uranium. For the purpose of setting the maximum acceptable gamma activity specification, one earload shall be considered a unit. The maximum acceptable gamma activity for a unit, due to fission products, shall be 300 percent of the gamma activity of aged normal uranium.	
3.22.2 Beta Activity	
Beta activity due to fission products shall be determined on each earload composite. The maximum acceptable beta activity due to fission products shall be 100 percent of the beta activity of aged normal uranium.	
3.22.3 Plutonium Content	
Plutonium shall be determined on each earload composite. The maximum acceptable plutonium concentration shall be ten parts of plutonium per billion parts of uranium.	

Figure 2-8 Copy Of Early UO₃ Product Specification

basis. In 1971, it was recommended a Np specification of <1 ppm for a lot composite and <0.3 ppm on 10 lot composites [Elgert 1971]. And in 1982 a ⁹⁹Tc specification of 0.4 ppm was proposed [Miskho 1982]. At that time it was implied that there was a 400 ppm limit for ⁹⁹Tc, but it has never been part of the Hanford specifications for UO₃ product.

2.2.4.5 Operating History

The facility began test runs in January 1952, and was in full operation the following month. Thus, for the first time, a full cycle of uranium into and out of Hanford was completed. The first UO₃ calciners simply were large pots that heated the UNH in batches. During the initial operation of U-Plant, the UNH product resulted in UO₃ product which required additional purification to remove non-radioactive impurities. This material was shipped off-site for purification. By the end of 1953, however, process improvements in both the REDOX Plant and U-Plant resulted in UNH so pure that the UO₃ Plant product no longer had to be sent offsite for further purification. This UO₃ was able to be shipped and used directly at Oak Ridge.

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During 1954, UO_3 Plant capacity increased to 18 tons per day with the addition of two gas-fired calciners. Overall production in 1954 was limited, however, by the quantity of feed available from the REDOX and U-Plants. A key improvement that year was the use of vacuum cleaning techniques that enabled the plant to recover 90-95 percent of the oxide powder associated with failed filter bags.

During 1955, monthly UO_3 production averaged 106 percent that of 1954, while the unit costs per ton dropped to 92 percent those of the previous year. In 1956, equipment modifications, including new pot agitators, shafts, seals, bearings and supports, as well as new pot cover assemblies, off-gas systems, and gasketing were employed.

In the same time frame, plans went forward to construct the 224-UA Building, a major addition to the UO_3 Plant, with six large, new, continuous action calciners. These calciners were developed and designed at Hanford. They had large troughs with paddle agitators that turned and mixed the UNH/ UO_3 bed throughout the process. The caking and clogging problems that had plagued the pots were obviated by the below-grade valving of the continuous calciners. The new valves used air to agitate the powder, and provided a seal between the calciner and the powder pickup bin, while passing a continuous stream of UO_3 .

The new calciners also produced a pebbly product that consisted of spheres with an average diameter of 200 microns (about 1/100th of an inch), as opposed to the granular oxide product of the pots. The first three continuous calciners began operation in the last quarter of 1956 at the same time specially designed T-Hoppers began to replace the 900 pound drums as shipping containers. The T-Hoppers left the site on specially fitted rail flat cars and served as feed hoppers for the Oak Ridge plant customer.

Installation of the last three of the continuous calciners was completed in early 1957. At that time, the 18 pot units were retired from service, and all processing was done through the continuous calciners. Late that year, design was initiated for new facilities which were needed to segregate regular UNH feed from that generated by the processing of enriched metal (E-Metal) in the REDOX Plant. The UO_3 powder that resulted from processing the two types of UNH streams had significantly different nuclear reactivity levels. It was necessary to maintain separation of these streams for the customer at Oak Ridge. Routine transfers of enriched UO_3 from Hanford to Oak Ridge began in early 1959.

In 1958, U-Plant finished its uranium recovery mission and was shut down. However, the quantity of PUREX Plant UNH product being shipped to the UO_3 Plant far surpassed that which had come from U-Plant. Together, the UO_3 and PUREX Plants went on to set and surpass production records almost continually, while the REDOX Plant continued to supply a small stream of enriched UNH until its shutdown in 1967.

During 1959, the concentration equipment and the acid recovery system at the UO_3 Plant were automated. In 1960, the calciners were also automated so that they could be operated and shut down remotely. In 1963, production of UO_3 shipped offsite rose to

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was the technological answer to the need to increase plutonium production and uranium recovery to satisfy growing national defense needs.

2.2.5.1 Plant Description

Experimentation and design for the REDOX (reduction oxidation) Plant began in 1947 with actual construction beginning in late 1949. The facility commenced operations in 1952. The building is 467 feet long by 161 feet wide by 82 feet high. At one end of the building is a "penthouse", 132 feet high, which housed the extraction columns. It is constructed of thick, steel-reinforced concrete. The processing of irradiated fuel was accomplished in nine below-grade, thickly shielded, concrete cells. Support facilities, including the 222-S laboratory, were constructed in the same time frame. Like the earlier T- and B-Plants, the process first dissolved the irradiated fuel cladding and then dissolved the uranium core. The plutonium was separated from the uranium in a continuous process utilizing methyl isobutyl ketone (hexone). The plant design-basis production rate was about 3 MT uranium per day [Gerber 1996].

2.2.5.2 Material Flowsheet

The REDOX process offered substantial improvement over the T- and B-Plant separation processes by employing continuous solvent extraction. Under specific conditions, plutonium and uranium were found to extract into hexone solvent, allowing separation from the fission products, which remained in aqueous solution. Figure 2-9 shows a simplified flow diagram of the process.

The irradiated reactor fuel was removed from the shipping cask and transferred to a dissolving tank to permit selective removal of the protective aluminum cladding using a caustic solution. The coating waste was neutralized and sent to underground waste tanks for storage. The bare irradiated uranium slugs in the dissolver tank were then reacted with nitric acid to dissolve the uranium and accompanying plutonium and fission products. The resulting uranium nitrate solution was first treated with sodium dichromate and sodium hydroxide to adjust the plutonium valence state and then was contacted with an acidic hexone solvent in a countercurrent extraction column.

The uranium and plutonium formed a complex with the acidified hexone and were separated from the fission products. The aqueous waste was neutralized, treated and transferred to the waste tanks for storage. The plutonium/uranium/hexone complex was treated again to adjust the plutonium and uranium valences to permit separation of the plutonium from the uranium in the next countercurrent extraction column, where the plutonium was stripped from the hexone complex into a nitric acid solution. The uranium/hexone solution was further treated and the uranium removed by a nitric acid wash. Once the separation had been completed, the uranium and plutonium streams were separately treated in two more similar extraction/separation steps through countercurrent extraction columns to further remove impurities and provide product nitrate solutions meeting stringent impurity specifications. Off-specification product

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solutions were sent back into the extraction process for rework. The hexone solvent was washed, neutralized and re-distilled for recycle into the extraction process

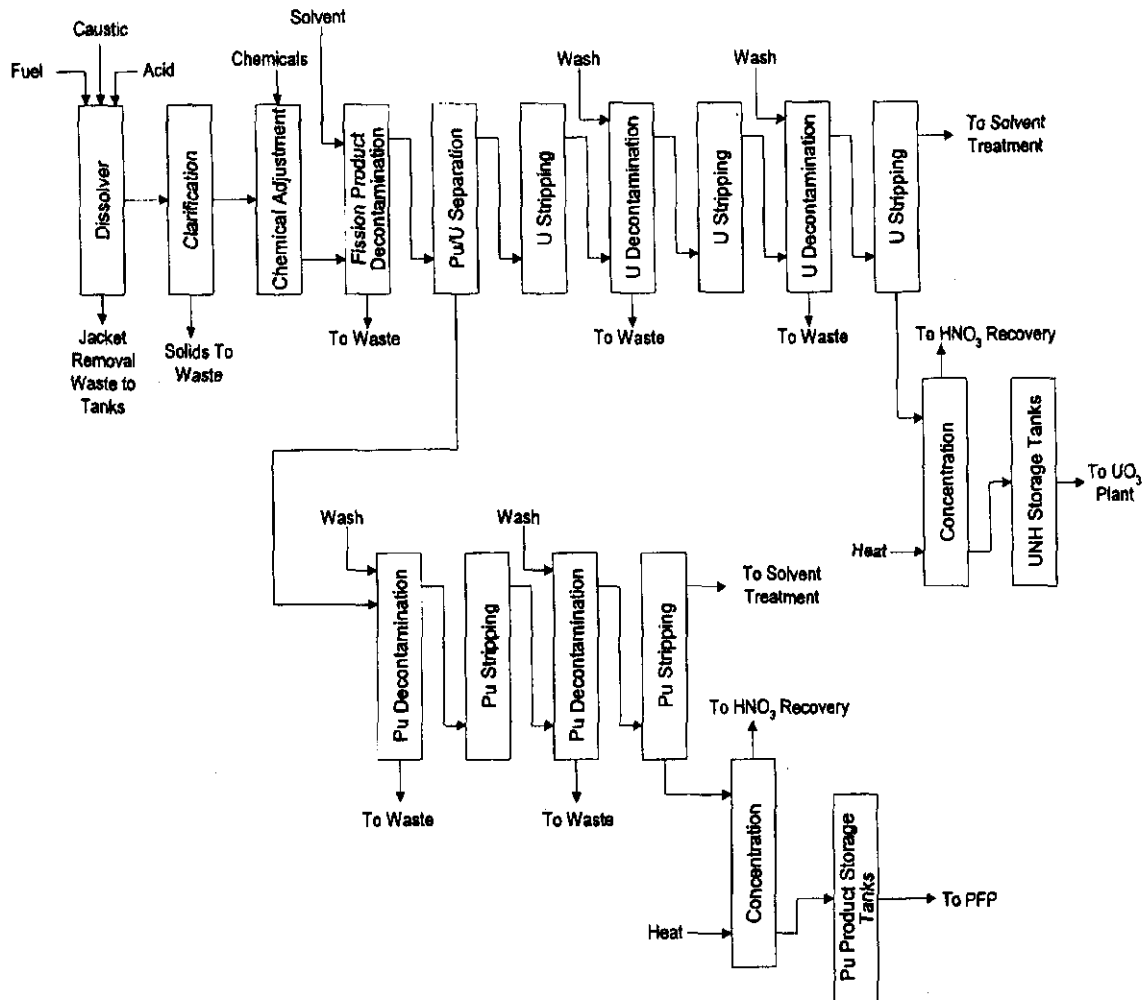


Figure 2-9 REDOX Process Flow Diagram (Simplified)

2.2.5.3 Feed Specifications

The irradiated fuel was aged before processing to eliminate some of the fast decay fission products and to assure that the product beta and gamma radiation limits were met.

2.2.5.4 Product Specifications

The uranium product specifications were driven by the UO_3 product specification requirements. The UO_3 Plant processing of UNH product from REDOX, did not further

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refine the uranium, thus the feed UNH from REDOX had to meet the final UO_3 product specification limits. The plutonium content of three trial lots of UO_3 prepared from material processed through the REDOX Plant in 1952 was less than 5 parts per billion parts (ppbp) uranium [Richards 1952]. It was also reported in June 1952 that the average plutonium concentration in REDOX recovered uranium was less than 1 ppb [HW-24605 1952].

2.2.5.5 Operating History

The REDOX plant commenced "hot" operations in January 1952. By April 1952 the plant achieved its design rate production throughput. At the end of 1952 the plant had exceeded its throughput rate by 50% for several months. A series of three "Capacity Increase" projects began in 1953 with the result that the production throughput rate being increased by a factor of 2-3 times that achieved in 1953. By the end of 1954 the throughput rate reached 8 tons per day. Additional improvements brought the throughput rate to 11-12 tons per day by 1958. Part of the capacity increases included construction of the 233-S Plutonium Concentration Building. Completed in 1957, this building carried out the third and final plutonium concentration cycle. Processing of enriched irradiated fuel assemblies (E-metal) began at REDOX in 1958. E-metal throughput initially was limited to 3 tons per day due to criticality considerations in the plant. After installation of multipurpose annular dissolvers and other process improvements, the plant throughput rate reached 12 tons per day for enriched fuel. The REDOX Plant operated until 1967 when it was shut down [Gerber 1996].

2.2.5.6 Current Status

The REDOX Plant is shut down and awaiting decontamination and decommissioning. The 222-S laboratory is currently in operation supporting waste management and environmental control processes.

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contaminated cask cars without compromising the ventilation system, and (3) a "soft wall" at the east end of the building that consisted of concrete blocks. These blocks could be removed for the installation of an additional crane, or to enlarge the building at some future date [Gerber 1996].

2.2.6.2 Material Flowsheet

PUREX operated under a number of different flowsheets that were the result of process and equipment improvements and changing reactor fuel compositions. Additionally, some campaigns involved neptunium and the processing of thorium based reactor fuels. A typical process flow diagram for PUREX processing of N-Reactor irradiated fuel is shown in Figure 2-10. Not shown on this diagram is the Plutonium Oxide Production and Rework Facilities that were added in the PUREX N-cell in the early 1980s. This process allowed conversion of recovered plutonium nitrate to plutonium oxide without having to send the solution to the PFP for conversion to the oxide.

The PUREX process was the next generation separations process. It utilized tributyl phosphate (TBP) to complex with plutonium and uranium under specific conditions and to cleanly separate the uranium and plutonium. The irradiated reactor fuel elements were removed from the storage basin and transferred into three dissolvers. The aluminum cladding was dissolved from the uranium fuel rod with sodium hydroxide. The cladding waste solution was treated to meet waste tank corrosion specifications and transferred to the waste tanks.

The PUREX Plant also processed zirconium clad fuel, which required the use of an ammonium fluoride and ammonium nitrate decladding solution at near boiling conditions. The zirconium-containing decladding waste from the dissolvers was centrifuged and treated to meet Tank Farm waste solution corrosion specifications. The centrifuged solids contained recoverable amounts of uranium and were accumulated and subsequently treated for uranium and plutonium recovery. Next, nitric acid was added in two increments to the dissolvers to dissolve the uranium, plutonium, and fission products into a solution that permitted transfer and separation. The dissolvers were maintained at elevated temperature to facilitate fuel dissolution.

Once the uranium and plutonium and fission products were dissolved, the solution was fed continuously into a pulsed extraction column where the uranium, neptunium, and plutonium were stripped away from the nitric acid solution into a normal-paraffin-hydrocarbon (NPH) solvent containing TBP. The fission products remained in the aqueous solution and were discarded after treatment to the waste tanks. The decontaminated solution of uranium, neptunium and plutonium in the TBP complex was next pumped through a second pulse column where the plutonium was selectively stripped out of the NPH/TBP complex into an aqueous nitric acid solution.

The NPH/TBP complex containing uranium and neptunium was sent to the next pulse column where the uranium and neptunium were stripped from the TBP complex using a weak aqueous nitric acid solution. The uranium/neptunium solution went to the

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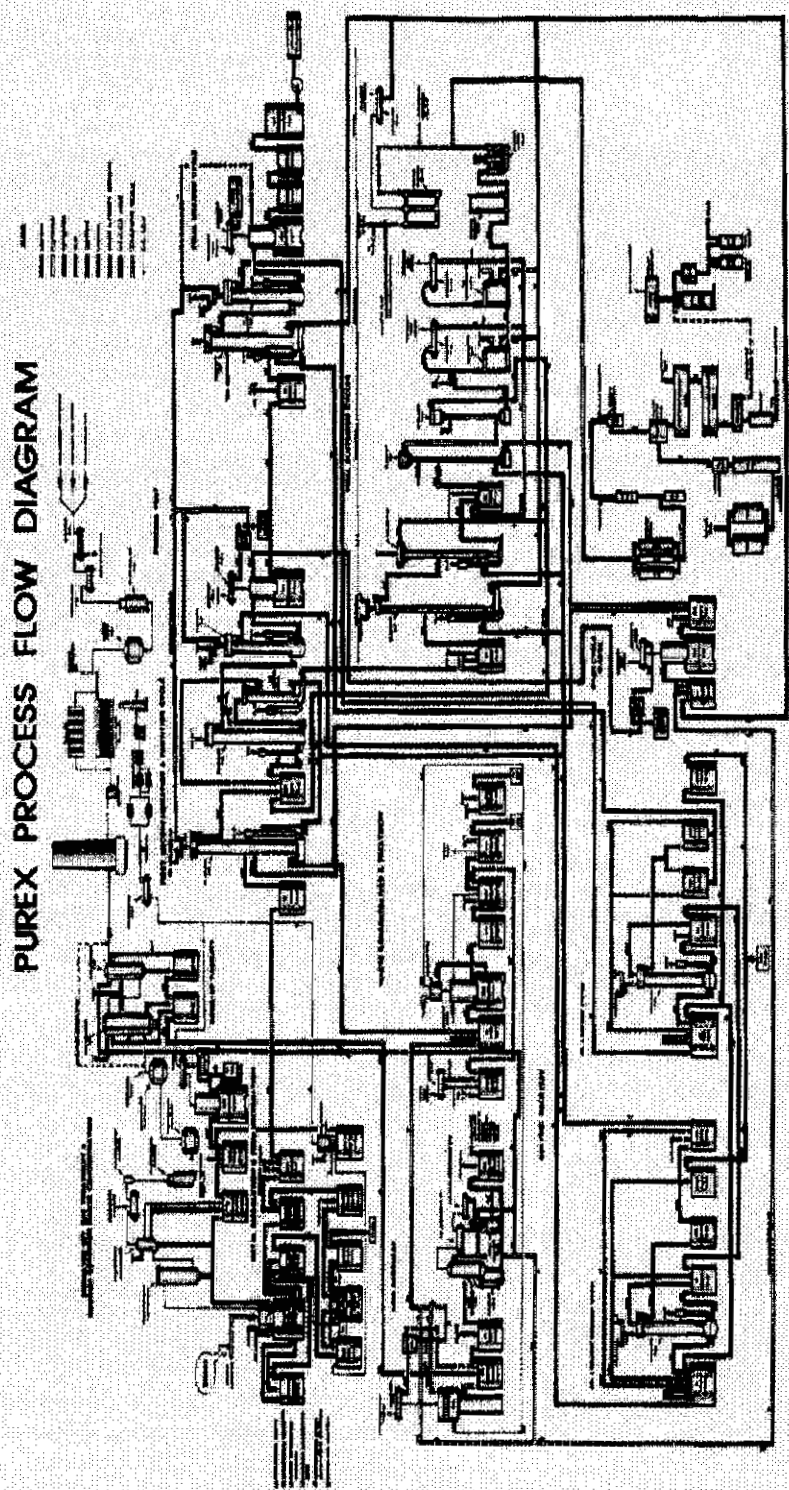


Figure 2-10 PUREX Process Flow Diagram
(excluding the Plutonium Production and Rework Facilities)

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evaporator for concentration before entering the final uranium cycle. The concentrated uranium/neptunium solution was routed to a feed tank for acid adjustment before entering a pulsed stripping column where the uranium/neptunium solution was contacted with NPH/TBP solution. In this column, the uranium was selectively extracted into the NPH/TBP while the neptunium remained with the aqueous flow. The NPH/TBP/uranium complex was then sent to another column for stripping uranium into an aqueous solution which was subsequently concentrated in an evaporator and, after sampling, transferred to the UO_3 feed tank.

The plutonium stream from the second pulsed column was sent to the second and third plutonium cycle extraction columns where it was contacted with NPH/TBP solution repeatedly to remove any residual fission products and trace uranium. Following the completion of the two cleaning cycles the plutonium solution was concentrated to meet product specifications.

Product solutions not meeting specification were recycled back through the process for cleanup. If needed, waste solutions from the columns were scavenged for uranium, plutonium and neptunium before treatment to meet waste tank acceptance criteria. The used NPH/TBP solutions were treated to remove impurities, re-acidified and returned to the process. Nitric acid off-gasses were recovered, concentrated into nitric acid, and returned to the process.

2.2.6.3 Feed Specifications

PUREX feed specifications were driven by the type of reactor fuel that was to be recovered for its plutonium content. The process was flexible and permitted feeds of different enrichment. Criticality control of the fuel dissolution process drove the size and configuration of the plant dissolvers. PUREX processed irradiated fuel initially containing up to 2.1% ^{235}U .

2.2.6.4 Product Specifications

The plutonium and uranium product compositions were derived from the feed requirements imposed by the UO_3 Plant and the Plutonium Finishing Plant (PFP). A typical specification for the UNH product solution is shown in Figure 2-11. For additional information on specification development, see Section 4.

2.2.6.5 Operating History

The PUREX Plant was originally designed to process up to 200 MT of irradiated uranium per month. It achieved this rate and higher over the years it operated. In 1956 alone, the PUREX plant processed 56 percent of the annual plutonium output of Hanford for that year, which was 59% above that of 1955. In September 1956, PUREX demonstrated a sustained, instantaneous rate of 16 MTU/day of irradiated uranium and an on-line efficiency of 99 percent. The following year, the total output of plutonium from Hanford increased by 54 percent over that of 1956, with PUREX processing 71

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percent of this new total. The plant demonstrated an instantaneous production rate of 20 MTU/day, with a reduction in the amount of residual fission product activity in the product of 25 fold below design specifications in 1957.

	108	HW-31000
<p>In terms of a Purex-Plant uranium product for inter-area shipment, the following approximate specifications should be met:</p>		
Pu	≤ 10 parts per billion parts of uranium	
Gamma activity due to fission-products	≤ 200% of aged natural uranium	
Beta activity due to fission-products	≤ 200% of aged natural uranium	
Total metallic impurities (T.M.I.)	≤ 200 parts per million parts of uranium	
<p><u>Note:</u> The fission-product specification for inter-area shipment is dependent upon the individual fission products present in the uranium product. If, as is expected, Pu constitutes a substantial fraction of the activity, it is permissible to exceed the final UO₂ specification since Pu is largely volatilized in the calcination of uranyl nitrate to the oxide.</p>		
<p>C. <u>PRINCIPLES AND OUTLINE OF THE PUREX PROCESS</u></p>		
<p>The Purex process is designed to separate uranium and plutonium from each other and from the fission products with which they are associated in irradiated slugs. The process is one of solvent extraction, in which the components are separated from one another by controlling their relative phase distribution between aqueous solutions and an immiscible organic solvent, tributyl phosphate dissolved in a hydrocarbon diluent. In the following subsections the basic principles of the process are briefly described and the several steps which make up the process are outlined. This section is intended only as an introduction to the process, more complete information being contained in Part II (Chapters II through X).</p>		
<p>1. <u>Basic Principles</u></p>		
<p>1.1 <u>Properties of uranium of process importance</u></p>		
<p>The Purex process utilizes the preferential extractability of uranyl nitrate by tributyl phosphate to separate uranium from plutonium and the fission-product elements.</p>		
<p>Metallic uranium is soluble in nitric acid to form an aqueous solution of UO₂(NO₃)₂·xH₂O. A rapid dissolution rate of the metal in nitric acid is favored by a high concentration of acid, and elevated temperature.</p>		

Figure 2-11 Example of PUREX Product Specification

In 1958, the decision was made to send virtually all of the standard irradiated uranium from the 100 Areas production reactors to PUREX, and to divert processing of enriched uranium containing 0.9475 wt% ²³⁵U ("94 Metal") to the REDOX plant. REDOX equipment was reconfigured and handling techniques were changed to accommodate the higher enrichment levels of the 94 Metal. Additionally in 1958, the PUREX facility began the recovery of neptunium (²³⁷Np) on an occasional batch basis from its normal product stream. The plant became the AEC's prime supplier of this isotope. For short periods of time, PUREX demonstrated the capacity to operate at 3.6 times its original

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design capacity. In late 1965, other PUREX systems were modified to allow the plant to sustain normal operations at a 4.0 capacity factor, or 33 MTU/day [Gerber 1996].

Beginning in 1963, the PUREX plant was modified to allow for the processing of various fuel types, including fuel from N-Reactor with elements much larger than the four to eight inch long "slugs" from Hanford's single pass reactors. In 1967, the REDOX plant closed, and PUREX became the sole, operating separations facility at Hanford. In 1972, the PUREX plant entered a temporary shutdown period that lasted for 11 years. All of Hanford's single-pass reactors had closed, and most of the available (aged) N-Reactor fuel had been processed. The initial PUREX shutdown was planned for 18 months, to allow the accumulation of N-Reactor fuel. During the shutdown period, other issues arose, including environmental concerns that led to providing upgraded filtration systems, seismic safeguards, backup power sources and many other projects. Increased safety concerns about shipping plutonium in nitrate form from PUREX in the 200-E Area to the Plutonium Finishing Plant in the 200-W Area, led to an additional delay. The shutdown period was extended while facilities were added within the PUREX plant to convert plutonium nitrate to plutonium oxide.

In 1983, the PUREX plant reopened with an operating limit allowing it to process up to 3,000 MTU/year of N-Reactor fuel, or about eight MTU/day. However, the highest sustained (but short-term) production rates in the ensuing years hovered near seven MTU/day, and the highest long-term rates were about three MTU/day. The PUREX facility closed for about six weeks in 1988, for a safety issue, and again for a year beginning in December 1988, after steam pressures briefly fell below levels necessary to support back-up safety equipment. Additional equipment repairs and improvements to waste handling systems also occurred during that closure period. After a stabilization run lasting only a few weeks, the plant again closed in early 1990 in order to prepare additional environmental and safety documentation and facility upgrades. In total, PUREX processed approximately 69 percent of all the irradiated fuel produced at Hanford. In October of that year, the PUREX Plant was placed on standby status by the Secretary of Energy, James Watkins. A final closure order was issued by the DOE in December 1992 [Gerber 1996].

2.2.6.6 Current Status

The PUREX Plant has been deactivated and is awaiting final disposition.

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The foundation and floors were poured concrete. The roof consisted of insulated metal paneling covered with felt and roll tarpaper and a tar and gravel surface. The structure was 300 feet by 140 feet, with a total area of 48,817 square feet. In 1980, in response to anticipated increases in production, a small addition was placed on the northwest corner of the 333 Building. It consisted of two stories; the ground level for an open bay shop and the second story for offices. The addition was 33 feet by 104 feet, and extended from the HVAC (heating, cooling and ventilating) supply units on the west side of the building to the north exterior wall.

The co-extrusion process was carried out with various equipment pieces, but the most prominent and unique of these was a Loewy Press that actually pressed all of the fuel components (U core and all of the cladding components) together in one unit. Each N-Reactor fuel element was 26 inches long, weighed approximately 52 pounds and had a tube-in-tube configuration with a coolant channel running down the entire length of the element. Projections also were welded onto each element, as the N-Reactor process tubes were smooth or "ribless." The co-extrusion process provided a better, more uniform bond between core and jacket than had been possible with older methods based on dipping. The new method was beneficial in smoothly cladding the inner and outer tubes.

2.2.7.2 Material Flowsheet

The basic process flow for fuel element preparation for the single pass reactors was to receive and inspect uranium billets from offsite for visual flaws and correct dimensions. Then a sample was irradiated in the 305 Building test reactor. If the reactor tests were acceptable, the lot was accepted and entered the manufacturing process. In the 314 Building, uranium billets were heated in a muffle-type furnace with an interior, inert gas atmosphere. The inert atmosphere, which involved either helium or argon, was used to reduce the oxidation of metal during heating. The uranium was then transferred through a closed passageway to the extrusion press, which also operated in an inert atmosphere. After being extruded, the rods were out-gassed, straightened, and sent to the 313 Building for machining and jacketing. In the 313 Building, uranium fuel rods were machined into fuel cores in lengths of either 4 inches or 8 inches, with 1.3-inch diameters. Known as "slugs," these cores were "canned" or jacketed into finished elements, and then tested and inspected in this building. This jacketed fuel was then sent to the 314 Building for autoclave and radiographic inspection. Figure 2-13 illustrates the fuel fabrication process for the single pass reactors.

Additional fuel fabrication activities were initiated in the early 1960s for the fabrication of N-Reactor fuel. This process, illustrated in Figure 2-14, is significantly different than single pass reactor fuel process. The fuel was larger in diameter, longer, and consisted of two fuel elements, one within the other. It also had a centered annulus that was created by the co-extrusion tube-in-tube process. The two sizes were made, tested and then assembled to form a finished fuel rod.

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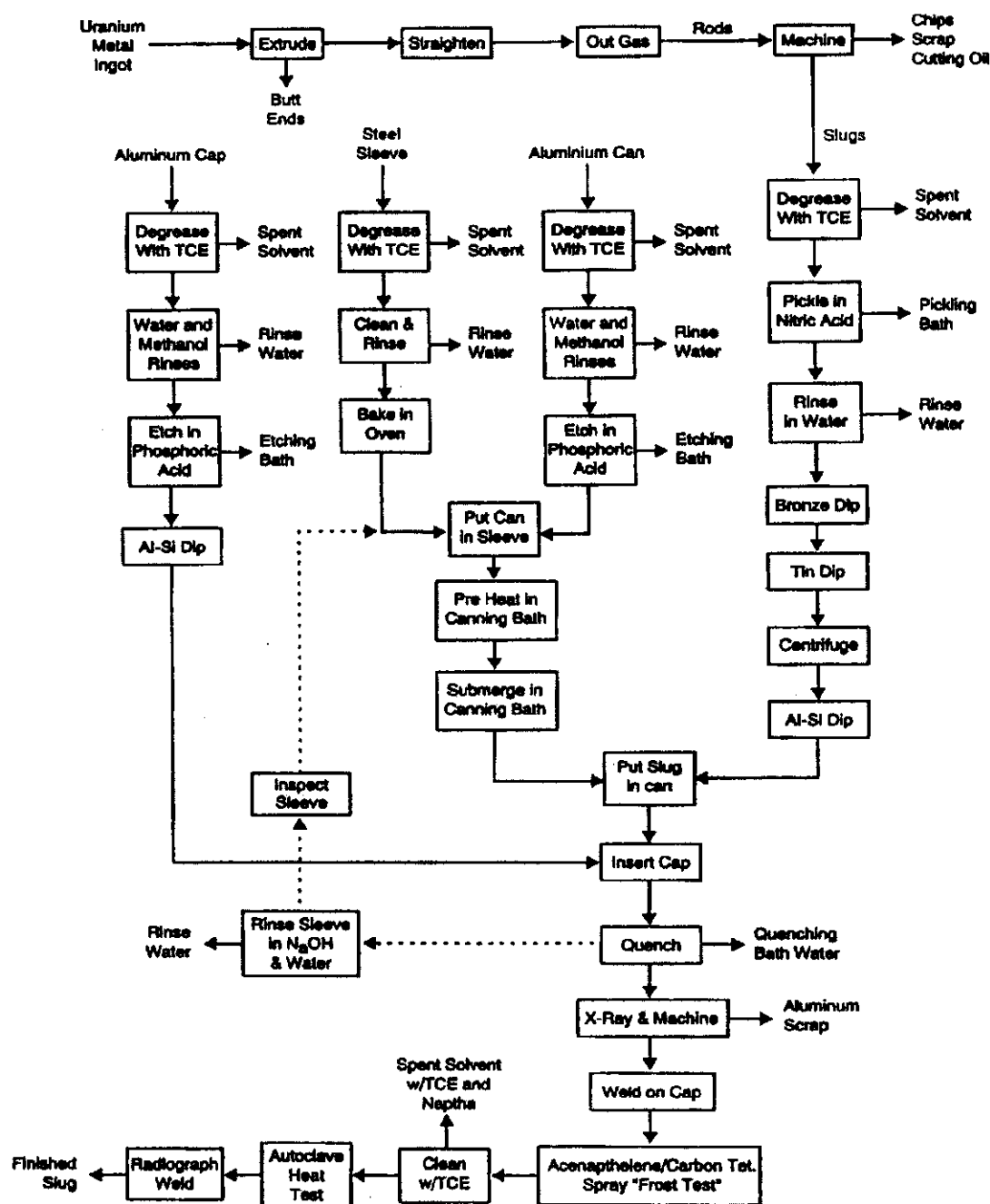


Figure 2-13 Fuel Fabrication Process for Single-Pass Reactors.

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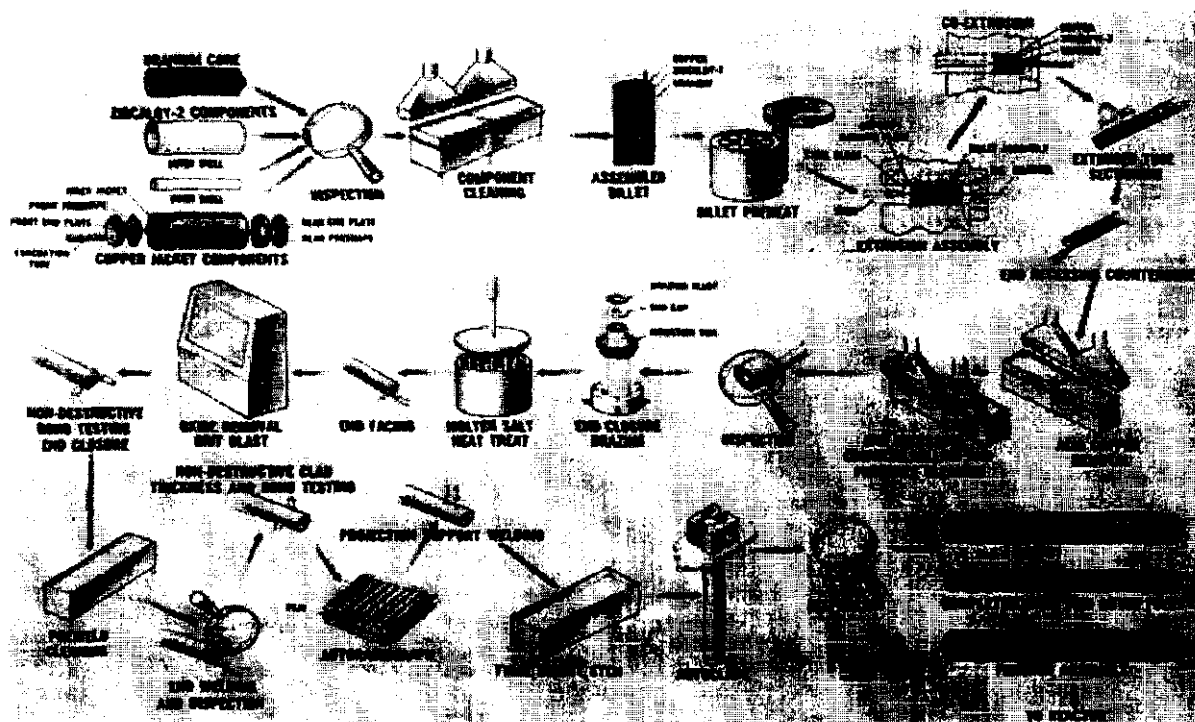


Figure 2-14 N-Reactor Fuel Fabrication Process

More detailed information on past fuel fabrication processes and activities at Hanford can be found in Appendix F.

Over the years, several other ancillary or off-shoot processes have taken place in the 313 and 314 Buildings. Among these have been uranium scrap recovery operations, experimental and/or small-scale fuel making ventures, and waste treatment activities. From its earliest days, concern of the Manhattan Engineer District (MED) about the adequacy of uranium supplies brought strict policies that mandated the reclamation of all possible uranium scraps at federal atomic sites. During the earliest fuel fabrication operations (1943 to mid 1944) at Hanford, difficulties with early fuel canning techniques produced thousands of rejected cores and other scraps, including lathe turnings and metal oxides that formed when canned slugs failed in autoclave tests. In June of 1944, Du Pont reported that "all available space" around the 313 and 314 Buildings was filled with cans of scrap, and the fabrication area fence had to be moved about 30 feet east of the fresh fuel storage building 303-J to allow for more storage space. In addition, beginning with the startup of extrusion press tests in January 1945, extrusion butt ends, oxides, and container residues collected, along with acids from the slug pickling process and from the slug recovery process were generated.

At first, the various types of scrap were shipped to offsite reclamation processing centers. By 1946, the accumulating volume of uranium scraps brought a change in policy at Hanford. This change was the result of scrap storage expense and risks associated with potential fire and security hazards during shipment. Consequently, a

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"chip recovery" operation began in the 314 Building to process scrap. It operated only a few days a month and involved collecting all chips and turnings from machining operations, sorting them, breaking them into small pieces, washing, drying, and then pressing them into briquettes. At first, the briquettes themselves were shipped offsite. In May, however, the MED ordered briquetting to be discontinued due to a number of uranium chip fires within the centrifuging step at other sites.

A "melt plant" was established in the 314 Building in late 1947. In that process, uranium scrap could be recovered as metal for fuel fabrication. This was accomplished by combining uranium tetrafluoride (UF_4 or "green salt") and either calcium chips or magnesium chips, placing this mixture in a dolomite-coated steel vessel, which was then heated until free molten uranium separated from magnesium fluoride or calcium fluoride, and then allowed to cool. The molten uranium settled into large buttons shaped like Derby hats (called "Derbies" by Hanford workers). The Derbies were separated from the slag (calcium fluoride and magnesium fluoride salt) utilizing a jackhammer to break up the slag. These Derbies were then melted in a vacuum furnace, and cast into ingots. These ingots were then rolled into new uranium rods, either offsite or at Hanford, and used to make additional fuel rods.

In the spring of 1946, an additional scrap recovery operation known as the "oxide burner" began on the north side of the 314 Building. All uranium-bearing powder and particulate matter that could be collected from the fuel fabrication facilities, as well as the tailings or settlings from washes and quenches, was burned to convert it to a stable oxide (powder) form. The UO_2 was then collected in 5-gallon containers for shipment offsite [Gerber 1992].

Between 1952 and 1954 both the vacuum furnace melting and the oxide burning processes were phased out and replaced with a process which slurried all the scrap into sodium diuranate, stored it in drums, and returned it to the Feed Materials Processing Center (FMPC), Fernald, Ohio, for recovery [Gerber 1996].

The concentration of any transuranics and fission products in the scrap from fuel fabrication activities would have been the same as that of the uranium metal received at Hanford for fuel fabrication. During our review of historical documentation for this study, we have found no information to suggest that the scrap processing activities would have altered the ratio of impurities to uranium.

2.2.7.3 Feed Specifications

Natural uranium was used for billet fabrication until about 1952 when the use of recycled uranium was phased in. The billets were accepted for use based upon material certification and chemical analysis received from the manufacturer. The scrap material was returned on the basis of the same billet receiving certifications and analysis data since the fuel fabrication process did not make any significant chemical changes.

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2.2.7.4 Product Specifications

Product specifications for the reactor fuel assemblies were based upon conforming to the design requirements of the reactor. Fuel was inspected dimensionally and by radiograph, ultrasonic, and other standard quality measurements. Chemical constituent control was confirmed by supplier certifications. Welds were examined and weld integrity verified.

2.2.7.5 Operating History

The earliest uranium received for the fabrication of reactor fuel arrived at Hanford in October 1943 as extruded rods. The rods were delivered to the Riverland Yards, which were an official part of Hanford and located just east of the Midway power substation and west of the 100-B Reactor Area. Because railroad track had not yet been completed to the 300 Area, the rods were taken by rail to the Construction Camp about 20 miles north of Richland, and then trucked to the 300 Area. Once railroad service to the 300 Area was connected in January 1944, uranium was delivered to the fabrication area by rail.

Newly arrived uranium rods were unpacked and visually inspected (in sample amounts) for cracks and for overall dimensions. A random amount from each lot was taken to the 305 Test Pile Building just west of the 313 Building, and irradiated at a low level to check for warping, cracking, and embrittlement under irradiation. If the sample withstood the process in good form, the entire lot was accepted. Operations began in December 1943. The first uranium fabrication operation at Hanford was machining, in which bare uranium rods were machined to specific core dimensions in the 313 Building. The following month, operators began degreasing the machined cores before inspection, using a commercial product that contained primarily trichloroethylene, Detrex™, a solvent degreaser. Core canning operations actually began in the 313 Building in March 1944.

In the 314 Building, autoclaves for fuel element testing started to operate in July 1944. A scrap recovery process began the following month. Out-gassing and straightening operations started in the 314 Building in September 1944, but Hanford's uranium rods still were being extruded offsite. Beginning in November 1944, uranium was transported to Hanford as billets, which were stored until the extrusion process began to operate in the 314 Building in January 1945. The press testing phase lasted into mid-spring, and then fuel operations commenced. Improved performance ended required shift work in the metal preparation buildings in June 1945, and work proceeded on a straight, 6-day-per-week schedule. From that time until 1948, a complete cycle of metal preparation was conducted at Hanford.

2.2.7.6 Current Status

Fuel and target fabrication in the 300 Area ceased permanently in 1987 with the closure of the N-Reactor. Some of the facilities have been decommissioned. Based on the

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HANFORD SITE HISTORICAL OVERVIEW

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integrated site baseline priorities, cleanup and restoration of the 300 Area presents many challenges. A portion of the 313 building now produces commercial aluminum extrusions. Other buildings are expected to be leased if suitable tenants can be found and after the facilities are decontaminated to guideline levels which allow occupancy by non-rad workers.

2.2.8 Other Uranium Handling Facilities

Several other Hanford facilities were involved in handling depleted uranium. These are discussed below. Much of this work supported reactor research activities at both this and other DOE sites.

2.2.8.1 308 Building Mixed Oxide Fuel Fabrication

The fabrication of mixed oxide (plutonium oxide and uranium oxide) fuel in the 308 building utilized both depleted and natural uranium. Operations were conducted in gloveboxes to protect the workers from inhaling the material.

2.2.8.2 306 Building

Depleted uranium oxide fuel pellets were fabricated in rooms 158 and 159 of this facility. These operations were conducted in open-faced hoods.

2.2.8.3 234-5Z Plutonium Finishing Plant (PFP)

In 1972, the flowsheet and piping in the PFP were modified to allow separation and recovery of plutonium from some plutonium-uranium materials. The separated UNH stream was accumulated and trucked to the PUREX plant. At PUREX, the UNH was offloaded and discarded to the underground waste storage tanks since there was no need for the material.

2.2.8.4 321 Building

Development and testing of a continuous calciner for production calciners to replace the batch kettles at the UO_3 Plant was performed in the 321 Building. The calciner development proved successful and 6 units were installed in 224UA building. About 200,000 pounds of uranium, as UNH, was trucked from the 200 West area to the 321 Building and processed in calciner development activities. The UO_3 product powder was subsequently returned to the UO_3 Plant and blended into the plant's product stream.

2.2.8.5 303-M Uranium Oxide Facility

A small 303-L Building was constructed in 1961 for burning uranium metal scraps to an oxide form that would be suitable for shipment to the FMPC for recovery. Burning was stopped in 1971 due to operating problems. The building was removed in 1976. In

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1983, a new building, 303-M Uranium Oxide Facility, was constructed on the same site. This high bay, 51 ft by 35 ft by 25 ft (high) concrete structure with a one story extension 20 ft by 13.5 ft on the north side was used to convert more than 115 tons of uranium scrap to oxide during its brief operating period of 1984 to 1987 [Gerber 1992]. A drawing of this facility is shown in Figure 2-15.

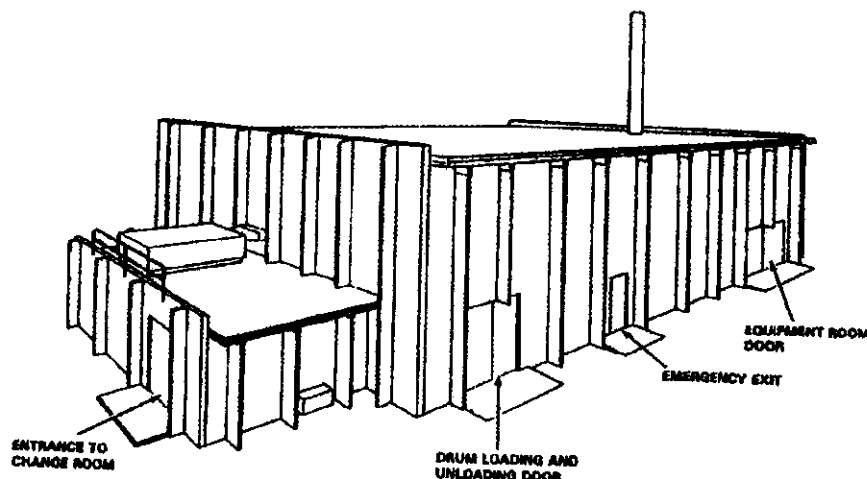


Figure 2-15 303-M Uranium Oxide Facility
[Prezbindowski 1983]

2.3 Activity Summary

Hanford operation started in 1943 to support the defense effort of World War II. After W.W.II, it continued to support the growing defense effort. In doing so, the initial facilities were expanded and new facilities built to meet the congressional mandates for defense. In 1993, all production efforts supporting defense were terminated and the site was redirected to a clean-up mission. This clean-up mission is under way and some facilities have already been decontaminated and either removed or turned over to private industry for new uses.

The first major facilities at Hanford included B-, D-, and F-Reactors, T-Plant, B-Plant, a Fuel Fabrication complex and a plutonium purification plant. In addition to these major facilities, supporting structures, steam plants, housing, water, and personnel support facilities were simultaneously built. The first reactor was started in September 1944. The first batch of plutonium was shipped from the site in February 1945 to Los Alamos where it was converted into the first atomic device.

After the war, it was recognized that the uranium needed to continue and expand plutonium production was in short supply and new sources of uranium would be

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As increased plutonium product was required to meet defense needs, the PUREX process was developed to support the planned production increases and the PUREX separations plant was built, coming online in 1956. PUREX performance permitted the shut down of the REDOX and T-Plant facilities. The PUREX uranium output was also sent to the UO₃ Plant for conversion to oxide. In 1972, PUREX operations were shut down. In 1983, the PUREX plant was reactivated to meet national plutonium requirements created by congressional direction. The UO₃ Plant was also brought back on line to process the UNH output of the PUREX plant. In 1993, all plutonium operations were halted and PUREX Plant and the UO₃ Plants were shutdown to await decommissioning. Figure 2-16 summarizes the operational periods for the major Hanford Plants.

300 Area Fuels Fab.		[REDACTED]		
REDOX		[REDACTED]		
U-Plant		[REDACTED]		
UO ₃ Plant		[REDACTED]		[REDACTED]
PUREX		[REDACTED]		[REDACTED]

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Section 3 Recycled Uranium

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3.0 Recycled Uranium

3.1 Uranium Recycle Description

This chapter is designed to quantitatively define the recycled uranium flows to and from Hanford. The transactions into and out of Hanford will focus on the 300 Area Fuel Fabrication complex of facilities and the UO₃ Plant (224-U Building).

3.1.1 Hanford Key Interfaces for Recycled Uranium

For the Uranium Recycle Project, the Hanford Site is designated as a "Source Site". A source site is viewed as one at which uranium fuel is irradiated, chemically separated, and shipped to offsite locations. These offsite locations are referred to as "Tier 1" sites. Tier 1 sites are those which received recycled uranium directly from the Hanford Site. From the Hanford perspective, uranium transactions offsite are divided into "Major Tier 1" sites and "Minor Tier 1" sites. The distinction is made primarily as it relates to the quantities of recycled uranium shipped and/or received. The Major Tier 1 and Minor Tier 1 sites (from Hanford's perspective) are identified below:

Major Tier 1 Sites:

- Paducah Gaseous Diffusion Plant (GDP), Kentucky
- Fernald, Ohio (FMPC), previously National Lead of Ohio (NLO)
- K-25 Gaseous Diffusion Plant & Y-12 Plant, Oak Ridge, Tennessee

Minor Tier 1 Sites:

- All others (see Appendix B tables for these sites)

Major Tier 1 site locations are shown in Figure 3-1. Figure 3-2 shows the locations of many of both the Major and some of the Minor Tier 1 site locations. Figures 3-3A through Figure 3-3D show the flow of material through the complex for various time periods [DOE/EM-0319 1997]. There have been no reviewed records which indicate transfers of recycled uranium directly to the Portsmouth GDP.

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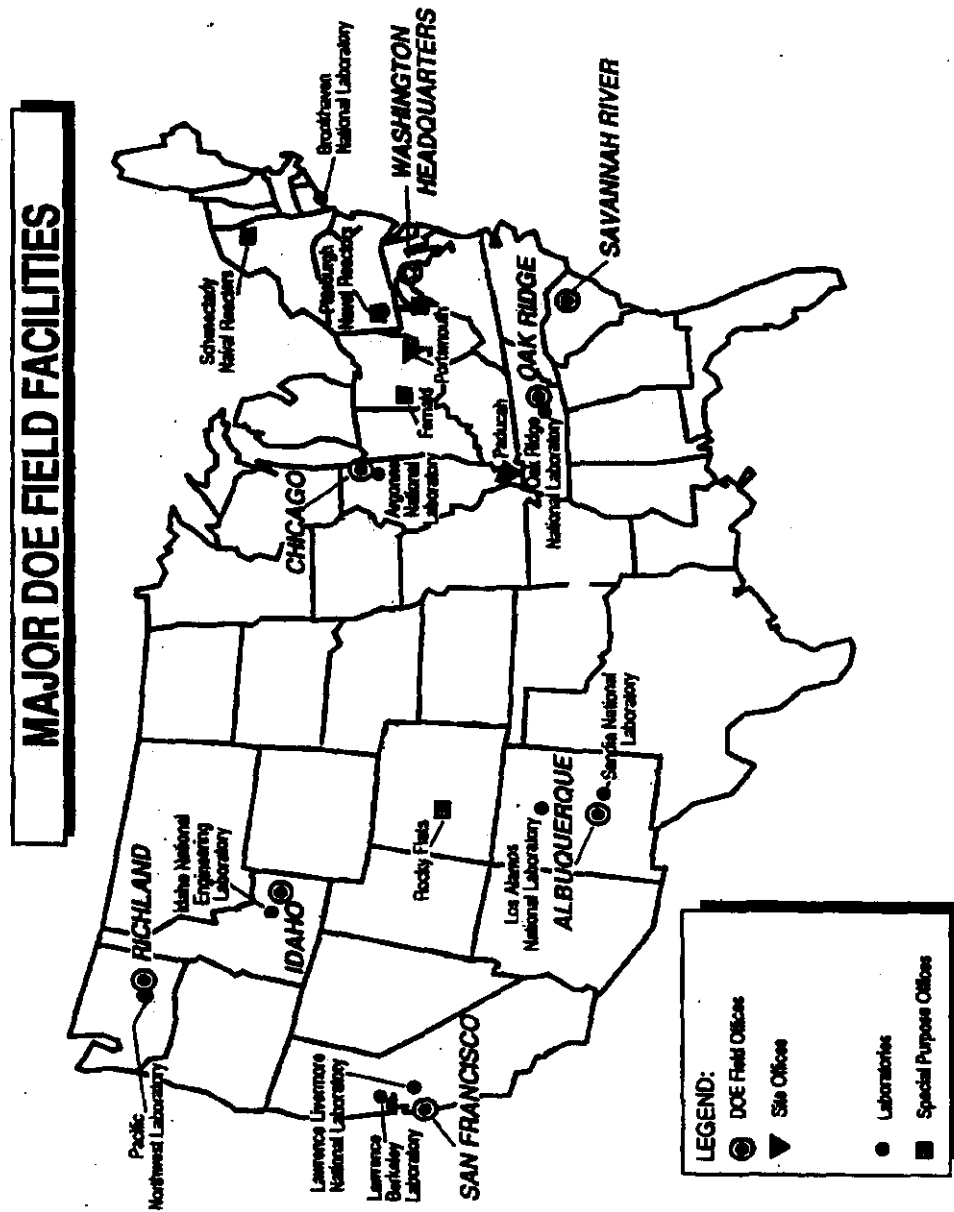


Figure 3-2 Major DOE Field Facilities

Section 3 Recycled Uranium

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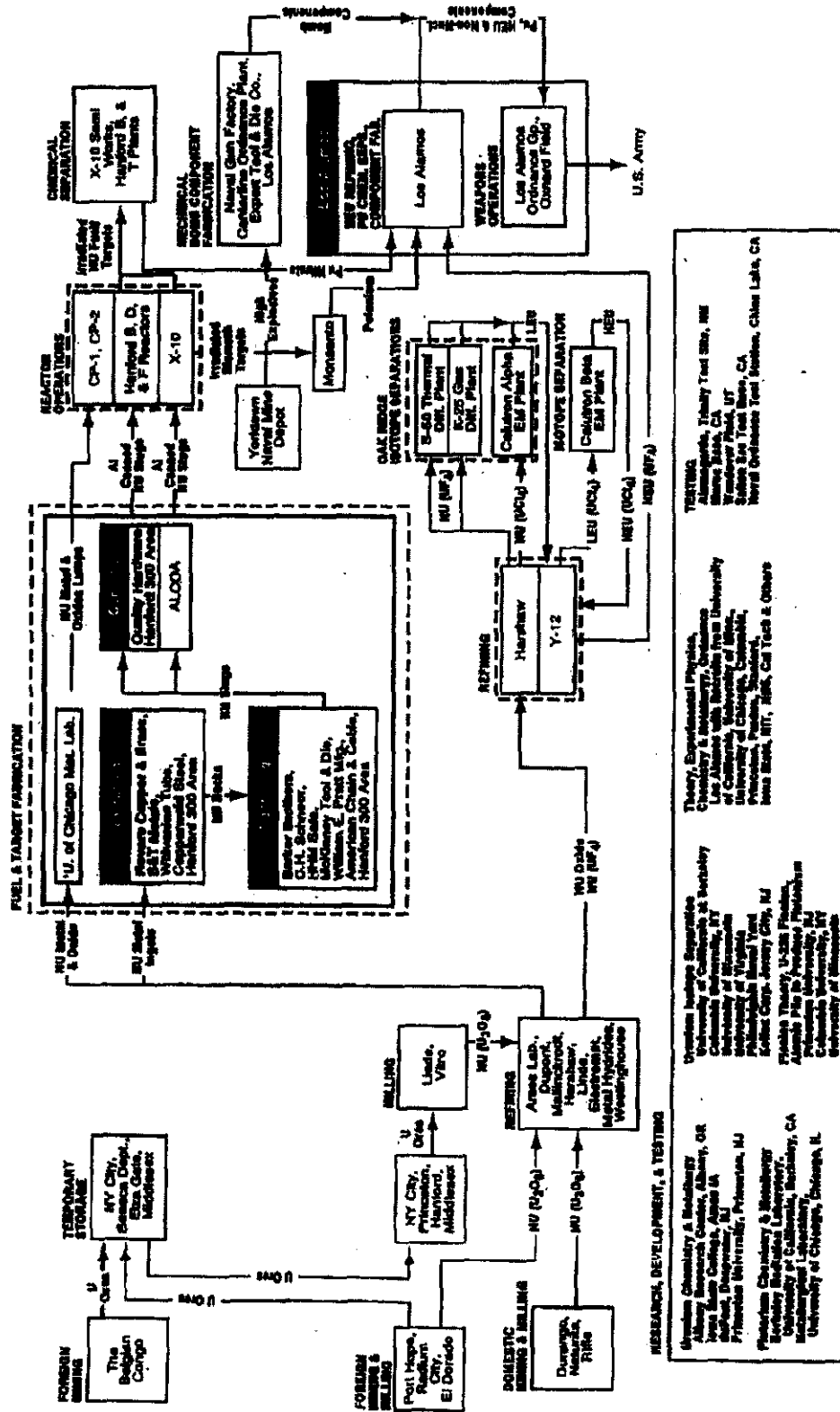


Figure 3-3A Material Flow – Manhattan Engineer District: 1942 – 1946 [DOE/EM-0319 1997]

DOE/RL-2000-43

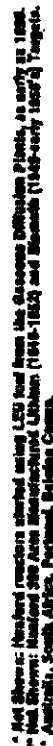


Figure 3-3B Material Flow – Atomic Energy Commission: 1946 – mid 1950s [DOE/EM-0319 1997]

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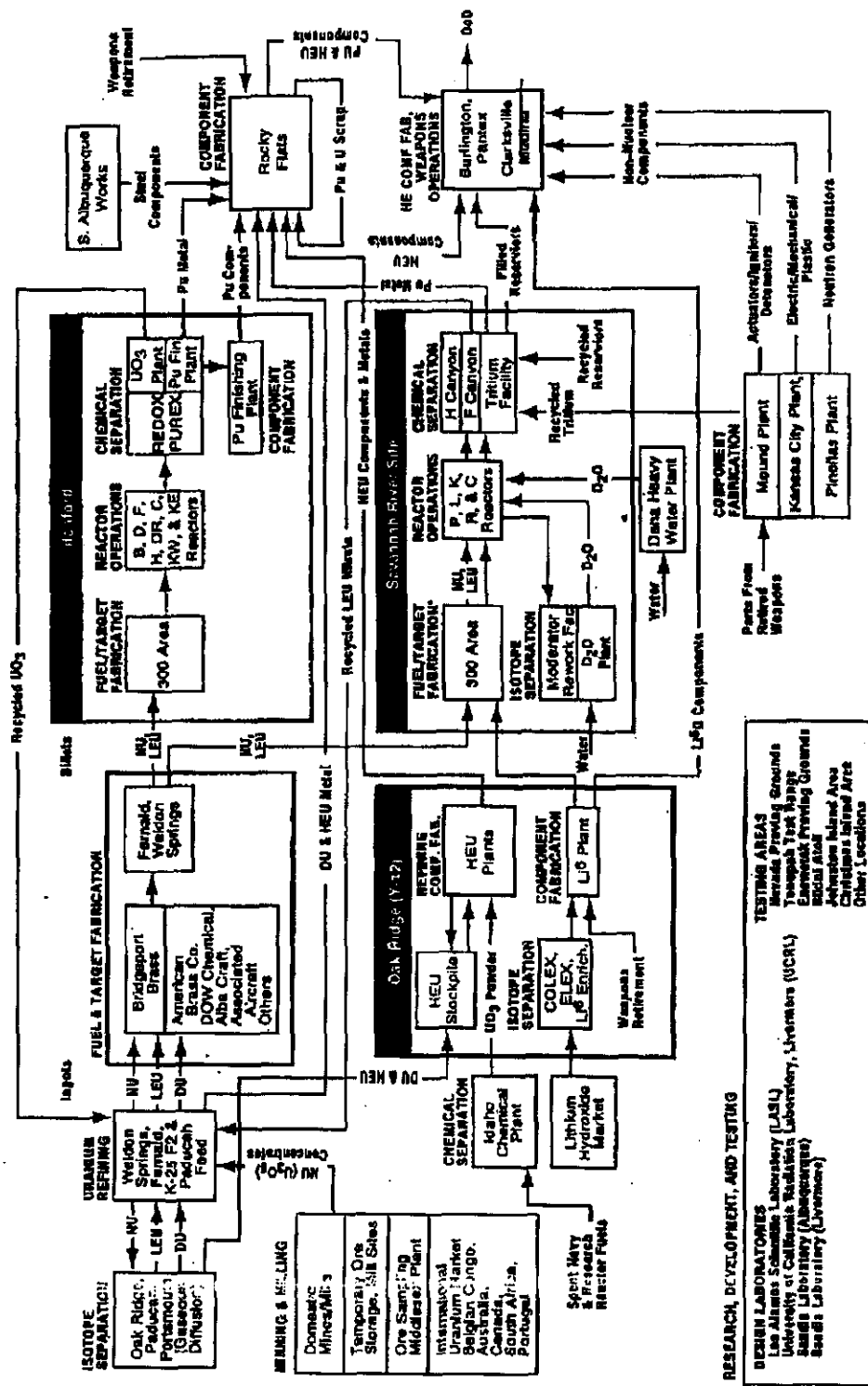


Figure 3-3C Material Flow – Atomic Energy Commission: mid 1950s – mid 1960s [DOE/EM-0319 1997]

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Figure 3-3D Material Flow -- Atomic Energy Commission/Energy Research and Development Agency/Department of Energy: mid 1960s -- late 1980s [DOE/EM-0319 1997]

Section 3

Recycled Uranium

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3.1.2 Beginning of Hanford In-Scope Recycled Uranium Transactions

3.1.2.1 Key Hanford Historical Dates for Recycled Uranium

Beginning of Recycled Uranium Shipments **OUT** of Hanford:

Depleted Uranium:	Mar 1952	UO ₃ product to Oak Ridge K-25
Normal Uranium:	July 1952	Metal scrap returns to offsite fuel reprocessors
Enriched Uranium:	July 1952	Research & development quantities
Enriched Uranium:	Mar 1959	UO ₃ LEU product to Oak Ridge K-25 (Production Channel)

Beginning of Recycled Uranium Receipts **INTO** Hanford:

Depleted Uranium:	July 1952	Hanford UO ₃ heels in returned drums from Oak Ridge K-25
Normal Uranium:	July 1952	Metal billets from offsite fuel fabricators
Enriched Uranium:	July 1952	Research and development quantities
Enriched Uranium:	July 1960	Metal LEU billets from Fernald (Production Channel), at parts per trillion Pu (from cascades)
Enriched Uranium:	Oct 1963	Metal LEU billets from Fernald (Production Channel) (at parts per billion Pu)

3.1.2.2 Production Channel Material Transactions

3.1.2.2.1 Shipments

For UO₃ finished product from the Hanford production channel, the first lot of UO₃ was rail shipped to K-25 on January 25, 1952 and consisted of 8 drums of Lot 001 [Richards 1952b]. The second shipment (Lot 002, 7 drums) was shipped to K-25 on February 11, 1952 [Richards 1952]. Both of these lots were produced from natural uranium and contained no fission products. They were "cold" test runs to validate the UO₃ conversion process. This material was shipped to K-25 to make sure the physical (particle size) and metallic impurities were within Oak Ridge acceptance criteria. As the "cold" UO₃ was examined and found acceptable, Hanford began spiking the feed stream with UNH from irradiated fuel.

Production records indicate shipment of recycled uranium trioxide product to the Oak Ridge K-25 GDP first occurred on March 10, 1952. Examples of the historical transfer documents, with attendant analytical data, are shown in Figures 3-4, 3-5, and 3-6. This March 1952 UO₃ shipment is consistent with Hanford production history indicating UO₃ test runs in January 1952 and full operation in February 1952.

Section 3 Recycled Uranium

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nu - 2 - 1 - 7
HAN-43666

Dr. Frank Burt

- 2 -

March 11, 1952

Component or Property	Reported as	Lot 007	Lot 008	Lot 009
UO ₃	%	97.9	97.7	97.4
H ₂ O	"	0.21	0.23	0.26
NO ₃	%HNO ₃	0.05	0.61	0.55
U ₃ O ₈	%	0.12	0.1	0.11
Na	ppm	3000	3000	2500
FOH	"	1500	1500	1500
Fe	"	700	950	550
Mn	"	20	20	20
Mo	"	50	50	< 50
Cr	"	2	2	5
W	"	< 100	< 100	< 100
Si	"	10	10	50
B	"	< 0.2	< 0.2	< 0.2
S	"	27	8	< 1
Al	"	5000	5000	5000
Particle Size	% thru 80 Mesh	98.2	99.0	97.7
Bulk Density	g./cc.	1.99	2.07	1.75
Surface Area	sq. m./g.	1.3	1.035	1.2
Pu	ppb	< 5	< 5	< 5
f.p. activity, α	"	< 5%	< 5%	< 10%
f.p. activity, β	"	< 70%	< 100%	< 56%
U ₂₃₅	% of U	0.68	0.68	0.67

* %, relative to (beta or gamma) activity of an equal weight of natural uranium

Figure 3-4A (Continued) First Hanford Shipment Containing
Transuranics and Analytical Data
[Richards 1952a]

Section 3 Recycled Uranium

DOE/RL-2000-43

DECLASSIFIED

EW-23846

Box 130263
1849

March 19, 1952

This document contains...
Pages No. of
Copies, Series

Dr. Frank Hurd
Carbide and Carbon Chemicals Corporation
K-25 Plant
Oak Ridge, Tennessee

Dear Dr. Hurd:

UO₂ TRIAL PRODUCTION LOTS O10, O11, O12, and O13

We are shipping by rail (car Milwaukee 1011, GHL AT-26979) lots O10, O11, O12, and O13 of UO₂ prepared from nominal 600 MHD/t material processed through the Redox Plant. This shipment, consisting of 32 drums and 4 boxes, left on March 19 and should arrive on March 24, directed to K-25 Plant, Oak Ridge, att'n F. H. Anderson - J. W. Arendt. Results of analyses of a composite sample of each lot are given below.

Classification cancelled (Change to

UNCLASSIFIED)

By Authority of RLD-CG-5

Section 1.4.3

By CA Bauman-PNL

Verified by W HC-CO 11/14/94
For: JEM:blip

Yours very truly,

R. E. Richards, Manager
Separations Technology Unit
Technical Section, Engineering Dept.

By J. B. Work
J. B. Work

- cc: 1-2 Dr. Frank Hurd, K-25
3 AP Huber, K-25
4 MS Humes, K-25
5 Sylvan Crower, K-25
6-7 Hanford Operations Office - Atomic Energy
Commission - Att'n: DC Springs
8 AB Gruninger - OSI Greager
9 JE Meider - TM Hecoff
10 RS Bell - VR Chapman
11 VD Donibee
12 JB Work - RE Richards
13 FW Woodfield
14
15
16
17
18 Pink
19 Yellow

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11-95

MATERIALS Known

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Figure 3-4B Second Hanford Shipment Containing Transuranics and Analytical Data
[Work 1952]

Section 3 Recycled Uranium

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FEI-23846

Dr. Frank Burt

- 2 -

March 19, 1952

Component or Property	Reported as	Lot 010	Lot 011	Lot 012	Lot 013
UO ₃	%	97.3	97.6	98.0	98.1
H ₂ O	"	0.27	0.24	0.24	0.23
NO ₃	Fe NO ₃	0.65	0.55	0.62	0.54
U ₃ O ₈	%	0.24	0.46	0.35	0.45
Na	ppm	2500	2500	5000	2000
PO ₄	"	< 15	< 15	< 15	150
Fe	"	174	177	133	116
Mn	"	< 10	< 10	< 10	< 10
Mo	"	< 50	< 50	< 50	< 2
Cr	"	1	10	1	< 1
W	"	< 10	< 10	< 10	< 10
Si	"	10	20	50	25
B	"	< 0.2	< 0.2	< 0.2	< 0.2
S	"	7	< 1	< 1	< 1
Al	"	2500	2000	2000	1500
Particle Size	% thru 80 Mesh	95.5	96.4	99.1	80.7
Bulk Density	g./cc.	2.2.	1.69	1.75	2.10
Surface Area	sq. m./g.	1.1	1.15	1.0	1.0
Pu	ppb	< 5	< 1	< 5	< 1
f.p. activity, β	*	< 5%	29%	18%	18%
f.p. activity, δ	*	83%	105%	82%	77%
U ₂₃₅	% of U	0.67	0.66	0.66	0.66

* %, relative to (beta or gamma) activity of an equal weight of natural uranium

Figure 3-4B (continued)
Second Hanford Shipment of UO₃ - Analytical Results
[Work 1952]

Section 3 Recycled Uranium

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<p>ISSUING FILE</p> <p>CIRCULATING COPY RECEIVED MAR 1 1971 REL. TO JO DOCUMENT CONTROL 271-U Bldg. 200 West</p>		<p>TITLE AND AUTHOR</p> <p>URANIUM OXIDE - SOURCE DATA - FY 1971 June, 1971 JG Murphy</p>		<p>WESTINGHOUSE HANFORD COMPANY DECLASSIFICATION REVIEW 1ST REVIEW - DATE 9/12/95 AUTHORITY FBI ADD NAME: RW Hadd Sec SPEC 2ND REVIEW - DATE 12/13/95 AUTHORITY ADD NAME: [Signature]</p>	
<p>IN OR AN</p> <p>OTHER OFFICIAL CLASSIFIED INFORMATION</p> <p>THIS MATERIAL CONTAINS INFORMATION AFFECTING THE NATIONAL DEFENSE OF THE UNITED STATES WITHIN THE MEANING OF THE ESPIONAGE LAWS, TITLE 18, U.S.C. SECS 793 AND 794, THE TRANSMISSION OR REVELATION OF WHICH IN ANY MANNER TO AN UNAUTHORIZED PERSON IS PROHIBITED BY LAW</p>		<p>THIS DOCUMENT MUST BE KEPT UNATTACHED FROM AN UNAUTHORIZED PERSON MAY HAVE ACCESS TO IT. WHEN NOT IN USE, IT MUST BE STORED IN AN APPROVED LOCKED REPOSITORY WITHIN AN APPROVED GUARDED AREA. WHILE IT IS IN YOUR POSSESSION AND UNTIL YOU HAVE RETURNED IT TO YOUR CLASSIFIED DOCUMENT RESPONSIBILITY STATION, IT IS YOUR RESPONSIBILITY TO PROTECT IT AND ITS CONTENTS WITHIN THE LIMITS OF THIS PROJECT FROM ANY UNAUTHORIZED PERSON. ITS TRANSMITTAL TO, AND STORAGE AT YOUR PLACE OF RESIDENCE IS PROHIBITED. IT MAY BE DUPLICATED ONLY IN ACCORDANCE WITH EXISTING SECURITY REGULATIONS. ALL PERSONS READING THIS DOCUMENT ARE REQUESTED TO SIGN IN THE SPACE BELOW.</p>			
<p>ROUTE TO:</p> <p>JG Murphy</p>		<p>FILE ROUTE</p> <p>271-U Bldg. MAR 10 1971</p>		<p>SIGNATURE AND DATE</p> <p>[Signature] 7/1</p>	
<p>[Redacted]</p>		<p>RECORD COPY</p>			
<p>[Redacted]</p>		<p>DECLASSIFIED RECORDS SCHEDULE DRS 14.6A</p>			
<p>64-5100-150 (8-67) REPLACES STANDARD 5050</p>		<p>(CLASSIFICATION)</p>			

Figure 3-5 Example of Historical Transfer Forms-Cover Page

DOE/RL-2000-43

ALBANY, N.Y., June 26, 1948. — JAMES W. HANCOCK, JR., 40, 1000 E. 10TH ST., ALBANY, N.Y., HAS BEEN CONVICTED OF A FEDERAL CRIME AND IS BEING SENTENCED TO PRISON FOR TWO YEARS.

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[REDACTED]

208

(CLASSIFICATION)

UO₃ - PRODUCT ACCEPTANCE

LOT NUMBER 1-5-2 & 1-5-3			
TRANSFER SERIES HVA-FYA-133	GROSS WEIGHT	Lot-1-5-2 28 210	
		Lot-1-5-3 56 234	Lbs.
SHIPPED TO Union Carbide Corporation	TARE WEIGHT	3 820	
		7 570	Lbs.
GBL NUMBER E-8607-725	NET WEIGHT	24 390	
		48 664	Lbs.
MATERIAL Depleted UO ₃	DATE SHIPPED	6-1-71	
NO. OF CONTAINERS Lot-1-5-2 2 cont. Lot-1-5-3 4 cont.	NET SAMPLE WEIGHT	Lot-1-5-2 3 Kgs. Shipped 5-28-71	
CAN NUMBER UP-508825	AVERAGE PERCENT U	82.04	
		81.90	
SEAL NUMBERS HDE-6803 & 6804	AVERAGE PERCENT U-235	0.658 Est.	
SUBMITTED BY: <u>G.B. Kuklinski</u> ATLANTIC RICHFIELD HANFORD COMPANY		DATE 6-2-71	
ACCEPTED BY: <u>Al M. Langston</u> U.S. ATOMIC ENERGY COMMISSION		DATE 6/7/71	
Shipment Pending on Lot-1-5-3			
<div style="text-align: center; margin-top: 20px;"> <div style="border: 1px solid black; display: inline-block; padding: 5px 20px;">[REDACTED]</div> </div>			
<div style="text-align: center;"> <div style="border: 1px solid black; display: inline-block; padding: 2px 10px;">[REDACTED]</div> </div>		<p>THIS DOCUMENT CONSISTS OF 1 PAGES, NO. 1 OF 2 COPIES. SERIES [REDACTED]</p>	

HANFORD CATEGORY C-48

Figure 3-6 Example of Historical Product Acceptance Form
Hanford Depleted UO₃ to Paducah (circa 1971)

Section 3

Recycled Uranium

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In March 1959, General Electric was authorized by the AEC to begin routine shipments of low-enriched (0.94% ^{235}U before irradiation) UO_3 to the K-25 facilities in Oak Ridge [Gifford 1959]. Hanford LEU UO_3 shipments began soon thereafter. From this March approval-to-ship to the end of June 1959, Hanford produced and shipped approximately 288 MTU of the low-enriched (0.85%) UO_3 to Oak Ridge. Although the K-25 facility was the first recipient of Hanford recycled uranium, the vast majority of the UO_3 product was shipped to the Paducah site beginning in FY 1954 through FY 1972.

The third major recipient of Hanford recycled UO_3 was the Fernald site, which began receiving research quantities of depleted UO_3 in FY 1953. Although Fernald received small quantities of Hanford depleted UO_3 , they were the major recipient of Hanford low-enriched recycled UO_3 beginning in the early 1960s through March 1989. These shipments originated from the Hanford chemical processing contractors (GE, Isochem, ARHO, RHO, WHC). Some small quantities of Hanford UO_3 which did not meet K-25 acceptance criteria for non-radioactive chemical purity were sent to Harshaw for purification. The majority of Hanford UO_3 shipped from Hanford to the K-25 plant was later shipped from K-25 to Paducah.

3.1.2.2.2 Receipts

Beginning in the late 1940s, Hanford received uranium product to support fuel fabrication activities. Metal feedstock was received from Mallinckrodt (St Louis and Weldon Spring, Missouri), and Simonds. Fuel samples were exchanged with many sites as this new technology was rapidly growing. With the Fernald Plant coming on line in March 1953, an increasing quantity of uranium was received and shipped between Hanford and Fernald. Hanford receipt of recycled uranium is assumed to begin in July 1952 (FY 1953) as material shipped from Hanford offsite between March through June 1952 could not have reasonably been received, reprocessed, and returned as feedstock from offsite until that time. In discussions with Fernald staff, normal (recycled) uranium metal feedstock initially received at Hanford could be expected to have contained only parts-per-trillion quantities of plutonium. Further discussions concerning the Hanford receipts are detailed in Section 3.2. Figure 3-7 (based on a 1949 document) shows the flow of uranium received into Hanford's 300 Area Fuel Fabrication facilities.

3.1.3 Out-of-Scope Uranium Transactions

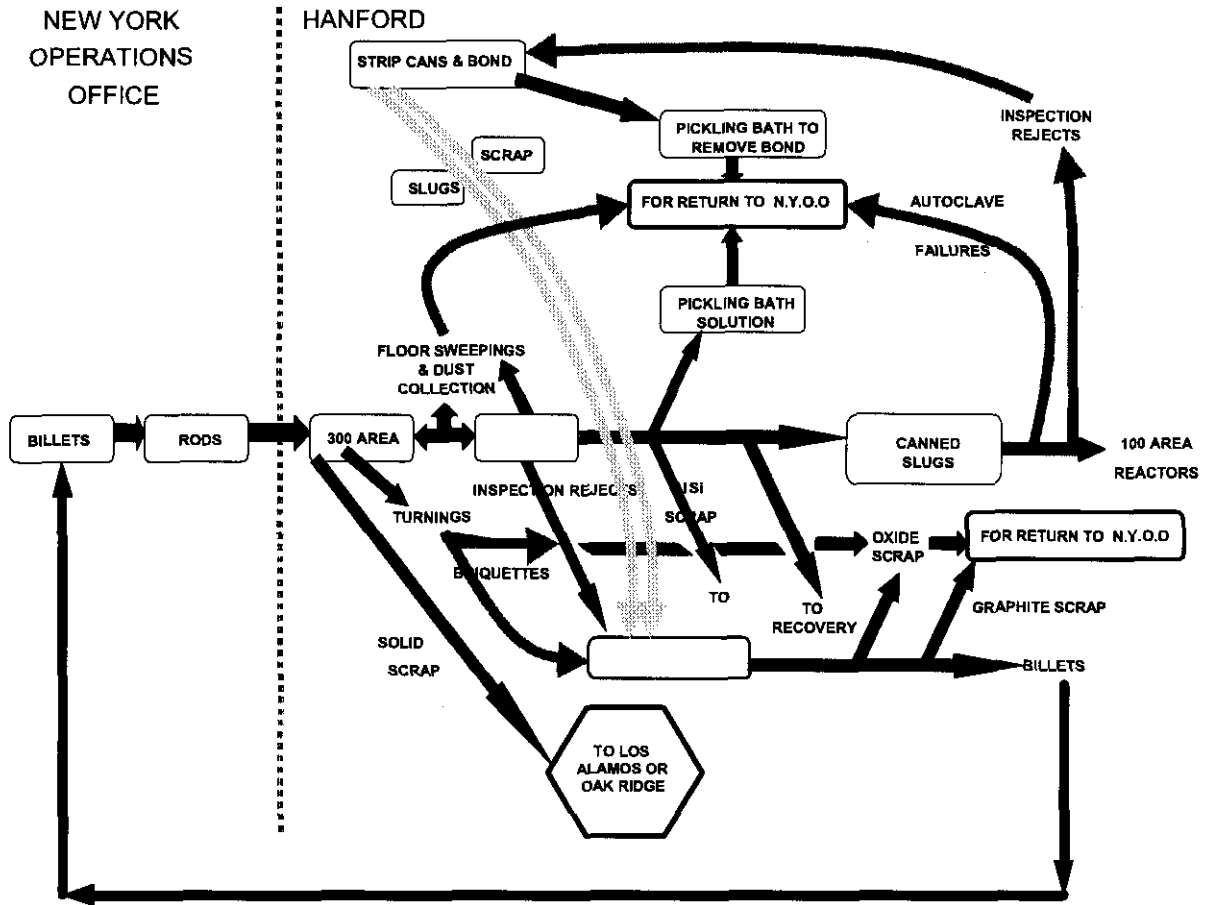
3.1.3.1 Hanford Production Channel

Prior to March 1952, uranium shipments were confined to natural uranium scrap from Hanford's 300 Area Fuel Fabrication activities or metallurgical and process research involving UNH solutions. Natural uranium metal rods were received, principally from the New York Operations Office contractors, and processed at the 300 Area. The unirradiated scrap generated, in various forms, was sent offsite for reclamation. The finished fuel, termed "slugs" were "canned" and sent to the Hanford reactors for subsequent irradiation. Significant effort was made at Hanford in the early 1950s to reclaim and reuse as much of the generated uranium scrap due to the shortage of

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feedstock within the production complex. The fuel fabrication process had no input points at which transuranics could be introduced into the unirradiated fuel manufacturing process. There has been no evidence of any transuranic contaminants being introduced into the fuel within the Hanford manufacturing process.



**Figure 3-7 1949 Schematic Diagram Showing Uranium Flow in 300 Area
(based on HAN-25257, dated May 25, 1949)**

3.1.3.2 Out-of-Scope Research and Development Programs

As the development for increased uranium fuel productivity and chemical integrity continued during the late 1940s and early 1950s, small amounts of uranium were diverted from the production channels for research and development. The three areas of R&D were 1) Exponential Pile Program; 2) Fuel Development Metallurgy; and 3) Separations Technology. One such research program, referred to as the Pile Enrichment program, involved transfers of unirradiated slugs between the Y-12 Plant

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and Hanford. Hanford received the bare slugs from the Y-12 Plant, canned them, and returned slugs, scrap metal, and reject slugs to Y-12. There is no indication that these slugs contained recycled uranium, and are therefore considered out-of-scope transactions.

A subsequent part of the R&D program sent irradiated slugs to the Idaho Chemical Processing Plant (ICPP). These J-1 slugs were also irradiated at H reactor and the J-2 slugs at C reactor. The "C" slugs were irradiated at C and H reactor. As the ICPP came on line, shipments of these "J" irradiated slugs began in late calendar year 1951 and were reported in a 1952 Material Balance Report, FTS-953 [Donihee 1952]. As spent fuel, the irradiated slugs sent to Idaho are considered out-of-scope for this project.

Another mid-1960 AEC research program, termed the Plutonium Credit Activity, involved shipment of Hanford irradiated fuel to Nuclear Fuel Services (NFS) in West Valley, New York [DOE 1999]. Uranium contained in this spent fuel is also considered out-of-scope for this study.

3.1.3.3 Recycled Uranium Timeframe Summary

Summaries of recycled uranium transfers at Hanford have been separated into two distinct timeframes. The period from January 1952 through June 1970 (FY 1970) represents the initiation of Hanford processing of recycled uranium from one or more separation plants. (In 1967, REDOX (S-Plant) shut down.) The second period from July 1970 through the present (March 1999) represents a period in which the PUREX plant (when operating) was the sole separation plant for Hanford's Defense missions. This later period is also one in which Hanford supported multiple non-defense missions, such as the Fast Flux Test Facility, under multiple Hanford contractors.

Quantities of uranium shipped and received are presented in Sections 3.2 and 3.3 and further detailed in Appendix B.

3.1.4 Data Presentation – Isolation of Specific Timeframes

This narrative section is prepared to explain the Hanford Recycled Uranium Project team's approach to quantitatively define recycled uranium materials that were shipped into and out of the Hanford Site since its inception in 1943 until March 30, 1999. To simplify reporting, Hanford shipments and receipts include the aggregate of the fuel fabrication/reactor operations contractors (Douglas United, UNI) and the chemical processing contractors (Isochem, ARHO, Rockwell, Westinghouse, Fluor) after contractor turnover from General Electric (GE, 1965-66 turnover). The Pacific Northwest National Laboratory (PNNL) and its predecessors offsite shipments and receipts are addressed separately.

In an effort to simplify the data investigation, the team chose to separate the Hanford Site uranium transactions to correspond to the following four timeframes:

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- Late 1940s – December 31, 1951: Hanford Site external shipments and receipts from December 1947-December 31, 1951 encompasses the General Electric Company (GE), which solely operated the fuel fabrication, reactors, and chemical separations plants. This first timeframe was isolated to define a demarcation between In-Scope and Out-of-Scope uranium transactions. All transactions within this timeframe have been evaluated as Out-of-Scope to this project. These transactions, detailed in Section 3.2 and 3.3, were almost exclusively natural uranium product and scrap transfers between the New York Operations Office (NYOO) and its contractors and Hanford's Fuel Fabrication facilities.
- January 1, 1952 – June 30, 1965: This timeframe represents the beginnings of Hanford transactions involving recycled uranium under a single GE Company contractor. This period also represents a high production timeframe. As the research for safer and more efficient plutonium production continued, more offsite facilities become recipients and suppliers for recycled uranium into and out of Hanford. In the early 1950s, the major NYOO contractors were replaced primarily by the Fernald and Weldon Spring (Mallinckrodt) facilities as the major suppliers of Hanford metal feedstock and recyclers of Hanford scrap.
- July 1, 1965 – June 30, 1970: This timeframe represents a transitional period of Hanford contractor turnover from the GE Company to multiple contractors and the beginnings of implementation of a DOE-wide Nuclear Materials Management and Safeguards System (NMMSS). PNL, assumed the management of Hanford Laboratories in 1965 as an independent research entity from Hanford Operations.
- July 1, 1970 – March 30, 1999: This timeframe includes the period when the PUREX Plant became the sole producer of UNH for Hanford. The NMMSS MC&A system became operational (complex-wide). Recycled uranium transactions between Hanford and Paducah and Oak Ridge were minimal, and the vast majority of transactions for Hanford were with Fernald (NLO, FMPC, FEMP) and Reactive Metals Incorporated (RMI, Ashtabula Extrusion Plant).

3.1.5 Hanford Historical Timeline References

In tracing the historical transfers, the key activities and timeframes listed below were identified as potentially significant for the purposes of this study. (A more complete Hanford historical timeline of events is provided in Appendix H.)

- Events Related to Hanford:

1950:	Paducah Gaseous Diffusion Plant sited
1951:	Savannah River Plant sited
1951:	Fernald Feed Materials Production Plant (Ohio) sited
1952:	Fernald production begins
1953:	Paducah GDP becomes operational

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- Hanford Contractor timeline:

December 21, 1942: Du Pont signed to construct/operate atomic plants

September 1, 1946: General Electric Company (GE) assumes control as overall Site Contractor

1965 to 1966: GE replaced by multiple contractors

September 1965 - 1973 **Fuel Fabrication & Reactor Operation:**
Douglas United Nuclear (DUN-joint venture subsidiary of Douglas Aircraft Co. and United Nuclear Corp.)

1973-1979 United Nuclear Industries

1979-1987 United Nuclear Corporation (UNC)

1987-1996 Westinghouse Hanford Operations (WHC)

October 1996 - Current Fluor Hanford Incorporated (FHI)

Chemical Separations, Processing & Production
January 1966 - September 1967 Isochem (joint venture subsidiary of U.S. Rubber Co. and Martin Marietta Corp.)

September 1967 - October 1967 Atlantic Richfield Hanford Company, chemical processing operations

October 1977 - July 1987 Rockwell Hanford Company, chemical processing operations

July 1977 - October 1996 Westinghouse Hanford Operations, reactor operations and chemical processing

October 1996 - Current Fluor Hanford Incorporated (FHI)

Research & Environmental Monitoring
January 1965 - 1977 Battelle Memorial Institute (BNWL) (became PNL)

1977 - Current Pacific Northwest Laboratory (PNL)(became PNNL)

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- Government Agencies Having Control of Hanford Site:

1943 - 1946	U. S. Army, Manhattan Engineer District
1947 - 1974	Atomic Energy Commission (AEC)
1/1/75 - 9/30/77	Energy Research and Development Administration (ERDA)
10/1/77 - Current	U. S. Department of Energy (DOE)

3.1.6 Key Dates/Assumptions for Uranium Transactions

May 5, 1950:	First shipment of unirradiated EU "J" slugs to Y-12
January 1952:	First recorded shipments of irradiated EU slugs to Idaho ICPP
January 1952:	Depleted UO_3 product was shipped (no fission products)
March 10, 1952:	First recorded shipment of UO_3 product to K-25 GDP with fission products
July 1952:	First assumed return of recycled uranium into Hanford
July 1958	Scheduled start of enriched UNH input into UO_3 Plant [Gustafson 1957]

March-June, 1959: First production and shipment of enriched UO_3 to K-25

3.1.6.1 Beginning Shipment of Recycled Depleted Uranium Trioxide (UO_3)

As previously noted, the first shipment of recycled UO_3 produced at Hanford was shipped to Oak Ridge, Tennessee on March 10, 1952. Trial Production Lots 007, 008, and 009 were prepared from nominal 600 MWD/t material processed through the REDOX Plant. This initial truck shipment consisted of 24 drums and was sent to the K-25 Plant. The analytical results of composite samples for each lot were also provided [Richards 1952] and are shown in Figure 3-4. Further discussion of Hanford analytical data and product quality is detailed in Section 4.0.

3.1.6.2 Initial Shipments of Recycled Low-Enriched Uranium Trioxide (UO_3)

The first shipments of low-enriched (0.8 -0.9% ^{235}U) UO_3 to Oak Ridge were approved by the AEC on March 3, 1959. Shipments were initially to be made to the K-25 Facility [Gifford 1959].

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- Enriched Uranium: July 1952 research and development quantities
- Enriched Uranium: July 1960 metal LEU billets from Fernald from production channel (Pu in parts per trillion U) (from cascades)
- Enriched Uranium: October 1963 metal LEU billets from Fernald (Pu in parts per billion U)

3.2.3 Receipts Prior to July 1952 (Out-of-Scope)

In the late 1940s, Hanford receipts were natural uranium billets and rods from various metal fabricators under the management of the New York Operations Office (NYOO). Many of these same contractors were the recipients of Hanford shipments of scrap generated during the fuel fabrication activities and are detailed in Section 3.3. In the late 1940s and early 1950s, a majority of the Hanford billets were supplied by Mallinckrodt Chemical Works (MCW) and originated from three types of MCW cast ingots which included 1) ingots cast from natural uranium derbies; and 2) ingots recast from ingot croppings; and 3) ingots recast from reject slugs, rod ends, and rolling mill scrap [Greninger 1953]. Any uranium received at Hanford before July 1952 would not have contained reactor-produced fission products or radionuclides. There would have been no ^{236}U in these uranium receipts but would have contained the same distribution of uranium isotopes as present in natural or enriched uranium from a GDP cascade.

3.2.4 Beginning Receipts of Recycled Uranium at Hanford

The beginning receipts of metal feed stock with trace transuranics into the 300 Area is assumed to begin in July 1952 (FY 1953). This assumption is based on the logic that transuranics in the March 1952 UO_3 shipped offsite, could not have reasonably been processed and re-introduced into the returning metal billets until July 1952. Throughout the 1950s, Hanford continued to receive substantial metal feedstock from the NYOO contractors (Mallinckrodt Chemical Works, Simonds Saw & Steel, etc.). The largest shipper of metal feedstock during the mid-1950s and until the 1980s was the National Lead of Ohio Company (NLO) plant in Fernald, Ohio. NLO was renamed the Feed Materials Production Center (FMPC) in the 1950s. FMPC is now managed by the Westinghouse Materials Company of Ohio. FMPC is a Major Tier 1 site, being both the recipient of Hanford fuel fabrication scrap and UO_3 product and the supplier of metal billet feedstock. FMPC produced, via plants 6 and 9, normal and low-enriched ingots that were finished into billets at Reactive Metals Inc. (RMI) and shipped to Hanford's 300 Area. Informal discussions with Fernald staff, indicate that there were no input points at RMI that could introduce transuranic contaminants into the billets shipped to Hanford. RMI was essentially a heat treating and extruding facility.

3.2.5 Quantities of Recycled Uranium Received from July 1952-March 30, 1999

The summary of in-scope recycled uranium received at the Hanford contractor(s) starting in July 1952 until March 31, 1999 totaled approximately 109,200 metric tons. Of

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this total, approximately 85% was received from the three Major Tier 1 sites (~92,800 MTU). Yearly summaries for these three Major Tier 1 sites are detailed in Tables 3-1, 3-2, and 3-3. Summary fiscal year tables for all receipts by Hanford contractors are provided in Appendix B, Tables 3.2.1 through 3.2.8. These Appendix tables are divided into distinct timeframes to simplify transactions associated with the Hanford contractor turnover which occurred continually from 1965 onward. Summarized in Table 3-1 below is the total recycled uranium received from offsite sources at Hanford.

Table 3-1 Total Recycled Uranium Received from Offsite Sources

Timeframe:	MTUs Received: All Offsite Sources:	MTUs Rec'd Major Tier 1:	MTUs Rec'd Minor Tier 1:
FY 1953-FY 1965	77,603.7	72,869.5	4,734.2
FY 1966-FY 1970	19,119.5	19,109.6	9.9
FY 1971-3/31/99	12,420.4	788.0	11,632.4*
	109,143.6	92,767.1	16,376.5

*The majority of post FY 1971 receipts were from RMI Extrusion Plant (FTA) which supplied the Hanford fuel fabricator (United Nuclear, HXA).

3.2.6 Hanford Receipts of Recycled Uranium from Paducah

Hanford received residual UO_3 in returned containers from FY 1954 through FY 1964. The receipts from Paducah are detailed in Table 3-2.

3.2.7 Hanford Receipts of Recycled Uranium from Fernald and RMI (Ashtabula)

Hanford received metal billets from Fernald and Reactive Metals Incorporated (RMI), Ashtabula. Hanford also received residual UO_3 in returned shipping containers from Fernald. In 1983, incoming materials into the 300 Area were primarily 0.95% and 1.25% ^{235}U billets from RMI in Ashtabula, Ohio. The receiving rate was nominally 4 ½ loads per month at 18 metric tons uranium per load [Heaberlin 1983]. The receipts from Fernald are summarized in Table 3-3.

3.2.8 Hanford Receipts of Recycled Uranium from Oak Ridge

Hanford received relatively small quantities of UO_3 as heels in returned shipping containers and uranium metal for research programs. These receipts are summarized in Table 3-4.

3.3 Recycle Uranium Shipments

3.3.1 Recycled Uranium Streams Shipped Offsite:

Two major recycle uranium streams were shipped offsite from Hanford's beginnings until March 1999. The first of the two major streams was byproduct from the fuel

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fabrication activities in the 300 Areas. The other major stream was the UO_3 product produced at the UO_3 Plant in the 200 West Area.

Hanford UO_3 shipped after March 10, 1952 contained recycled uranium. The Major Tier 1 sites of Paducah, Fernald, and Oak Ridge received the vast majority of Hanford recycled uranium. Paducah received the majority for these three sites with approximately 74,500 MTU shipped out of Hanford from FY 1952 through FY 1973. After FY 1973, the majority of recycled uranium was sent to Fernald. Table 3-5 provides a brief summary of recycled uranium shipments from Hanford. Tables 3-6, 3-7, and 3-8 show these shipments to the Major Tier 1 sites in detail. Appendix B Tables 3.3.1 through 3.3.8 show the details of Hanford shipments to all off-site locations.

Table 3-5 Summary of Recycled Uranium Shipments from Hanford

Timeframe:	MTUs Shipped All Offsite Sites	MTUs Shipped Major Tier 1	MTUs Shipped Minor Tier 1
March 1952-FY65	67,740.4	64,593.0	3,147.4
FY 1966-FY 1970	28,292.4	28,289.6	2.8
FY 1971-3/31/99	13,759.6	11,263.6	2,496.0
Recycle Total	109,792.4	104,146.2	5,646.2

3.3.2 Uranium Shipments from 300 Area Fuel Fabrication Activities

Specific uranium forms being removed from the fuel fabrication shops included reject metal rods, uranium oxide, "eggs", "slugs", metallic chips and fines, and floor sweepings. As much uranium was recovered as was possible in the early years due to shortages in uranium feedstock supply. "Eggs" were a term for metal samples cut off from the ends of newly arrived billets and tested for impurities before the billets were fabricated into fuel elements. "Slugs" were an early term for uranium fuel elements in the form of short cylinders clad or encased in corrosion-resistant metals. The 1949 schematic in Figure 3-4 shows the various flows of the generated scrap from the fuel fabrication activities. The four major NYOO sites receiving Hanford scrap were Mallinckrodt Chemical Works (Simonds Saw & Steel (Lockport, New York), Vitro Manufacturing (Cannonburg, Pennsylvania), and Harshaw Chemical (Cleveland, Ohio)). Simonds performed metal rolling of the uranium billets, Mallinckrodt reprocessed sweepings, metal solids, "eggs", and rejected slugs. Vitro reprocessed Hersey Bag Filters (from UO_3 plant) and miscellaneous scrap oxides. Uranium billets and metal turnings were also shipped to National Lead of Ohio (NLO).

Beginning in 1952, Aluminum-Silicon (Al-Si) alloy scrap (from the fuel Fabrication process) was also shipped to the U.S. Bureau of Mines (Albany, Oregon) because that facility had developed a method for recovering the tin. The tin crystals contained uranium.

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In the late 1940s, as part of Uranium Sample Exchange Programs, Hanford shipped metal billets to Mallinckrodt Chemical Works (St. Louis) for metallic impurity comparisons [Rebol 1949].

Until the end of June 1952, all Hanford outbound shipments were of unirradiated natural uranium scrap or research materials generated at the 300 Area Fuel Fabrication or Hanford research laboratories. The primary recipients for the reprocessing of this scrap were Mallinckrodt Chemical Works, Simonds Saw & Steel Company, Vulcan Crucible Steel, Joslyn Manufacturing, and Vitro Manufacturing. The majority of the receipt sites were under the management of AEC's New York Operations Office (NYOO). As the metallurgical and chemical refinements to the Hanford fuel cycle continued, small quantities of unirradiated natural uranium were also sent to various laboratories for research. Shipments to the New York contractors was phased out in the early 1950s as the Oak Ridge-managed plants at Oak Ridge, Tennessee and Fernald, Ohio became the primary recipients of the fuel fabrication scrap. All production channel shipments of natural uranium from the late 1940s through June 1952 are therefore assumed to be out-of-scope for this report.

For the purposes of this project, it was assumed that offsite scrap shipments of recycled uranium from fuel fabrication activities began in July 1952 (FY 1953). This is based on the assumption that transuranics from UO_3 , or within irradiated slugs shipped offsite, could not have been processed and re-introduced into the returning metal billets until July 1952.

In the 1980's, all the Fuel Fabrication scrap was sent to National Lead of Ohio (NLO). Scrap forms included sludges, fines, and burned oxide (began in 1984). Approximately 181 MTU of 0.95% and 26 MTU of 1.25% as scrap was forecasted to be generated per year. A scrap generation rate of 21% of input was forecasted [Heaberlin 1983].

3.3.3 Hanford Shipments of Recycled Uranium in Trioxide Product

3.3.3.1 UO_3 Finished Product

For UO_3 finished product, the first shipment of UO_3 was rail shipped to K-25 on January 25, 1952 and consisted of 8 drums of Lot 001 [Richards 1952b]. The second shipment (Lot 002, 7 drums) to K-25 was shipped on February 11, 1952 [Richards 1952]. Both of these lots were produced from natural uranium and contained no fission products. They were "cold" test runs to validate the UO_3 conversion process. This material was shipped to K-25 to make sure the physical (particle size) and metallic impurities were within Oak Ridge acceptance criteria. As the "cold" UO_3 was examined and found acceptable, Hanford began spiking the feed stream with UNH derived from irradiated fuel.

3.3.3.2 Introduction of Fission Products

The introduction of fission products into the UO_3 product is indicated in production records that show a March 10, 1952 beginning for truck shipments, in drums, of

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recycled uranium trioxide product to the Oak Ridge K-25 Gaseous Diffusion Plant (GDP). (Copies of these historical transfer documents, with attendant analytical data, were previously shown in this report as Figures 3-4 and 3-5.) This March 1952 UO_3 shipment is consistent with Hanford production history indicating UO_3 test runs in January 1952 and full operation in February 1952. The primary recipient of early 1950s Hanford UO_3 was to be the Harshaw Plant [Sturges 1952], but shipments were diverted to Oak Ridge facilities as their feedstocks became depleted. In March 1959, General Electric was authorized by the AEC to begin routine shipments of low-enriched (0.94% ^{235}U before irradiation) UO_3 to the K-25 facilities in Oak Ridge [Gifford 1959]. Hanford LEU UO_3 shipments began soon thereafter.

3.3.4 Out-of-Scope Research and Development Spent Fuel

The irradiated fuel research and development program, referred to as the Pile Enrichment program, involved the receipt of unirradiated slugs from Y-12, irradiation in Hanford reactors, and shipment to Idaho.

The J-1 slugs were irradiated at H reactor and the J-2 slugs at C reactor. The "C" slugs were irradiated at C and H reactor. Early in calendar year 1952, as the Idaho Chemical Processing Plant (ICPP) came on line, shipments of these "J" irradiated slugs began [Sturges 1953]. *These transactions between Hanford and Idaho are considered out-of-scope for this study.*

Prior to and continuing into 1952, Hanford also transferred small research quantities of aqueous uranyl nitrate hexahydrate, processed through REDOX and U-Plant, to Mallinckrodt Chemical Works and the Oak Ridge K-25 facility for subsequent conversion to UO_3 [Richards 1950]. Although uncommon, UNH solutions were shipped offsite by rail in tanker cars. In 1952, Hanford shipped UNH to Brush Beryllium Company in Luckey, Ohio [Freitag 1952]. This company stored the UNH until it could be transferred to Harshaw for conversion to UO_3 .

3.3.5 Post Fiscal Year 1970 Shipments

After FY 1970, Hanford shipments continued to Fernald. In the early 1970s, Hanford missions also became more diversified with uranium materials being allotted by Defense Programs to support Research and Development projects such as the Fast Flux Test Facility (FFTF). Additionally, some of the Hanford recycled legacy metal and scrap was shipped outside the United States to support Mutual Defense Agreements and Hanford environmental management missions. Approximately 1,000 MTU were shipped abroad between 1993 and 1996 to support these governmental agreements [De-Minimis 2000].

3.3.6 Shipment Packaging and Scheduling

In the early 1950s, UO_3 product was shipped in steel 55-gallon drums via both truck and rail. Beginning in 1956, T-Hoppers based on a Union Carbide Nuclear Company design

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In the 1980s, UO_3 process pipeline storage capacity was 45.6 tons of UO_3 . Yard storage of UO_3 in 55-gallon drums or T-Hoppers was virtually unlimited (>1,500 MTU). Loading could keep up with maximum production rates.

3.3.7 Transaction Material Control and Accountability (MC&A):

Beginning in the early 1950s, shipment and receipt requests were approved through AEC correspondence. This correspondence was as both letters and teletypes between the various field offices. Transactions were recorded on AEC 101 forms. In later years, this form evolved into the current DOE 741 form but the basic function has remained unchanged. An example of the transfer forms and product acceptance forms are shown in Figures 3-4, 3-5, and 3-6 respectively. Key portions of the form included transfer authority, material type and description, authorized shipper signature, and authorized receiver signature. Accountability values were based on the net weight of the UO_3 which were, in turn, determined by chemical analysis of composite samples with the ^{235}U content determined by mass spectrometry analysis of the representative composite samples. As the planning for shipments evolved, any Hanford shipments which Hanford testing indicated were out-of-specification were reviewed, by formal correspondence, by the receiver site and approved prior to any physical transfers.

In the early 1980s, as the Rockwell Hanford contractor readied for the restart of the PUREX Plant and UNH shipments to the UO_3 Plant, the PUREX Material Control and Accountability Plan [Larson 1982] was prepared in which three analytical quality control programs were implemented for the laboratory measurement systems. The three systems were:

- Maintenance of control charts for each laboratory system
- Strict adherence to the Control of Analytical Measurement Systems (CAMS)
- Statistical tracking and evaluation per the Laboratory Accountability Measurement Program (LAMP) [RHO-MA-138 1978]

3.3.8 Sample Exchange Programs and Sample Shipments

In the late 1940s, the AEC understood the need for establishing a complex-wide set of uranium specifications and measurement methodology. Early specifications for depleted UO_3 were led by Oak Ridge and concurred with by Mallinckrodt, Harshaw, Hanford and NLO. As detailed in Section 4.1, early Hanford laboratory analyses were performed in accordance with HW-24403 (sections 472.2, 285.1, 660.22, 845.10, & 845.14) [McIntosh 1952]. Specifications for enriched UO_3 were based on K-25 operating experience and implemented at Hanford [Smith 1959].

Early in the 1950s, samples were exchanged for comparison and standardization. A triad of measurement programs provided standards and limits for the uranium

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transactions. The three programs included 1) the AEC-wide Measurements Program; 2) Fissionable Standards Samples Committee; and 3) Sample Exchange Program.

In the 1950s, Hanford's UO_3 Plant Control Laboratory composited samples of each ten (10) drum lot for each carload of UO_3 product shipped. Samples were analyzed at Hanford and one-half of each sample was sent to the receiving site for check analysis. About 25-30 samples monthly were exchanged with K-25 and Harshaw [Hauff 1952].

For inbound billets in the 1950s, uranium metal quality control of non-radioactive constituents was maintained through an analytical checking arrangement with Mallinckrodt with up to 10 samples per month exchanged. Hanford sampling of metal occurred before it was placed into storage [Hauff 1952]. The Hanford specification [Groot 1952] for receipt of uranium metal was strictly enforced with "...no deviations from these specifications will be accepted without prior approval" [McCullough 1952].

3.4 Recycle Uranium Scrap, Waste, and Conversion

3.4.1 Introduction

In the sub-sections below are summaries of Hanford's past waste handling activities that are relevant to recycled uranium. Also included is a discussion of the uranium consumed in the production reactors. Waste and scrap streams from the 300 Area Fuel Fabrication facilities, the separations plants and the UO_3 Plant are discussed. Each of these processes has been previously described in Section 2.0

The uranium waste streams were examined for possibilities of disposition and uranium content. Overall, less than two percent of the uranium handled in all aspects of operation was discharged as waste or local environmental releases.

3.4.2 Reactor Fuel Element Fabrication

Uranium-containing wastes were generated during the fabrication of reactor fuel elements. For the majority of the fuel fabrication activities, uranium slugs received at Hanford were first cleaned and then canned in aluminum cans. For a short time period, Hanford received metal ingots that were extruded, rolled, and cut into slugs or "cored" fuel rods for canning. With the start-up of Fernald, Hanford received billets that were coextruded, sectioned to specified lengths, and finished. The various unit operations included a number of cleaning, degreasing, acid leaching, and autoclave operations using nitric acid, hydrofluoric acid, sulfuric acid, organic solvents, and water. The liquid streams from these operations were treated to recover uranium. The uranium-containing sludge recovered from the treatment activities was processed to recover uranium. After treatment, these liquid wastes were routed to ponds and trenches. During 1984, the reported amount of uranium discharged, via liquid waste, was 0.004 percent [Hillesland 1984].

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Scraps in the form of chips and turnings from the lathes, rejected fuel slugs and the "butts" from the extrusion processes were salvaged and recycled. The chips and turnings were sorted, broken into smaller pieces, washed, dried, and pressed into briquettes. Initially, the filtered solids and dust were put into an oxide burner and converted to oxide. Later the chips and fines were drummed and sent to Fernald for recycle. Some of the fines and dust were cemented in drums and sent to solid waste disposal. (Additional information on scrap handling can be found in Sections 2.2.7.2, 2.2.8.5, and in Appendix F.)

Airborne effluents from uranium sawing and lathe operations were exhausted through an exhaust system equipped with a water spray scrubber to remove uranium particles, chemical vapors, gases, fumes and smoke particles. A typical annual emissions report from the 333 Building [Riches 1979] stated that the uranium concentration from the cutoff saw exhaust was 4.6×10^{-9} lb/ft³ in a total air volume of 2.9×10^9 ft³.

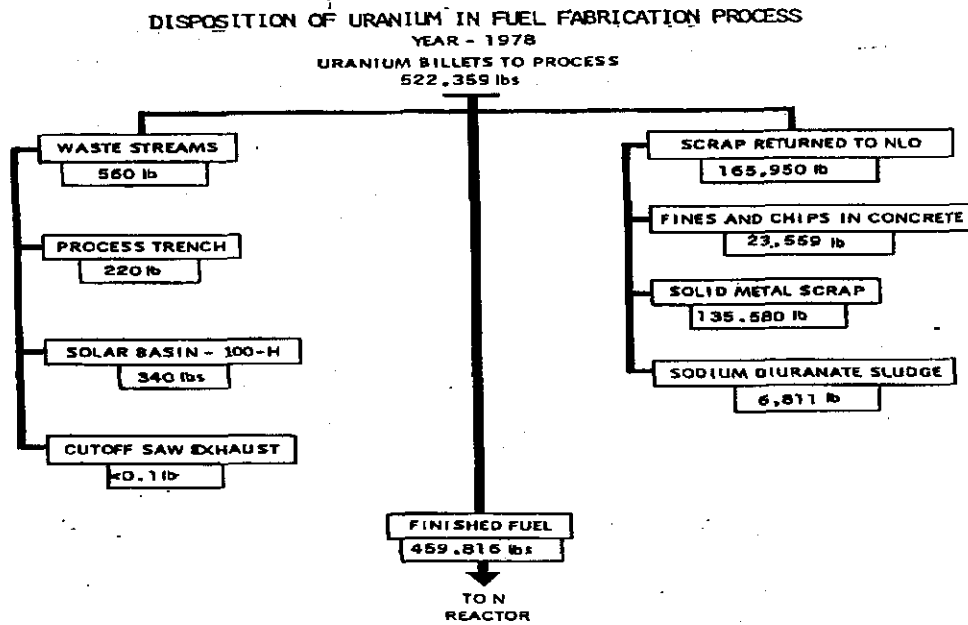
Solid uranium wastes, which included materials in failed and replaced equipment and normal line-generated process waste, were sent to Hanford burial grounds in the 300 and 600 Areas.

The description which follows, is based on the DOE Environmental Assessment [DOE/EA-0030 1980] and provides summary level information of scrap and waste streams from the 300 Area Fuel Fabrication facilities.

Uranium processing and effluent streams follow four principal material flow paths as related to fuel manufacturing. These are finished fuel, in-process storage, scrap returned to National Lead of Ohio (Fernald) for recovery, and waste streams. These streams are shown in Figure 3-10.

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Uranium Flow Path in Fuels Fabrication Process CY-1978.
(This figure presents the uranium flow and inventory
in the Fuels Manufacturing Process during CY-1978.)

**Figure 3-10 Uranium Flow and Inventory in Fuels Manufacturing Process
(CY1978) [DOE/EA-0030 1980]**

3.4.2.1 Scrap Returned to Fernald

Uranium metal scrap and sludge from uranium-bearing acids were returned to Fernald for reprocessing. Uranium scrap sources included uranium chips and saw fines, solid metal scrap, and sodium diuranate sludge.

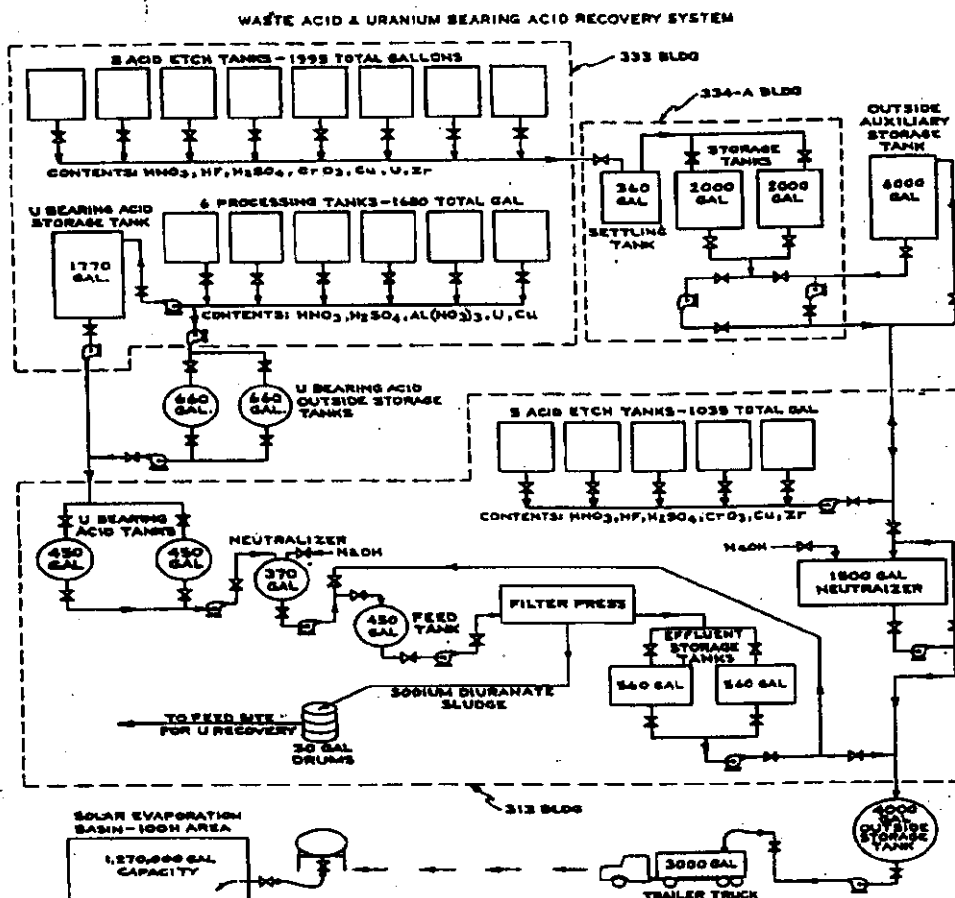
3.4.2.2 Liquid Effluent - Chemical Waste Containing Uranium

A chemical waste system was used in the 300 Area to receive and dispose of all concentrated liquid chemical wastes, including three liquid waste streams containing uranium. As shown in Figure 3-11, the system provided for collection, neutralization, and transportation of the wastes to concrete basins in the 100-H Area where the liquids would evaporate to form a solid salt cake. Later, as part of the Hanford Site response to the Comprehensive Environmental Response, Compensation and Liability Act of 1980 (CERCLA) the residual material was stabilized, removed, and buried at the Hanford Environmental Restoration Disposal Facility (ERDF).

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The only routine chemical wastes and uranium particulates to enter the process sewer from Buildings 313 and 333 were from process sewer rinse tanks, air scrubbers, wash stations, cut-off saws, and the concretion facility in the 304 Building.



Waste Acid and Uranium-Bearing Acid Recovery System

Figure 3-11 Waste Acid and Uranium-Bearing Acid Recovery System
[DOE/EA-0030 1980]

Chemical solutions that contained appreciable amounts of uranium were collected in holding tanks, pumped to Building 313 and neutralized with sodium hydroxide. The precipitate was shipped to Fernald for recovery.

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Chemicals used and transferred were controlled, and liquids discharged to the process sewer were neutralized. Neutralized waste storage tanks (surrounded by dikes) held 10,000 gallons of liquid waste.

3.4.2.3 Airborne Effluents

All plants had filtered ventilation and air monitoring devices to assure safety of personnel and that atmospheric releases were controlled. It is noted that prior to 1948, T-Plant and B-Plant did not have exhaust filters and had to restrict dissolution activities to periods when atmospheric conditions would permit maximum dilution of the radioactive and non-radioactive off-gases. Ventilation systems were provided at process locations to collect and remove airborne uranium particulates and smoke and to discharge the filtered air outside the buildings.

3.4.2.4 Solid Waste

Solid waste contaminated with uranium was packaged for transfer, by truck, to the burial sites in the 200 and 600 Areas. Some of the solid burial sites in the 600 Area contain unreported quantities of uranium waste. The Waste Information Data System maintained by the Environmental Restoration Contractor has information on each waste site on the Hanford Project. Included in the description of each site is the concentration, when known, of the chemical and radionuclide concentration.

3.4.2.5 300 Area Process Trenches

The chemical wastes and uranium within the process water that entered the process sewer from fuel fabrication were diluted in the sewer before being discharged into the two process sewer trenches.

3.4.3 Hanford Separation Plants

T-Plant, B-Plant, and the REDOX, U-Plant, and PUREX separations plants routinely discharged uranium in a number of waste streams to the environment, waste storage tanks, and to the solid waste burial ground. Plant operations were designed to minimize loss of product and for protection of workers and the environment. As earlier stated, both the T-Plant and B-Plant processed irradiated fuel to recover plutonium from the uranium and fission products, which were transferred to underground tanks. REDOX and PUREX recovered both plutonium and uranium as primary products. U-Plant reclaimed the uranium from the waste that had been discharged from the T- and B-plants.

Solid wastes, such as failed equipment and line-generated wastes, were sent to the Hanford Burial Grounds.

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3.4.4 Recovery of Uranium in the U-Plant

After uranium had been removed in the TBP process at U-Plant, residual liquid was returned to the waste tanks and chemically struck to cause precipitation of the fission products. Clarified liquid was then pumped to the BC cribs located just south of the 200 East Area. Approximately thirty million gallons of waste liquors containing about 5,700 kgs of uranium were thus disposed.

Other wastes from the TBP process were disposed to the liquid and solid waste pathways described in Section 3.4.6.

3.4.5 Uranium Trioxide (UO₃) Plant

The major unit operations performed at the UO₃ Plant were concentration of uranyl nitrate hexahydrate (UNH), calcination of UNH to UO₃, packaging of the UO₃ product, and nitric acid recovery. Uranium-containing wastes were generated during routine operation. The waste streams included solid wastes which were buried, the liquid effluents discharged to the ground, and gaseous effluents released to the atmosphere.

The UO₃ process condensates were pumped to the 216-U-12 Crib, though some went to the acid absorber tower for use as reflux water. Uranium-contaminated liquid wastes including steam condensate, chemical sewer, and cooling water were discharged to the U-10 pond.

The vapors leaving the concentrators contained water and very dilute nitric acid which were condensed and discarded as waste. Calcination of the UNH produced oxides of nitrogen, oxygen and water. The gaseous products were drawn through an off-gas scrubber, a gas cooler, and an absorption tower before being discharged to the atmosphere. A portion of the recovered nitric acid was circulated back through the acid scrubber and the remainder was pumped to storage for shipment back to the PUREX Plant. The nitric acid had a low residual level of UNH. The flowsheet [Raab 1978] indicated that the UO₃ content of the scrubber off-gas was negligible. The UO₃ product was conveyed to a cyclone separator where the UO₃ powder and the transporting air were separated. The air was filtered first through two bag filters and then a final filter before discharge to the atmosphere.

Solid contaminated uranium waste, consisted typically of failed equipment and normal line-generated process waste. These solid wastes were buried in the 200 Area waste burial grounds.

Gaseous wastes from concentration, calcination powder handling, and acid recovery operation were filtered and discharged to the atmosphere. Radioactive elements in this stream included uranium.

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200 Area is based on an estimated 77.9 Ci of uranium discharged to cribs and trenches [Diediker 1999]. An earlier estimate [TRAC-0151-VA 1991] reported 143 MTU based on an estimate of 137 Ci of uranium in the liquid waste. The quantity of uranium in solid waste is based on the reported estimate in each burial ground as of the end of 1998 [Hagel 1999]. The estimate includes a small contribution from solid uranium bearing waste from offsite. The quantity of uranium in liquid wastes to the ground for the 300 Area is based on the data reported in the 1988 hazards ranking report [Stenner 1988]. The waste in the North and South ponds has been excavated and shipped to the Environmental Restoration Disposal Facility. The quantity of uranium in solid waste in the 300 Area is based on data provided by the Environmental Resource Center and reported in the Waste Information Data System. The 300 Area generated solid U waste was actually buried in or moved to the 600 Area burial. Several of the solid waste burial sites in the 600 Area contain unreported quantities of uranium waste.

3.4.7 Uranium Losses Through Transmutation and Fission

Uranium fuel fabricated in the 300 Area Fuel Fabrication Facility was irradiated in one of nine reactors that were operated at Hanford. The reactors primarily produced plutonium for the Defense Program, but a number of other products were produced to support ongoing Defense and Nuclear Energy Programs. During reactor operations uranium was fissioned to produce fission products and uranium was transmuted to other radionuclides, including plutonium.

An estimate of the quantity of uranium consumed in the reactors has been made on the basis of the quantity of plutonium produced at Hanford, the change in the percentage of ^{235}U in the uranium fuel to the reactors, and the percentage of ^{235}U in the uranium fuel discharged from the reactors. Between 1945 and 1989 Hanford produced 67.4 MT Pu. [DOE DP-0137 1996] This would have required the consumption of an equivalent quantity of ^{238}U . Normal uranium (0.711 wt % ^{235}U) or low enriched uranium (0.94-1.25 wt % ^{235}U) was the feed to the reactors. The uranium recovered from processing was slightly depleted in ^{235}U . Assuming that 10% of the uranium received at Hanford for fuel fabrication was returned as fabrication scrap without cycling it through the reactors, an estimated 66 MT of ^{235}U was fissioned in the reactors. If it is also assumed that 10% of the plutonium produced was also fissioned or transmuted, then ~140 MTU was consumed in the reactors. This calculation results in a net loss of uranium in the overall uranium site balance.

3.5 Overall Recycled Uranium Site Mass Balance

In the attempt to segregate out the Hanford Site recycled uranium component, a mass balance including both in-scope and out-of scope uranium was developed. Development of this material balance was very complex because uranium transactions internal to Hanford activities needed to be clearly separated from non-Hanford transactions. In establishing a mass balance, both the Hanford Site contractors and the Pacific Northwest National Laboratory (PNNL) needed to be integrated into the calculations. Two issues related to shipper/receiver correlation of historical transactions

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make a precise mass flow extremely difficult. The first issue is that while MC&A records indicate shipments to offsite locations, it was not uncommon for shipments to be diverted, during transit, to secondary locations to address feedstock shortages. Secondly, for fiscal year transaction reconciliation between sites, quantities leaving one site near the end of a fiscal year may not be received and entered into the receiver site's MC&A records (booked values) until the next fiscal year. Further difficulties with establishing precise mass flows at Hanford are in establishing the accuracy of estimates for normal operational losses (NOL), accuracy of measured discards, accuracy of estimated discards, reconciliation of Inventory Differences (ID) from continual contractor turnovers, accuracy of past decay calculations, and accuracy of Material Unaccounted For (MUF) explanations. An example of one difficulty was when Hanford, within a semi-arid environment, shipped UO_3 powder to the southeast. During transit and upon arrival at the southeast receipt location, the UO_3 absorbed moisture, resulting in larger receipt quantities measured than were reported shipped from Hanford.

Table 3-10 and Figure 3-12 summarize these mass flows. At the right of each entry in Table 3-10 is a reference number which maps to the index below for further details to entered quantities and attendant reference documents. As the table indicates, approximately 115,955.4 metric tons of uranium (all types) were received at Hanford (Hanford and PNNL) from January 1948 through March 30, 1999. Approximately 112,287.3 metric tons were shipped within this same period. Approximately 4,006 MTU remains in the Material Control and Accountability (MC&A) inventory and approximately 2,314 MTU was lost to waste and reactor consumption. This leaves a difference of about 664.1 MTU between receipts, on-site holdings, uranium consumed, and shipments. This difference is primarily attributed to uncertainties in the quantities of uranium in waste, that which was consumed in the reactors, and the limited data from the pre-1948 operating period. As indicated in Figure 3-12, the recycled uranium component of the receipt total is approximately 109,143.6 MTU (~94%). The recycled component of the shipment total was approximately 109,792 MTU (~98%). Approximately 6,180 MTU is at the Hanford site in the form of current inventory or waste. An additional approximately 140 MTU was fissioned or transmuted in the production reactors.

Index Mapping for Summary Table 3-10:

Entry #	Table Reference (Receipts)	Entry #	Table Reference (Removals)
1.	Appendix B, Table 3.2.1	8.	Appendix B, Table 3.3.1
2.	Appendix B, Table 3.2.1	9.	Appendix B, Table 3.3.2
3.	Appendix B, Table 3.2.1	10.	Appendix B, Table 3.3.3
4.	Appendix B, Table 3.2.2	11.	Appendix B, Table 3.3.4
5.	Appendix B, Table 3.2.3		Appendix B, Table 3.3.5
6.	Appendix B, Table 3.2.4		Appendix B, Table 3.3.6
	Appendix B, Table 3.2.5		Appendix B, Table 3.3.7
	Appendix B, Table 3.2.6	12.	Appendix B, Table 3.3.8
	Appendix B, Table 3.2.7	13.	Section 5, Table 5.1.1
7.	Appendix B, Table 3.2.8	14.	Section 5, Table 5.1.2
		15.	Section 3.4

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Table 3-10 Hanford Mass Balance-Total In-Scope & Out-of-Scope

Hanford Ending Inventory				Quantity	Units
	31-Dec-47	In-Process (Fuel Fab, Rctrs, Storage, etc.)		1,400.3	MTU
	31-Dec-47	In Hanford Waste Tanks		1,915.7	MTU
Receipts:					
HANFORD	Receipts: 1-Jan-48	31-Dec-49	Aggregate Receipts (All U Types)	3,402.3	MTU
	<i>From Offsite</i>	1-Jan-50	EO FY 1965	Aggregate Receipts (All U Types)	81,013.2 MTU
		FY 1966	EO FY 1970	Aggregate Receipts (All U Types)	19,119.5 MTU
		FY 1971	31-Mar-99	Aggregate Receipts (All U Types)	12,142.1 MTU
			<i>Hanford Receipt Subtotal</i>	115,677.1	MTU
PNNL	Receipts: FY 1965	31-Mar-99	Aggregate Receipts (All U Types)	278.3	MTU
	<i>From Offsite</i>		<i>PNNL Receipt Subtotal</i>	278.3	MTU
			Receipt Subtotal	115,955.4	MTU
			Receipt & 47 Ending Inventory	119,271.4	MTU
Shipments:					
Hanford Shipments	1-Jan-48	EO FY 1951	Aggregate Shipments (All U Types)	1,601.6	MTU
	<i>Offsite</i>	FY 1952	EO FY 1965	Aggregate Shipments (All U Types)	68,282.6 MTU
		FY 1966	EO FY 1970	Aggregate Shipments (All U Types)	28,643.5 MTU
		FY 1971	31-Mar-99	Aggregate Shipments (All U Types)	13,515.7 MTU
			<i>Hanford Shipment Subtotal</i>	112,043.4	MTU
PNNL Shipments:					
	<i>To Offsite</i>	FY 1965	31-Mar-99	Aggregate Shipments (All U Types)	243.9 MTU
			<i>PNNL Shipment Subtotal</i>	243.9	MTU
			Shipment Total	112,287.3	MTU
3/31/99 Inventory:					
Hanford			Current Unirradiated In-Scope Inventory	1,862.6	MTU
Hanford			Current Irradiated & MOX Out-of-Scope Inventory	2,137	MTU
PNNL			Current Inventory	6.4	MTU
			<i>Inventory Subtotal</i>	4,006	MTU
			Subtotal Transaction Difference	2,978.1	MTU
Waste & Fission Loss:					
Hanford			Uranium in Waste Tanks	958	MTU
			Uranium in Solid Waste	1,054	MTU
			Uranium in Ponds, Cribs, & Ditches	162	MTU
			Uranium Lost thru Pu Production & Fission	140	MTU
			Total Difference	664.1	MTU

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The earliest specifications on UO_3 product for recycle required analyses to be performed for chemical and physical properties before it left the Hanford Site. Typical analyses included beta and gamma activity as a percent of aged natural uranium, metal impurities, density, particle size, plutonium, and sulfur. Prior to initiating the addition of sulfur, a chemical reactivity measurement was included to predict the effectiveness of the conversion of UO_3 to UF_4 in subsequent processing. Neptunium-237 analysis was not requested on the UO_3 product until about 1978 and technetium-99 was not included in analysis requirements until 1985. Although specification threshold concentrations were proposed, neither isotope was included in the specifications. As a result, there is a wide variation in the quantity of data available for Pu, ^{237}Np , and ^{99}Tc contaminants in Hanford UO_3 . This section focuses on the concentration of the three primary constituents of concern, Pu, ^{237}Np , and ^{99}Tc , although concentration data for other constituents are discussed.

4.1 UO_3 Process Specifications

Processing specifications of the UO_3 Plant interacted with those of the separations plants (U-Plant, REDOX, and PUREX) because the UNH product from the separations plants was the feed to UO_3 Plant. The UNH did not leave the separations plant if chemical analyses showed the product to be outside the threshold concentrations in any constituent of concern unless the UO_3 Plant manager granted prior approval. These threshold values were generally consistent through the years of UO_3 operation and are described below.

4.1.1 Feed Specifications

Feed specifications for the initial operation of the UO_3 Plant are provided in the UO_3 flowsheet included in Section 2.2.4.3. They indicate that the feed material from REDOX and U-Plants were tightly controlled, as the UO_3 process provided no further purification of the uranium. The need for process control was recognized in the late 1940s and early 1950s, before the UO_3 Plant came on line. Once decisions were made in the late 1940s to "enrich the depleted uranium back to normal concentrations" [Greenwalt 1947], questions were raised about "firm specifications for the final uranium product to be delivered from either the REDOX and the TBP Plants or an uranyl nitrate-oxide conversion plant at the Hanford works" [Greninger 1950]. The separation plants were originally designed on the basis that the recovered uranium would be sufficiently decontaminated with respect to Pu and gross beta and gamma activity to permit essentially direct physical handling of the final product in its last form at Hanford. It was also recognized at this early stage that subsequent processing at other plants might result in fractionation or concentration of either fission products or Pu and cause a need for more highly specific or greater decontamination than would be required at Hanford. A Pu concentration limit was defined in 1948 [Gamertsfelder 1948] based on the

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tolerance level in breathing air. The conclusion was stated "that material with a purity limit of one part in 100 million parts uranium could be handled essentially as natural uranium." In a 1951 letter [Gamertsfelder 1951], the limit was again considered and it is stated that "reclaimed uranium should contain no more than one part plutonium in 7.8×10^6 parts uranium" in order that the hazard due to breathing air contaminated with reclaimed uranium should be no more than 10% greater than for ordinary uranium. This value allowed a 10 ppb limit to be established and maintained throughout the history of the Hanford production era with greater than a ten fold conservatism factor built in.

4.1.2 Product Specifications

Threshold concentrations of constituents in UNH were included in technical manuals for both REDOX and U-Plants based on expected performance of the processes to purify the UO_3 product. Both manuals set Pu concentration levels at 100 ppb [HW-18700 1951 and HW-19140 1951] but these threshold concentrations were not accepted by Oak Ridge, the Site responsible for setting standards for the UO_3 Receiver Sites. Only product meeting the 10 ppb Pu specification were to be shipped. Negotiations continued between the sites into 1953 by which time the processes demonstrated the ability to meet a more stringent quality requirement.

Although firm specifications were reissued many times, (see Table 4-1) and changes were made in the beta, gamma, chemical reactivity, and metal impurities thresholds, the approved Pu specification value did not change. In 1951, Hanford proposed a Pu specification of 50 ppb but Oak Ridge held firm in maintaining the 10 ppb limit [Sapirie 1951]. Communications between Oak Ridge and Hanford continued into 1953 with adjustments to accept metal impurities up to 200 ppm with stipulations, but "in regards to the plutonium content, the specification of less than 10 ppb should not be exceeded and, if possible, the plutonium level should be even lower. Oxide received from Hanford has, in general, contained less than 5 parts per billion plutonium" [Sapirie 1953].

The product specifications for the UO_3 Plant provided chemical and radiological requirements that had to be met. In 1953, based on operating experience, Hanford and Oak Ridge representatives agreed upon the properties of the Hanford UO_3 to be included in a firm specification. These properties included fission product activity, Pu content, purity (uranium content), particle size, and volatile impurity content. "The maximum acceptable Pu concentration shall be ten parts of plutonium per billion parts of uranium. Plutonium shall be determined on each carload composite" [Smith 1953]. Table 4-1, UO_3 Process Specifications, provides a list of the firm specification documents in place during the life of the UO_3 Plant. The information in these documents show that the required Pu concentration specification remained unchanged.

4.1.3 Proposed Specifications

Product specifications for ^{233}U , ^{232}U , ^{237}Np , and ^{99}Tc were also discussed but not adopted. In 1962, it was proposed that the maximum concentration of ^{233}U be set at 90 ppm on a ^{235}U basis, and ^{232}U be set at $1.10\text{E-}2$ ppm on a ^{235}U basis [Judson 1962]. In

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transferred from the separations plants to the UO_3 Plant with prior approval if the UO_3 Plant had material on hand that could be blended to bring the out-of-specification material to within specification concentration.

Figure 4-1, an example page from one of the specification documents, is included for completeness. It defines the threshold quantity of impurity that was allowed and the analytical method that was used to generate the result.

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Analysis	Method Ref.
Sodium	1000.1, 769
Calcium	1000.1
Aluminum	1000.1
Iron	1000.1, 515.25
Chromium	1000.1, 341.1
Nickel	1000.1
Density, bulk	647.2
Density, packed	647.5
3.22 Properties with Established Limits	
3.22.1 Gamma Activity	
Gamma activity due to fission products shall be determined on each carload composite. For the purpose of setting the average acceptable gamma activity, shipments of uranium oxide will be considered in designated blocks of ten consecutive carloads. The average acceptable gamma activity due to fission products for a block shall not exceed 100 percent of the gamma activity for aged normal uranium. For the purpose of setting the maximum acceptable gamma activity specification, one carload shall be considered a unit. The maximum acceptable gamma activity for a unit, due to fission products, shall be 300 percent of the gamma activity of aged normal uranium.	
3.22.2 Beta Activity	
Beta activity due to fission products shall be determined on each carload composite. The maximum acceptable beta activity due to fission products shall be 100 percent of the beta activity of aged normal uranium.	
3.22.3 Plutonium Content	
Plutonium shall be determined on each carload composite. The maximum acceptable plutonium concentration shall be ten parts of plutonium per billion parts of uranium.	

Figure 4-1 Example Page of Specifications for UO_3 Plant

4.2 Recycle UO_3 Processing

Each of the separations processes (i.e. U-Plant, REDOX, PUREX) sampled the UNH product prior to sending it to a load-out tank for transfer to the UO_3 Plant. This internal transfer was not made until the analytical results were completed. If the UNH material was out of specifications in any respect, the material was recycled back to the partitioning cycle and reworked before being transferred to the UO_3 process [HW-25744 1952]. Weekly and monthly reports contain several examples of this rework being necessary during the early years of Hanford operations. In the REDOX process, this rework was most commonly necessary to reduce the fission product activity rather than

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for reducing the levels of the three primary constituents of concern. There was a final silica gel extraction for removal of zirconium/niobium-95 ($^{95}\text{ZrNb}$), if those radionuclide concentrations were found to be high. This step was omitted if the UNH was found acceptable without employing this process step. Laboratory oriented, research and engineering flowsheet improvements were provided on a continuous basis through the operational life of each process. In addition, Process Engineering Support monitored and evaluated process operations on a daily basis. A few examples of these activities are recorded in the weekly and monthly reports and are included below to demonstrate the attention that was paid to UNH product quality in the separations plants and the UO_3 product. In spite of this strict adherence to UNH specifications, five references have been found that document the shipment of UO_3 product with Pu concentrations outside the 10 ppb limit. These citations are included in Section 4.2.1. Although Hanford documents indicate these shipments were made after approval was obtained from of the receiver site, copies of receiver site acknowledgement of these notifications have not been located.

4.2.1 Processing Issues

During processing at the separations operations (REDOX, PUREX, U-Plant), upsets occurred that caused the UNH product to be outside the acceptable specifications in one or more constituents. Provisions were made during the construction of these facilities for taking remedial actions without exacerbating the entire process. Provisions were made for storage and rework of the UNH prior to transfer to the final loadout tanks. The U-Plant process provided alternative decontamination of REDOX UNH from impurities prior to transfer to the UO_3 process. There are also records that document the transfer of UNH produced in the U-Plant process to REDOX for decontamination of $^{95}\text{ZrNb}$. In PUREX, provisions were made for rework of the UNH prior to transfer to the staging tanks if the product was found to be outside the specification.

4.2.2 Typical Hanford Responses

The following items are presented to provide examples of the types of issues addressed by the Process Operations and the laboratory-oriented Research and Engineering organizations: (These items are not listed in chronological order and only items that pertain to UNH product quality have been included. All items found addressing Pu issues have been included.)

- Provisions were made to "ship all UO_3 which fails to meet specifications for impurities, other than radioactive contaminants, to the Harshaw Chemical Company" [Shaw 1952]. A number of railcar shipments (numbers 77-87, 90, 93, 94, 96, 98) are recorded from July through September 1953) as being sent under this directive. Sodium contamination was a continuing problem in the UNH recovered from the waste tanks by the U-Plant. Iron concentrations above acceptable thresholds from corrosion were a recurring issue. These recurring non-conformance issues continued throughout the 1950s.

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- In 1953, a weekly report for REDOX [Christy 1953] states that six batches of UNH in excess of gamma specifications were sent to storage for decay of ^{237}U .
- A firm specification was included in the REDOX and PUREX process operations that limited the concentration of plutonium allowed in the UNH product. Several references are recorded that indicate UNH transfers were held up awaiting analytical results on the process samples to verify that the product met the specification [HW-50584-DEL 1957].
- Several examples were encountered of UO_3 produced from PUREX UNH that was outside the acceptable threshold for iron concentration [HW-48835-DEL 1957]. The excessive iron contamination in the final UO_3 was found to originate in the UO_3 Plant rather than in PUREX.
- An extensive investigation of analytical methods was initiated to resolve an apparent discrepancy of plutonium values in uranium as reported by REDOX and PUREX Laboratories. Subsequently, a real discrepancy was found to exist between results obtained by two different methods employed in the different laboratories. An analytical procedure was accepted that both laboratories subsequently used [HW-48835-DEL 1957].
- Conversion of UNH to UO_3 was frequently hampered by foaming in the pot calciners. The identified source of this issue was the organic extractant, used in the U-Plant process, that contaminated the aqueous UNH feed. This issue was resolved when the continuous calciners were put into service.
- A shipment (carload #8) was made to Harshaw that contained 30 ppb Pu in 1952 [Richards 1952d].
- Three cars of continuous calciner powder were outside shipping specifications, however, they were accepted by the customer prior to shipment. The reason one car (UA-16) contained 16 ppb plutonium concentration was unexplained since the UNH feed was determined to be within the 10 ppb limit [HW-48835-DEL 1957]. An investigation was initiated that resulted in a modified procedure [HW-50584-DEL 1957] that eliminated the bias due to neptunium coextracting with the plutonium in the final uranium analyses. The quantity of powder represented is not given specifically but (at this time) the usual shipment contained 10 drums, each containing 900 pounds, which comprised one carload. These shipments were made prior to use of T-Hoppers which contained 4.5 metric ton of UO_3 . The UO_3 in one car exceeded the iron limit of 50 ppm and the third car exceeded the particle size specification of 98% passing a 40 mesh screen.
- In December 1953, a shipment of UO_3 product was made to Paducah, after acceptance by Oak Ridge, with 19 ppb Pu [Christy 1954].

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- In January 1954 an additional two cars, numbers 148 and 149, of UO_3 product were shipped to Paducah with 13 and 12 ppb Pu respectively. Lot 148 resulted from poor quality REDOX UNH and Lot 149 resulted from poor U-Plant UNH [Christy 1954a].
- A T-Hopper (T 58) was returned to the Hanford Site with residual material from the shipment of GDP tails from Paducah to Fernald. Typical quantities of residue in these containers is approximately two kilograms. Information from Fernald indicates that the ash heel in this T-Hopper was 900 grams, was 40.69 wt. % uranium, and contained ~7,760 ppb plutonium, and ~25,300 ppb neptunium on a uranium basis. T-Hopper T 58 was subsequently refilled with UO_3 and is in storage at Hanford.

4.3 Analytical Laboratories

In the Hanford separations areas, buildings with the designation of "222" were laboratory facilities (222-B, -T, -S, and -U Labs) that supported the separations facility with the corresponding letter designation. As the only laboratory incorporated into its process building, the PUREX laboratory did not carry its own building designation. Analytical services for the UO_3 Plant were provided by the 222-S Laboratory after the Metal Recovery and TBP processes were discontinued and the U-Plant laboratory was closed. These measurements continued in 222-S Laboratory until the UO_3 process was put in standby in 1972. During restart of the UO_3 process in 1983, the Plutonium Finishing Plant (PFP) Laboratory provided analyses for a short period until the testing could resume at the 222-S Laboratory. It continued there until the UO_3 process was closed again in the late 1980s. The exception to this statement is that all uranium isotopic analyses and total metal impurities measured by emission spectroscopic analyses were performed at the PFP laboratory.

4.3.1 UO_3 Product Sampling and Subsampling

Reliable analytical measurements were dependent on the adequacy of sampling and subsampling of the stream to be characterized. A new continuous sampler was designed for use when the continuous calciners were installed [Gustafson 1957]. This sampler was to replace a screw-type, continuous sampler used in the 224-UA unloading system to sample material produced in the pot calciners. A proportional sample was collected for analysis while each T-Hopper or pallet of four drums was being filled with UO_3 product.

Continuous collection is generally recognized as an appropriate methodology for reliably sampling a stream that may have variability in composition. By collecting a portion of the bulk product as it is made or moved, variations in any constituent of concern will be sampled in relation to the extent that the constituent is present in the overall product.

When a sample arrived at the laboratory, it was placed on a tumbler-mixer and thoroughly homogenized before any aliquots were extracted for any purpose. After

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homogenization, a subsample was removed from each sample to build a composite representing a "lot" of material. A "lot" normally represented ten T-Hoppers or sixty drums of UO_3 powder. This composite was again tumbled to effect thorough mixing before any subsampling was performed for either archive or analyses. In addition, a subsample of each sample was collected, sealed to exclude moisture, and stored separately for archive. Analytical measurements were then made of subsamples taken from the lot composite.

4.3.2 Analytical procedures

Procedures were developed for monitoring impurity levels and product quality through the separations processes while flowsheet testing of those processes was in progress. These procedures were transferred to the analytical laboratory supporting each process. Changes to these procedures were controlled and implemented only after closely controlled tests were conducted to verify performance. New methods of analysis were implemented to provide improved productivity or quality over the method being replaced. As higher resolution instrumentation became commercially available, especially for radionuclide characterization, these instruments were placed in use only after rigorous acceptance testing and approval of the change by the Process Engineering of the UO_3 Plant. The original procedures to be used were collected in a procedure manual [McIntosh 1952]. That document replaced a preliminary manual, HW-12864, 1950. The Product Specifications documents identified the analytical procedures to be used for monitoring the product quality of UO_3 shipped off-site, as shown in Table 4-1. The process control laboratories for REDOX, U-Plant, and PUREX were allowed to modify the procedures used on the UNH with technical justification but not without complex wide acceptance. Procedure differences were present between the laboratories and generally, the UO_3 Laboratory procedures were the last to be changed because of the time required to obtain approvals.

Uranium concentrations were measured in UNH feed by density and nitric acid concentration. Because the UNH was very uniform, and had low impurity levels, the concentration was directly proportional to the density. This measurement method was very precise and accurate as long as the stream was within accepted impurity concentration thresholds. Other methods were used in the separations processes, such as X-ray photometer and spectrophotometry. These methods provided a more robust measurement in the event the uranium concentration did not meet the specifications or impurities were unexpectedly found by other analyses to be significant and affect the density methodology.

A gravimetric analysis was performed on the UO_3 product in which the UO_3 was converted to U_3O_8 . This treatment eliminated contributions from water and corrections were made to account for the total metal impurities and sulfur associated with this compound.

The analysis methodology used for plutonium contamination was included in the original document [HW-12864 1950] although in 1960, this method was modified to improve the

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separation of plutonium from uranium and other alpha emitters [HW-65402 1960]. The formal mechanics of these methods are shown in a letter recommending substitution of an improved plutonium method from the previous accepted methodology [Harmon 1957]. This modification was included in the 1960 UO_3 process specification. Most (but not all procedures) of the established specification analyses were later documented in ARH-85 1970.

Plutonium was isolated from other alpha emitters and interferences by extracting it into thenoyl-trifluoro-acetone from a mildly acidic sample. The method is dependent on plutonium being in the extractable +4 oxidation state. The separation from uranium is effected by washing the extracted uranium from the organic phase with high concentration nitric acid. Alpha activity from ^{237}Np contributed to the plutonium alpha activity which was measured for determining the plutonium content. This resulted in a high-biased plutonium analysis. An estimate of the potential bias introduced can be calculated using the ratio of specific activities of ^{239}Pu ($1.30\text{E}11$) and ^{237}Np ($1.56\text{E}9$) and the relative concentrations of each (10 ppb and 500 ppb respectively). The Np could have introduced a high bias to the Pu result by as much as 30 percent if the Np came through the procedure quantitatively. If the Pu concentration was already near the 10 ppb limit, this contribution could be considered significant. A modification was made to the method in 1960 to improve separation of plutonium from neptunium. The modification included a reduction step that also reduced the oxidation state of neptunium and only the plutonium was reoxidized for extraction. The basic technology of the analytical method remained consistent throughout the rest of the history of the UO_3 process.

Prior to the mid-1950s, beta and gamma activity of UO_3 powder was determined using a Geiger-Mueller tube with a mica end window. During the gamma measurement, the beta activity was shielded out with an aluminum-lead-aluminum absorber. This provided a best estimate relationship of beta activity to gamma emissions. The Shonka instrument, a high-pressure ionization chamber, replaced the G-M tube. Absolute measurements were not possible on a control basis with the Shonka. Empirical controls and relationships to aged natural uranium were re-established to define relative changes in product quality. It was assumed at that time, based upon process knowledge, that product UO_3 would exceed the gamma activity threshold before reaching the beta threshold. Results from these instruments were likely biased high due to decay daughters of strontium-89 and strontium-90, if present, because of their high energy beta emission. In 1966, a modification was presented for measurement of beta and gamma activity with instrumentation that could attribute the gamma activity to specific fission products. Gamma scintillation counters replaced the Shonka after negotiations were concluded between Hanford and the recipients of the UO_3 product [Knights 1966]. In 1967, the UO_3 product specification established an upper limit of 15 $\mu\text{Ci} / \text{lb. U}$ for $^{95}\text{ZrNb}$, 50 $\mu\text{Ci} / \text{lb.}$ for the combination of ^{103}Ru $^{106}\text{RuRh}$, and 2 $\mu\text{Ci} / \text{lb.}$ for all other isotopes excluding ^{99}Tc . Ten lot average values were also established with the values for $^{95}\text{ZrNb}$, ^{103}Ru , and $^{106}\text{RuRh}$ and others limited to 10, 25, and 0.5, respectively [Knights 1966].

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Introduction of the Beckman, Wide Beta II[®] Counter, provided the ability to measure beta activity directly rather than calculating the beta activity from the beta/gamma measurements. Since ruthenium isotopes produced the majority of beta activity, the beta specification threshold was eliminated as a routine measurement. Although the Wide Beta II[®] instruments exhibited superior sensitivity to the prior instruments, the beta measurements did not adequately represent the Tc concentration in the sample. Chemical separations were required to isolate the ⁹⁹Tc from all other beta emitters before quantitation. This was not requested until the restart of the UO₃ Plant in 1984. In later tables of this section, both the previous beta/gamma percent and specific radionuclide activities are presented.

Neptunium and technetium methods were not included in the original Hanford Works documents, nor are there single accepted methods for the measurements available today. For that reason, technologies developed at the on-site laboratories were applied after extensive testing and application to the UNH matrix. Reliable neptunium measurements were not made on UO₃ until 1969 and technetium analyses were not performed until 1985. Chemical standards with the isotope were included during the analyses to verify method performance.

An emission spectrograph provided the total metal impurities measurements on all UO₃ product. This methodology was originally procured for quantitation of Pu product in the PFP and when the need for analyses of UO₃ arose, aliquots were sent to PFP for analyses. The same logic was used for the uranium isotopic analyses that were also performed at PFP.

4.3.3 Analytical Methods and Errors

Each analytical method has an uncertainty associated with the measurement that can be attributed to either random or fixed errors. Both types must be considered and with appropriate data can be evaluated individually using statistical methods. The total uncertainty of a measurement is the combination of the two types. Fixed errors are those usually associated with the chemistry of the method such as extraction coefficients, volumes of vessels, and sampling. Random errors are those that are not repetitive such as degradation of chemicals used in the measurement and inadvertent use of incorrect supplies.

Combining both types of errors provides an error band that estimates the minimum and maximum concentration of a measured constituent that may be present in a sample. Certain of these parameters are easy to assess, but others are much more difficult to establish and monitor over time. During the development of a procedure, the fixed error contribution is defined and documented. The method is only put into service if it meets the measurement criteria established to support the use of the resulting data.

Early analyses of Pu were biased high from the effect of Np being coextracted during the separation of Pu from the sample matrix. This contribution was small when the UO₃ was not recycled and the ²³⁷Np concentration was small compared to the concentration

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of Pu. As preparations were made to separate ^{237}Np as a co-product, its contribution increased. The accuracy of the plutonium measurement was estimated to be $100 \pm 20\%$ at the 99% confidence interval. Radioanalytical precision available at the time ranged from plus or minus 2 to 4% for gross beta, and plus or minus 0.2% to 10% for gross gamma depending on sample size [HW-19140 1951]. Emission spectrographic instrumentation provided data that was reliable within step ranges (20-50 ppm, 50-100 ppm), for each element. Unique quantitation values were not available on multi-element instrumentation until inductively coupled plasma atomic emission spectrometry became available in the late 1960s. Consequently, there are variations in early documented metal impurity data that may not be reproducible with current instrumentation.

4.3.4 Quality Assurance

Analytical procedures were written that implemented the standard methods documented in the manuals HW-12864, HW-24403, and HW-65402, cited earlier. Although laboratory-specific procedures were given different identification numbers, the technology remained the same. Details were modified to conform to specific instructions from the UO_3 Process Engineering organization, implementation of new instrumentation, or to provide more specificity to the measurement.

4.3.4.1 Quality Assurance Program

An extensive quality assurance program was developed for use by the laboratories performing specification analyses in support of the UO_3 process. This program was primarily focused on the measurement of uranium, but included very limited standards data to monitor the performance of analytical methods for other constituents as well. The program consisted primarily of blind standards and in-house referee analyses. The percent average recovery and precision of the average (95CL) was reported and used by the nuclear material control organization to apply a bias correction to the uranium shipment data if necessary [Rochon 1972]. The uranium measurement threshold limit for percent average recovery was about 100 ± 0.5 percent, but concern was raised if the result exceeded 100 ± 0.1 percent. A chemist was assigned to oversee and approve all results generated by the laboratory technicians. This methodology was directly applicable to the UNH received by the UO_3 Plant.

The measurement for uranium in UO_3 was sufficiently reliable that it did not require monitoring. Temperature and laboratory balances used were routinely calibrated according to accepted standards of the time. However, in 1961-1962, there were shipper-receiver discrepancies recorded in the uranium analyses. Investigation of the discrepancy was resolved by finding that hygroscopic UO_3 picked up water during shipment and storage before the measurement was made at the receiver site. These differences were less than one percent but resulted in a significant bias in uranium material balance.

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4.3.4.2 In-House Standards Program

An in-house standards program included synthetic UNH solutions (UNH from natural uranium) to which known quantities of the impurities of concern were added. These standard solutions were analyzed as a sample at a periodicity that would provide statistically significant quantities of data within the monitoring period, normally one month. During high production periods, these standard results were plentiful, however as the process was shut down for extended periods, data were not generated in sufficient numbers to be statistically evaluated for all constituents. Although sample preparation was performed in a laboratory room dedicated to uranium, analytical instruments were shared with other operations of the analytical laboratory. Standards results were not necessarily exclusive to the UNH or UO_3 product. The percent average recovery of the Pu measurements was maintained at 100 ± 10 percent at the 95 percent confidence interval. Neptunium quantitation was held to 95 ± 15 percent.

Early in the 1950s, samples were exchanged for comparison and standardization. A triad of measurement programs provided standards and limits for the uranium transactions. The three programs included: 1) AEC-wide Measurements Program; 2) Fissionable Standards Samples Committee; and 3) Sample Exchange Program. These programs were supported by the AEC and its contractors and provided different emphasis. They continued until sufficient reliability was demonstrated that significant differences in analytical results would not be encountered.

Also in the early 1950s, Hanford continued to support quality assurance efforts to validate both the measurement techniques and product quality. Early correspondence between the New York Operations Office (NYOO) and Hanford indicates that UO_3 data between Oak Ridge and Hanford correlated very closely in that:

- For both labs, the precision of mass spectrometry was 0.006%.
- For both labs, the sampling was by aliquot and was nearly foolproof.
- Comparison between Hanford General Electric (HGE) and Carbide and Carbon Chemicals (CCC K-25 Oak Ridge) shipment analysis showed five cases of agreement at 0.64%; one case of agreement at 0.65%; and two cases of difference of 0.01%. Averaging the above shows that in eight cases there is a difference of 0.00250% between the two laboratories. This represented 9.48 pounds of ^{235}U out of 399,699.06 pounds of UO_3 shipped. The conclusion is that HGE analyses for Harshaw shipments are of the same reliability as above.

An independent referee program was continued throughout the uranium recycling effort to monitor the analytical processes at the participating sites. The program established that aliquots of each container and lot composites were prepared and sent to the Site receiving the lot shipment. Since analytical measurements were performed at the receiver's site on material from the same composite, this effected a double-blind

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external referee program. Shipper-receiver differences were tracked by the Nuclear Materials Accountability personnel. If there were significant differences, the event was investigated at both locations and the differences resolved. Limited data is available that shows some differences in concentrations of plutonium were present on individual lots as expected, but were probably within the combined (two site) error band of the measurement. This inter-site comparison has not been performed.

Agreement on the Pu concentration between Hanford and Oak Ridge was more problematic in the 1952-1953 time period [HW-27314 1953]. Hanford generated data was apparently significantly lower than the Oak Ridge analysis of the same UO_3 powder. However, in one sample of four, the Pu concentration agreed. Documentation with resolution of this issue was not found.

4.4 UO_3 Analyses:

Production of UO_3 product for recycle was continuous from inception in 1952 until the PUREX and UO_3 Plant were placed in stand-down in 1972. By the time these processes were restarted in 1983, additional measurements were made that included ^{237}Np and ^{99}Tc although there was no threshold specification on the allowable concentration. Data is separated in the following sections only because of the discontinuity of operations. The sources of data used in the following subsections are gleaned from records as near to the final UO_3 shipment report as possible. The only data sources available prior to 1984 for ^{237}Np and ^{99}Tc concentrations are the specifications imposed on the process facilities, REDOX or PUREX, and a few special analyses performed on UNH at the UO_3 Plant.

4.4.1 Impurity Concentrations in UNH

As described earlier, the UO_3 process made no significant changes in the impurity concentrations of the incoming UNH. Sulfur was added to the UNH after the mid-1950s to improve the conversion of UO_3 to UF_4 at the receiving site. The concentration was varied from about 300 to as much as 3000 parts sulfur per million parts uranium according to the requests of the receivers. Radioisotopes and volatile compounds were present in the UNH feed to the UO_3 Plant. Some minor concentrations of ruthenium and other volatile fission products were volatilized during the calcination process strictly due to their volatility. The conversion of UNH to UO_3 evolved large quantities of nitrogen oxides. The majority of volatilized NO_x and some fission products were collected in the off-gas treatment system and returned to the PUREX Plant. Low concentrations of fission products were also included in wastewater discharged to the soil column. Constituents that were not volatilized remained in the UO_3 product.

4.4.2 Analyses Performed and Results

After conversion of UNH to UO_3 , chemical analyses were performed on each lot, representative samples from which consisted of ten containers (when using T-Hoppers) or sixty drums. During heavy production, when both REDOX and PUREX were

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with a maximum of 7.5 ppb and minimum of < 1 ppb. The average gamma percent for this UNH was 87 % of aged natural uranium. These are limited data, but they indicate the ability of the U-Plant Process to produce UNH that comfortably met the 10 ppb specification.

4.4.4 Storage of UO_3 at Hanford

Due to shutdown of facilities producing UF_6 at Paducah, Hanford-produced depleted UO_3 was stored at Hanford [Sloat 1964]. The enriched (nominal 0.8 wt% ^{235}U) was milled and packaged in T-Hoppers while the depleted (nominal 0.6 wt% ^{235}U) was packaged in 55 gallon drums for storage when the T-Hoppers were not available.

Plutonium and fission product concentrations on lots of depleted UO_3 packaged in drums and stored at Hanford are summarized in Table 4-3; more detailed reports are presented in the Appendix C, Table 4-3. A data package [ISO-877 1967] provided this product information for the time period from May 1964 through June 1967. The data covers 352 lots of drummed material that were processed during the 32-month period. The average plutonium concentration for the 352 lots was 2.2 ppb uranium with a low of <1 ppb and a high of 8 ppb. It should be noted that the plutonium concentration exceeded 5 ppb on only six lots out of the 352 and these were only 6, 6, 6, 7, 7 and 8 ppb. None exceeded the 10 ppb specification.

The data package includes concentrations of specific radioisotopes, and gross beta and gamma radiation levels during periods when the measurements were made. The average beta radiation for 317 lots for which data is available was 6.53% of the beta radiation level of aged natural uranium and ranged from a low of 0.31% to a maximum of 36.5%. The average gamma radiation level was 78.2% that of aged natural uranium and ranged from a low of 3.9% to a maximum of 212%.

The gross beta and gamma data were not included for the last 35 lots; however, activity levels for specific isotopes ($^{95}\text{ZrNb}$, ^{103}Ru and $^{106}\text{RuRh}$) were documented for the last 52 lots in the data package. The average $^{95}\text{ZrNb}$ activity was 4.58 $\mu\text{Ci/lb}$ of uranium with a range of 1.23 to 38.76 $\mu\text{Ci/lb}$. It should be noted that the data is fairly consistent except for two entries that appear to be calculation errors that are off by a factor of ten. If these two entries (38.76 and 20.18) are corrected, the average activity becomes 3.56 $\mu\text{Ci/lb}$ of uranium and the range is 1.23 to 7.04 $\mu\text{Ci/lb}$. The average ^{103}Ru activity was 0.29 $\mu\text{Ci/lb}$ and the range was from unmeasurable to 2.79 $\mu\text{Ci/lb}$. For $^{106}\text{RuRh}$, the average activity was 0.44 $\mu\text{Ci/lb}$ of uranium and the range was from unmeasurable to 1.78 $\mu\text{Ci/lb}$.

This material whose analytical results are presented in Table 4-3 and some LEU was shipped by truck and rail to Paducah between 1969 to 1971. Gamma isotopic data (where available) are presented in this table in addition to the beta and gamma percentages (although a direct correlation can not be drawn without the calculations used to report the beta and gamma percent of aged natural uranium). UO_3 produced

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prior approval from the receiving site, very few if any of the lots with unrecovered data exceeded the 10 ppb threshold. Using the LEU data from 1962 through 1967, when only REDOX was processing LEU spent fuel, it is obvious that the REDOX process was well able to produce the UNH within the specification limit. Likewise using the analyses of depleted fuel from 1964 forward, PUREX was also capable of meeting the UNH plutonium specification. Consequently, there is a basis for predicting with assurance that nearly all shipments of UO_3 met the plutonium specification.

4.4.5 UO_3 Processed In/After 1984 Restart of PUREX/ UO_3 Plants

When the PUREX and UO_3 operations resumed in 1983, after about 10 years of stand-down, the UO_3 process support analyses were performed for the three impurities Pu, ^{237}Np , and ^{99}Tc and results of these analyses are summarized in Table 4-4. There is less consistency in these data because the measurement systems were inactive for the extended down period from the previous operating period. These data were also generated by two different laboratories on site. Data that has been found is included in Table 4-4 in its entirety. Data recorded in Table 4-5 is a special processing of 177 drums of UO_3 that is atypical in the ^{235}U content, but is included for completeness.

4.5 Neptunium-237

4.5.1 Neptunium Concentration in Recycled Uranium

From the earliest records of uranium production at the UO_3 facility, plutonium analyses were required, however the same was not true for neptunium. Production of ^{237}Np in the Hanford reactors was modeled based on reactor power levels and uranium isotopic data.

4.5.2 Neptunium-237 Formation

^{237}Np was formed in the Hanford production reactors by several possible neutron capture reactions in uranium. In natural uranium, the formation of ^{237}Np was due to two distinct reactions:

1. $^{238}\text{U} (n, 2n) \rightarrow ^{237}\text{U} (\beta) \rightarrow ^{237}\text{Np}$
2. $^{235}\text{U} (n, \gamma) \rightarrow ^{236}\text{U} (n, \gamma) \rightarrow ^{237}\text{U} (\beta) \rightarrow ^{237}\text{Np}$

The generation of ^{236}U in uranium recovered for recycle, materially added to the production of ^{237}Np . The ^{236}U reaction [Nilson 1961, Gestson 1967] was:

3. $^{236}\text{U} (n, \gamma) \rightarrow ^{237}\text{U} (\beta) \rightarrow ^{237}\text{Np}$

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installation of the Neptunium Recovery System and the Neptunium Purification System. During this cyclical operations mode, the ^{237}Np concentration in the recovered uranium varied by as much as an order of magnitude. Data presented in Table 4-4 shows this variability with concentrations ranging from 20 to 490 ppb.

The ^{237}Np is separated from the uranium by taking advantage of the relatively large differences in extractability of the two elements. The separation is further enhanced by deliberately saturating the organic solvent with uranium and maintaining a controlled uranium "loss" to the aqueous waste corresponding to about five percent of the input to the system. The organic uranium product stream normally contains less than five parts of plutonium and 100 parts of ^{237}Np per billion parts of uranium [Matheison 1968].

Analytical measurements of ^{237}Np in UNH were not routinely performed at the UO_3 Plant prior to 1969. Initial analyses were performed on the REDOX and PUREX UNH before shipment to the UO_3 for conversion of the UNH to UO_3 . Neptunium concentrations in the UO_3 powder shipped offsite were not routinely reported until the mid-1980s.

At the Paducah GDP, measurements were made after FY 1957 on monthly composite samples of received UO_3 [Smith 1984]. The average concentration of ^{237}Np in uranium oxide received from Hanford and Savannah River prior to FY 1967 was 240 ppb with a range of 10 to 600 ppb. For Hanford material after FY 1967 the average concentration of ^{237}Np was 120 ppb with a range of 50 to 270 ppb. The concentration of ^{237}Np in recovered low enriched UO_3 from Hanford was slightly lower, 50 ppb with a range of 10 to 110 ppb.

In 1978, the UO_3 chemical flowsheet included a proposed threshold for the neptunium content of incoming UNH to the UO_3 Plant. The value is given in grams/gallon and includes a minimum uranium threshold of 2.12 M. By calculation, the allowed concentration is 210 parts ^{237}Np per billion parts uranium. Table 4-7 lists available neptunium concentrations measured in the incoming UNH from PUREX to the receiver tank at UO_3 Plant in 1969 and 1970. These values agree with the limited data generated and reported at Hanford on UO_3 product produced in 1985-1986.

Available data [Smith 1984] suggests that ^{237}Np concentrations in UO_3 remained within the same wide range of values before and after recovery of ^{237}Np was initiated in 1959 as a co-product. Recovery of the ^{237}Np occurred within approximately the same time period that the ^{236}U concentration in reactor fuels was enhanced. There was therefore little net effect on the quality of UO_3 product.

In addition, analyses were reported from Paducah [Ritter K/ETO-30 (no issue date)] reports average Np receipts by year. The estimated Np received at Paducah with UO_3 was 18.4 Kg from 1953-1976 (no receipts shown for the years 1965-1968 and 1971). The annual quantity for the years 1953-1956 was estimated rather than measured but from 1956-1976 the quantity was measured. Assuming the quantity of reactor tails received is reported in English tons, the average Np concentration from 1953-1964 is 239 ppb \pm 1ppb. From 1969-1976 the average Np concentration varies from 11 ppb to

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ppm implied specification but above the proposed specification of 0.4 ppm. Special studies were performed in 1964 in which ^{99}Tc concentrations were found to be $1.74\text{E}5$ d/m/gram in PUREX-produced UNH and $2.9\text{E}4$ d/m/gram in REDOX UNH. This calculates to 450 ppb in PUREX and 760 ppb in REDOX UNH [Christy 1964]. The range of ^{99}Tc concentrations in the UO_3 in T-Hoppers currently stored at Hanford is between 2.3 to 12.4 ppm. In a review of the radiological effects of a UO_3 release scenario in the interim safety basis [Goldberg 1998], it was noted that ^{99}Tc was not included in the original source term calculations. A concentration of 0.001 weight percent ^{99}Tc "supplied by the customer" was ascribed to the stored UO_3 , consistent with the measured values.

4.6.3 Paducah Measurements of Technetium-99 in Recycled Uranium from Hanford

Measurements on "depleted reactor tails" received from Hanford were made at Paducah from 1959 onward [Smith 1984]. The ^{99}Tc data clustered in the range of 4 to 10 ppm on a uranium basis. The ^{99}Tc average was 7 ppm $\pm 30\%$. The few analyses reported for recovered enriched uranium showed an average ^{99}Tc concentration of 16 ppm with a range of 11-27 ppm.

4.6.4 Fernald Measurements of Technetium-99 in Recycled Uranium from Hanford

Measurements at Fernald on UO_3 lots from Hanford shipped in the 1980s are consistent with measurements at Hanford and Paducah and are in the range of 3 to 12 ppm [Lower 1995].

4.7 Uranium Isotopic Composition

4.7.1 Natural Uranium

Natural uranium contains three isotopes ^{238}U , ^{234}U , and ^{235}U which are present in the weight percentages 99.28 %, 0.005 % and 0.711% respectively. Irradiation of uranium in the Hanford reactors resulted in the generation of other uranium isotopes, in particular ^{236}U and ^{232}U . The preponderance of the uranium irradiated in the Hanford reactors was natural or normal and the remainder was low enriched uranium (LEU) primarily 0.94 wt% or 1.25 wt% ^{235}U . In the context of this document, natural uranium is uranium that has not been irradiated. "Normal" uranium is uranium that has been through a nuclear reactor and recovered from the spent fuel, but contains approximately the same concentration ^{235}U as occurs in nature. This ^{235}U concentration is attained either by blending uranium of different isotopic compositions or by processing in a GDP. Until normal U entered the metal fabrication process, reactor generated fission products would not be present in the fuel fabrication operations. It is believed that "normal" uranium was not received at Hanford for fuel fabrication before the start up of National Lead of Ohio. One year after UO_3 shipments from Hanford, the cascade feed at K-25 was composed almost entirely of reactor depleted uranium and therefore the quantities of normal uranium hereinafter will almost surely vary from theoretical isotopic ratio of 0.711% ^{235}U . In a letter [Gifford 1963], a statement is made that "...the next billets to be

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received from the feed material sites for NPR fuel elements will be recycle material." These billets were being produced at NLO.

4.7.2 Normal Uranium

Normal uranium received for reactor fuel tended to have a low concentration of ^{236}U , the ^{235}U concentration was depleted to only around 0.64 wt% due to the short irradiation time. The ^{236}U isotope and concentration did not build up very rapidly. In the gaseous diffusion process, the ^{235}U isotope is partially separated from other U isotopes and blended to produce the desired product isotopic composition.

4.7.3 Low Enriched Uranium (LEU)

LEU shipped to Hanford for fabrication into fuel elements was recycled uranium. The recycled uranium contained varying amounts of ^{236}U depending on the number of times it had been recycled through the reactors and the exposure time in the reactor. From 1975 through 1979, the 0.94% enriched fuel contained ^{236}U mostly in the range of 400 to 500 ppm and the 1.25 % enriched fuel contained between 350 to 400 ppm. From 1981 through 1986 the ^{236}U was mostly between 500 and 600 ppm for 0.947 enriched fuel and 500 to 800 ppm for 1.25% enriched fuel [Schmitroth 1995].

4.7.4 Typical Recovered LEU Uranium Oxide

The approximate isotopic content of LEU uranium oxide from N-Reactor operations contained 79 -154 ppb ^{232}U , 0.009 wt% ^{234}U , 0.88 wt% ^{235}U , and 0.07 wt% ^{236}U [Millward 1993]. Based on analytical measurements taken between 1972 and 1988, the recovered LEU contained an average of 0.0093 wt% ^{234}U with a range of 0.008 to 0.011 wt % ^{234}U , 0.860 wt% ^{235}U with a range of 0.748 to 0.957 wt% ^{235}U , and 0.071 wt % ^{236}U with a range of 0.06 to 0.08 wt% ^{236}U . Table 4-8 presents typical uranium isotopic distributions of LEU UO_3 product.

Table 4-9 shows a significant decrease in the ^{236}U concentration in depleted uranium in the ^{235}U concentration. These three lots are much different in isotopic content and would have been classified as depleted by the GDPs. They are atypical of normal production at Hanford. These three lots represent 177 fifty-five gallon drums that were in the Hanford Site Inventory in 1992 but have since been buried [Salley 1992].

Measurements of the uranium isotopic content of the recovered UO_3 produced at the UO_3 Plant were made on every lot of material shipped from Hanford. As shown in Table 4-10, the average ^{235}U concentration of the depleted UO_3 over the time period of 1952 through 1971 was 0.645 wt%, with a range of 0.62 to 0.68 wt% based on currently available data.

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Area Spectrographic Laboratory from those included in the methods compendium HW-24403 and in reports of the development documents [Daniel 1952]. This method development improved the measurement detection limits of three elements and added six elements that could be detected. It should be noted that the elements of concern were for those non-radioactive constituents that might perturb the nuclear reactivity of the fuel in the reactor, the physical characteristics of the fuel during fabrication operations, or form impurity inclusions in the fuel.

The importance of adhering to these critical specifications is apparent from the extensive "round robin" acceptance test program implemented in May 1951. This program was recommended by the Chief, Metal Branch, Production Division, NYOO, to the Hanford Operations Office (HO) in Richland [Morgan 1951]. These verification tests included selection, by the St. Louis Area Office, of two consecutive production lots (16 heats) each week. Samples from each of the 16 heats were to be sent to New Brunswick Laboratory (NBL), an AEC operated referee laboratory, where complete and precise analysis for all elements of concern was to be performed. The NYOO would collect the sample identification and coordinate shipment of the samples to Hanford where they would be analyzed. Data from both sites (NBL and HO) were to be collected and evaluated by the NYOO. "When sufficient information has been obtained to enable a reliable correlation to be made of the results, chemical analyses, and bare slug tests, an acceptance plan based on functional testing will be devised" [Morgan 1951].

Several letters issued in 1953 have been reviewed (between C. L. Karl and others) that address uranium metal specifications and uniformity of metal composition. Blending of feed materials (scrap, virgin derbies, and briquettes) into the metal appeared to have value rather than relying on one feed source alone. Although routine sampling and testing protocols were established with assistance from the Hanford Research Division during this period, the Hanford Site accepted the shipper's data for impurities. No records have been found that indicate that routine impurity analyses were performed after the uranium metal was received at Hanford for fabrication as fuel.

4.8.2 Metallographic Testing

Extensive micrographic examinations were performed on the uranium metal after fabrication into fuel elements to evaluate heating, rolling, and quenching effects on the grain size and orientation. These metallographic examinations of uranium metal and uranium compounds were performed in the onsite laboratory facilities during the 1950s [Bach 1950, Hartcom 1954, Gardner 1956].

4.8.3 Radiological Contaminants

Prior to 1952, no uranium oxide was recycled and as a result, Pu, ²³⁷Np, ⁹⁹Tc and fission product contamination were not present in the metal received for fuel fabrication. Between 1952 and 1962, UO₃ was processed through the gaseous diffusion plants, which significantly reduced the concentration of Pu and ²³⁷Np in the enriched product to

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levels reported to be in the parts per trillion. Special test measurements performed at Oak Ridge in 1973 estimated that approximately 85 % of ^{99}Tc received with recycle UO_3 is vaporized to the gaseous diffusion cascade. Measurements of the cascade tails suggests that essentially all ^{99}Tc entered the cascade. In 1963, a trap was installed to reduce the ^{99}Tc concentration in the cascade product, and the concentration was reduced in the enriched U fraction from an average of 3.2 ppm to an average of 0.15 ppm. Routine ^{99}Tc measurements were not initiated at Paducah until fiscal year 1972. In fiscal year 1974 the average ^{99}Tc concentration peaked at about six ppm and in 1982 the Paducah GDP product averaged below the detectable level of 0.01 ppm [Smith 1984].

It can be assumed that the ^{99}Tc concentration in U metal received at Hanford between 1953 and 1963 varied proportionally with the content of ^{99}Tc in the recycled UO_3 . After 1963, the ^{99}Tc content of the U metal would have been lower, but dependent on the effectiveness of the trap installed to remove ^{99}Tc in the GDP cascade.

4.8.4 Direct Blending

In 1962, the Fernald Plant proposed blending LEU oxide from Hanford with oxide containing 1.2% ^{235}U produced from UF_6 from Paducah to produce metal for reactor fuel [Keller 1962]. This process was to supercede generating the desired ^{235}U content within the GDPs. This direct blending of UO_3 containing the recycled radioisotopes would be expected to increase the radionuclide content of metal returned to Hanford from that produced only from GDP product. Since metal was produced by blending GDP enriched U and recycled UO_3 , the radionuclide content of metal could not exceed the radionuclide content in the UO_3 , since there were no concentration processes in the metal production.

The metal production site, Fernald, maintained a maximum acceptable concentration of 10 ppb for Pu, even though this was not included in the Hanford metal specifications. Concentrations of the Pu, ^{237}Np , and ^{99}Tc were not routinely monitored at Hanford on the received metal. One set of data (Transuranic Analyses for 0.95% ^{235}U Enriched Ingot Composites) is presented in Table 4-11. These data reflect composites of metal in the Hanford inventory after Hanford reactor operations ceased.

Although these data are incomplete, they indicate that U metal, even when produced by direct blending, remains below the 10 ppb in Pu content and the Np content is within the range of concentrations documented on the UO_3 product. The ^{99}Tc concentrations appear to be lower than the accepted concentrations values on the limited UO_3 data available.

Metal produced from natural uranium or normal uranium which has been processed through a GDP could be expected to have much lower concentrations of Pu and ^{237}Np . During the 10 year period 1972 through 1982, Smith [Smith 1984] reports average values of ^{99}Tc in Paducah GDP product from <0.01 to 6.1 ppm. That document also

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Hanford did not routinely measure the uranium metal receipts for the amounts of Pu, Np, and Tc constituents. Since Hanford has very little data on the constituent concentration in the uranium metal received for fuel fabrication, it was necessary to estimate concentration ranges of Pu, Np, and Tc in the received metal for this historical review, based upon data presented in Table 4-12, the Smith 1984 document, and communication with Fernald personnel during the course of this review. For the purpose of providing a rough estimate of the amount of constituents in the metal received at Hanford for fuel fabrication, the following ranges of constituents were used: Pu range of 0.01 - 6 ppb, with a mean of 3 ppb; Np range of 3 - 10 ppb, with a mean of 6.5 ppb; Tc range of 0.01 - 6 ppm, with a mean of 3 ppm. It is recognized that the selected range will have a significant impact on the amounts of constituents received, however refinement of these ranges would require a more thorough analysis of historical Hanford data in conjunction with an analysis of available analytical data from those sites who shipped uranium to Hanford. The ranges listed above were utilized in Tables I-12, I-13, and I-14 to estimate potential quantities of constituents in recycled uranium received at Hanford.

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Section 5 Current Inventory of Recycled Uranium

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(COEI) descriptors. As the table indicates the recycled uranium is stored at the 300 Area Fuel Fabrication complex and the UO₃ Plant.

5.2 Pacific Northwest National Laboratory Current Inventory of Recycled Uranium

Battelle Memorial Institute (BMI), assumed management of the Pacific Northwest National Laboratory (PNL) research and development activities for Hanford in 1965. PNL later received national recognition and became the Pacific Northwest National Laboratory (PNNL). One of the significant projects of the 1960s was PNL's design of the Fast Flux Test Facility (FFTF) in support of the Atomic Energy Commission's commercial nuclear power program. This project utilized uranium in the fabrication of mixed oxide fuel (MOX) containing uranium and fuel grade plutonium. The role of the PNNL in support of Defense missions has been significantly reduced from the 1960s. The narrative below provides a brief overview of the main PNNL project utilizing recycled uranium. The approximate 6.4 metric ton PNNL inventory is shown in Appendix D, Table 5.1.2. The receipts and shipments relating PNNL recycled uranium transactions with the three Major Tier 1 sites are shown in Section 3.2 and 3.3 tables.

The Kinetic Energy Projectile Project (KEPP) project is an ongoing project that started in 1974. This was an extension of work for others through Picatinny Arsenal, which supported other munitions prior to the depleted uranium (DU) phase. In 1974, the uranium used for PNNL research activities was DOE material. This feedstock started as green salt from Paducah, which was delivered to National Lead of Ohio (NLO/Fernald) for processing into the ingot stage. The ingots were then sent to Rocky Flats (RFETS) for alloying with titanium and processed into billet form. The billets were then shipped to PNNL for final processing and machining. Essentially all of the uranium involved in the project was and is DU alloyed with a small amount of titanium. This alloy is used because of its density and mechanical properties that provide strength to survive extreme launch conditions. After the research penetrators showed great promise for defeating current armor threats, production facilities had to be established. This led to facilities such as National Laboratory of Albany and NLO to produce production quantities. Continuing success led to the private sector getting involved through systems contractors such as Honeywell, etc. The initial private uranium producers were Nuclear Metals Inc and Aerojet Ordnance Company. Both private companies are involved in recycling of the penetrator alloy. The March 31, 1999 DU inventory at 306W building is approximately 4.9 metric tons with a storage limit in the 300 Area limited to 10 MTU. (In the past this limit has been 30 MTU.)

The receipts and shipments of normal, and enriched uranium into and out of PNNL were primarily for experimental fuel development of MOX fuel in support of the Liquid Metal Fast Breeder Reactor (Fast Flux Test Facility (FFTF)) program. The uranium itself was mainly supplied to Nuclear Energy (NE), via the DOE allotment process, from Defense Programs recycled production channels. As such, the entire PNNL uranium inventory, including DU, is considered in-scope.

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6.0 Discussion and Conclusions

Based on the review of the documents located, the RL Team concludes the following:

6.1 Recycled Uranium Shipped and Received

6.1.1 Operating History

Uranium recovery from irradiated fuel began at Hanford in 1952 using the REDOX process located in the 202-S Plant. Shortly thereafter, the U-Plant also began supplying uranyl nitrate hexahydrate (UNH) containing uranium recovered from the waste tanks. In 1956, the PUREX Plant came on line and also recovered uranium, as UNH, from irradiated fuel for calcination and recycle. The UNH product was piped from REDOX Plant and U-Plant and trucked from PUREX to the UO_3 Plant. At the UO_3 Plant the UNH was converted by calcination to UO_3 powder. The powder was sampled and packaged into either drums or specially designed "T-Hoppers" for shipment.

6.1.2 Recycled Uranium Specifications

Hanford received recycled uranium metal billets for reactor fuel rod manufacture starting about mid-1952; however, the recycled uranium used to produce these billets had been processed through the gaseous diffusion plants (GDPs) and was reported to contain approximately 30 parts plutonium per trillion parts uranium. After about 1963, as a result of a process change at Fernald involving blending, their metal billet plutonium specification rose to a not to exceed 10 parts plutonium per billion parts of uranium level. Hanford did not routinely perform a radionuclide analysis on the incoming billets, but relied on the shipper.

6.1.3 Recycled Uranium Shipments and Receipts

In all, Hanford shipped approximately **109,792** metric tons (MTU) of recovered (recycled) uranium. Of this, **74,491** MTU were shipped (as UO_3) to the Paducah GDP, **4,404** MTU were shipped to the K-25 GDP and Y-12 Plant, and **25,251** MTU were shipped to Fernald. Metal turnings and scrap produced during fuel rod manufacture were returned to Fernald for recovery into new fuel rod billets. Lesser quantities of recycled uranium were sent to Harshaw Chemical Co. and Mallinckrodt Chemical Works for further refining to remove non-radioactive contaminants. Additional recovered uranium, in minor quantities, was sent to over 100 other destinations to support various DOE missions.

Hanford received and processed approximately **109,144 MT** of recycled uranium, with approximately 85% (92,767 MT) being received from Paducah, Fernald and Oak Ridge. Uranium metal received for fuel fabrication before 1952 was made from natural uranium and is out of scope for this report. With the exception of the material remaining at Hanford, the majority of this material was used for fuel, irradiated in the Hanford

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reactors, processed in the separations facilities to recover the plutonium and uranium, and the recovered uranium shipped offsite for use or recycle.

6.1.4 Current Inventory

Excluding uranium in solid and liquid waste and releases to the cribs, Hanford currently has approximately **4,006 MTU** remaining in various forms including metal received for fabricating fuel, unused fuel, irradiated fuel, unirradiated uranium in mixed oxide fuel at the Fast Flux Test Facility and at the Plutonium Finishing Plant, and recovered uranium as UO_3 stored in T-Hoppers awaiting final disposition. The recycled uranium in the irradiated fuel (2,137 MTU) is outside the scope of this study.

~958 MTU is mixed with fission products and other chemical wastes in high-level waste storage tanks. **~1,054 MTU** is buried solid waste, and **~162 MTU** was released to the environment through various cribs, ponds, and ditches.

6.1.5 Shipper/Receiver Differences

During and after the May 17, 2000 Uranium Mass Balance Project workshop at Oak Ridge, TN, the RL team worked with other site representatives to compare shipping and receiving quantities of recycled uranium. For the Hanford shipments to Paducah, the percent variance between 1952 through March 30, 1999, was approximately 0.02% of the approximate 74,491 MT total. Percentage variances for shipments from Hanford to Fernald were approximately 0.7%. Percentage variances for Hanford receipts from Fernald were approximately 0.2%. Comparison between Hanford and Oak Ridge for shipments and receipts to the Oak Ridge aggregate of K-25 and the Y-12 will not be completed until the Y-12 transactions are prepared. The percentage variance for Hanford shipments to Savannah River (DuPont) were approximately 0.3%.

6.1.6 Inventory Difference

Hanford uranium shipments, receipts, and material in storage, and waste records indicate a small material difference of about **0.5 wt%** of the uranium received remains unexplained based upon the reviewed records. The calculation of this material difference includes an estimate that **~140 MTU** was consumed during reactor operations and the generation of plutonium. The material difference resides largely in the uncertainties associated with quantities of uranium in liquid and solid wastes in the waste tanks, and in the estimate of uranium fissioned and transmuted to operate the reactors and generate plutonium.

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6.2 Typical Impurities and Isotopic Composition

6.2.1 Plutonium

Hanford participated in development of the recycled UO_3 product specification starting in about 1950. By 1952, before commencement of uranium recovery operations, a preliminary specification requirement of less than 10 parts Pu per billion parts uranium was established. This limit was firmly established in 1953 and remained in place for the entire UO_3 production period. The bulk of Hanford's shipments of recycled UO_3 powder to the GDPs contained only trace amounts (<10 ppb) of Pu. Based on the limited amount of historical documentation located, it still appears the preponderance of Hanford recycled UO_3 powder contained ≤ 5 ppb. Six shipments were identified in Hanford documents as containing about 12, 13, 16, 19, 22, and 30 ppb average, respectively; however, records indicate receiver sites were typically notified of these out-of-specification conditions and accepted these materials prior to their shipment from Hanford. These shipments represented ~ 193 MTU containing ~ 3.4 g Pu. Information from Oak Ridge indicates that their site received four shipments from Hanford for which their analyses indicated Pu concentrations of 13, 17, 17, and 28 ppb. These shipments are in addition to the shipment containing 22 ppb mentioned above. Hanford analyses on these four shipments indicate the Pu concentrations were all <10 ppb. Available documentation indicates that Hanford and Oak Ridge both acknowledged this discrepancy. This is further discussed in Section 6.4.3. These four shipments totaled ~ 123 MTU and contained ~ 2.3 gPu. The limited analytical data located is insufficient for RL to determine the total quantity of plutonium shipped to the various GDPs; however, it appears that approximately 110 to 550 grams of plutonium were shipped with the 109,792 MTU, based upon an assumed average plutonium concentration range of one to five ppb. The mean of this range of Pu concentrations is 330 grams of Pu.

Hanford did not routinely analyze the incoming uranium metal for radioactive constituents, but relied on the shipper's guidelines for the metal product. It appears, based on the information available, that metal received prior to 1963 had been processed through the Paducah GDP and contained on the order of 10 parts or less plutonium per trillion parts uranium. Metal that was received in 1963 and beyond had been directly blended at Fernald and contained <10 ppb Pu.

6.2.2 Neptunium and Technetium

Hanford did not routinely analyze the UO_3 product for neptunium or technetium before 1980, as there were no related product specification requirements. For Hanford recycled UO_3 powder, technetium and neptunium limits were not considered required. Until 1967, Hanford analyzed the recycled UO_3 powder for total beta and gamma activity and conformed to the required specification levels of less than 100% of the beta activity of aged natural uranium and less than 300% of the gamma activity of aged natural uranium, respectively. In 1967, the beta and gamma measurements were dropped in favor of specific isotopic measurements ($^{95}\text{Zr/Nb}$, ^{103}Ru , and $^{106}\text{RuRh}$) as

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discussed in Section 4.3.2. Information provided by Fernald indicates that metal produced from recycled uranium contained ^{99}Tc ranging from 3 - 12 ppm and ^{237}Np with an upper bound of 500 ppb.

No uranium processing activities at Hanford have been identified that would increase the plutonium component of the recycled uranium after it was separated in the reprocessing plants. In the course of this study, no documentation has been found that reports concentration of constituents in areas accessed during maintenance activities. However, time has not been available to perform a detailed assessment of all uranium processing steps utilized at Hanford to assure that impurities in uranium were never concentrated. The UO_3 Plant calciners, which converted the recovered UNH to UO_3 powder did operate at elevated temperatures in an oxidizing environment. It is reasonable to believe that any volatile fission products which may have been present in the recycled uranium could have been released through the off-gas system, plated out on equipment surfaces, accumulated in off-gas scrubber solutions, or have been released to the environment. Documentation has been found to indicate that some of the ruthenium volatilized during UNH calcination, with decontamination factors (DF) ranging from <1 to 6 (see Appendix G, Section G.5). No operations in the fuel fabrication processes have been identified which would be expected to have further concentrated the constituents, other than the burning of metal fines to uranium trioxide.

6.3 Activity Assessment and Occupational Potential Exposure

The DOE-HQ mass balance project is a Department-wide effort to review each site that was involved with recycled uranium to provide an estimate of specific activities involving recycled uranium, and to develop a preliminary estimate of the approximate number of employees whose work subjected them to potential exposure from the constituents in the recycled uranium. The estimate of occupational potential exposure (OPE) is based on guidance developed during a workshop meeting held at the ORNL in May, 2000. This OPE criteria is listed at the bottom of Table 6-1. The recycled uranium activity and OPE assessment for Hanford is provided below.

The operations, maintenance and waste handling operations of the facilities described in Section 2 contributed to some personnel exposure and environmental releases. However, distinguishing any such personnel exposures to trace quantities of transuranics and fission products in recycled uranium from those associated with other Hanford operations which involved the handling and processing of significant quantities of irradiated fuel, high-level waste, and plutonium, would be very difficult. Assessment of personnel uranium exposure is further complicated by the practice of transferring personnel between facilities to meet ongoing work needs. The facilities at Hanford that had the highest potential for uranium exposure were the fuel fabrication facilities where large amounts of uranium metal and scrap were handled, and the UO_3 Plant, which handled large quantities of dry UO_3 powder. Other facilities involved with the handling of separated recycled uranium (as UNH) and of any waste from recycled uranium processing also had the potential for contributing to some exposure.

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Time was insufficient to complete an in-depth activity and potential worker exposure assessment. However, a cursory estimate, utilizing assumptions of direct labor and maintenance staffing at the facilities of interest, was completed to provide an overview of recycled uranium work activities in these facilities and an OPE estimate.

The facilities that were identified to have involved the handling of recycled uranium included: (1) the 313-314 Building complex in the 300 Area which was primarily involved with the fabrication of aluminum-clad reactor fuels, (2) the 333 Building complex in the 300 Area which was involved in the fabrication of zirconium-clad reactor fuels, (3) the REDOX Chemical Separations Plant in the 200 West area where reactor fuels were dissolved for plutonium and uranium recovery (including transfer of the UNH to the UO₃ Plant for calcination), (4) the U-Plant in the 200 West area where pre-1952 tank wastes were processed for uranium recovery, (5) the UO₃ Plant in the 200 West area where uranium recovered as UNH was received, concentrated, calcined, and packaged for shipment and recycle, (6) the PUREX Plant in the 200 East area where irradiated fuels were dissolved for separation of plutonium and uranium (with the UNH being shipped by truck to the UO₃ Plant for calcination), and (7) the 183-H solar basin in the 100-H area that was used for a ten-year interval to evaporate dilute liquid wastes generated at the 300 area fuel fabrication plants.

The interval of operation for each of these facilities is illustrated in Figure 6-1, along with an estimate of probable direct labor staffing support. The staffing calculations were based upon estimates of the number of operators and craft personnel per shift, the number of shifts per week, and an estimate of the average length of time that an employee would work at the facility. This information was used to estimate the total number of people who worked at each facility during its operating lifetime. These staffing totals, by facility, were then used as inputs to Table 6-1 to allow an estimate of the number of people likely to have been exposed to various levels of constituents of recycled uranium. Below is a typical staffing estimate calculation for Figure 6-1:

As an example, consider the 313-314 Building complex which had a PQ operations shift schedule and operated for 25 Years. It is estimated that the following operational information would apply to these facilities:

25 Operators/Shift; 10 Crafts/Shift; and an average Employee Time at Facility of ~5 years.

Thus, the number of workers on any given shift that may have been in contact with uranium would be 25 fuel fabrication operators plus 10 maintenance craftsmen for a total of 35. The "PQ" shift arrangement provided for Monday through Friday coverage of the 8AM to 4PM shift and the 4PM to Midnight shift. If the average worker remained at the job for 5 years then,

$$(35 \text{ workers/shift}) \times (2 \text{ shifts/day}) \times \frac{25 \text{ years facility operation}}{5 \text{ years at that facility for a typical worker}}$$

= 350 involved employees at that facility over the lifetime of the facility.

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shown in Figure 6-1 to arrive at an estimate of the number of personnel who could have had some level of OPE. All of this information is presented in Table 6-1.

The data summarized in Table 6-1 indicates that a total of about 1,126 people are estimated to have worked in areas directly involving recycle uranium during the operational time of the Hanford Site. Using the given OPE assessment criteria, of the ~1,126 staff personnel, it is estimated that ~670 had "no significant exposure potential" while ~456 had "moderate occupational exposure potential." It should be noted that this is only a rough estimate based upon limited data, engineering judgements and assumptions, and the application of some broad general OPE criteria which were established specifically for the mass balance project.

Since 1946, Hanford has had formal personnel monitoring programs in place which are designed to identify uptake of radioactive materials by personnel. Any employee who was assigned to a work location where contact with radioactive materials was judged to be possible was required to participate in the bioassay program. Each decision/request for bioassay evaluation on an individual or group of people represented a rather conservative contemporary judgement for protection of employees from radioactivity. Not all of those who were placed on the uranium bioassay program were expected to have direct contact with the radioactive material. It is therefore reasonable to suggest that the number of employees sampled on the uranium-specific bioassay program represents a conservative and upper bounding estimate of the number of individual workers potentially exposed to recycled uranium at Hanford.

Site records show that more than 50,000 employees were hired at Hanford over the operating period of the installation. In contrast, since the sampling program was established in 1946, only ~4,200 Hanford employees have been subjected to uranium-specific bioassay sampling and evaluation. A preliminary review of the records of that program was conducted. Those records (which included estimates for the constituents of interest) identified no significant doses associated with the recycled uranium for any of the Hanford Uranium Bioassay program participants. Due to the eligibility criteria applied, it is considered highly improbable that any substantial number of un-assayed site employees could have had any significant uranium uptake. For this study on recycled uranium traffic at Hanford, the very brief and somewhat empirical staffing and timing models for the facilities that handled large quantities of recycled uranium suggests that ~1,126 of the 4,200 employees did in fact perform substantial duty in the facilities that included opportunities for rather close contact with recycled uranium. Of that number, perhaps ~456 individuals had "moderate occupation exposure potential" as assessed and defined in the criteria that was given.

For a more specific analysis of worker contact with recycled uranium at Hanford than that contained in this report, an examination of Hanford's uranium bioassay records, a more detailed review of plant-by-plant operations, abnormal events, maintenance, and facility upgrades, overlaid by staffing models and production intervals would be required. Corroboration of such an analysis by examination of various records and

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interviews of retired employees would be necessary, in order to validate the OPE estimates.

6.4 Data Validation

6.4.1 Recycled Uranium Shipped from Hanford

External validation of Hanford shipment and receipt quantities of recycled uranium were performed, to the extent possible, during and after the May 17, 2000 workshop previously described in Section 6.1.5. For those sites identified as Major Tier 1 sites (Paducah, Fernald, and Oak Ridge) in which comparison was possible, the percentage variances between Hanford and other site data was below 1% (0.02-0.7%) for transaction categories detailed in Section 6.1.5.

6.4.2 Impurities Shipped with UO_3 from Hanford

As shown in Section 4, fairly complete analytical data for Pu contained in shipments of UO_3 have been located for the years from 1963 to 1988. Data on shipments prior to 1963 are limited for the constituents of concern including Pu, ^{237}Np , or ^{99}Tc . Hanford data for the ^{237}Np and ^{99}Tc were not available on a routine basis until 1984. In an attempt to reconstruct the quantity of constituents that were present in Hanford produced UO_3 , source documents from other sites were used to make the estimates, i.e. Smith 1984 and Ritter K/ETO-30 and draft U Mass Balance reports of receipts from the Major Tier 1 receiver sites. The Smith and Ritter documents report the result of special studies performed at Paducah using Hanford UO_3 as a starting material. Analyses were completed at these sites on hundreds of samples between 1959 to 1973 for Pu, ^{237}Np , and ^{99}Tc .

Engineering estimates of the quantities of constituents (Pu, Np, and Tc) which were present in recycled uranium received, shipped, contained in waste, released to the environment, or contained in the current recycled uranium inventory at Hanford have been made. The detailed results of these estimates are provided in Appendix I of this report. Analytical data on the concentration of Np and Tc in Hanford recycled uranium is minimal, since there were no specification requirements for these elements. Reasonable analytical data has been identified which indicates that recycled uranium shipped from Hanford typically contained Pu in the range of 1 to 5 ppb, and limited analytical data indicate Np ranged from 20 to 500 ppb, and Tc ranged from 3 to 12 ppm.

Since it is not possible to provide a complete historical assessment of the constituent levels in all recycled uranium Hanford, rough estimates of annual quantities of constituents in recycled uranium shipped each year were developed by assuming that the constituent levels during the years when analytical data is not available were the same as those for the years when analytical data is available. The annual estimates provided were based upon the low, mean, and high values for the constituent ranges mentioned above. Summaries of the total amount of constituents sent to Paducah,

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Fernald, and Oak Ridge sites are shown in Figures 6-2, 6-3, and 6-4. The ~110,000 MT of recycled uranium shipped from Hanford is estimated to have contained from ~110 to ~550 g Pu, ~2.2 to ~28 Kg Np, and ~330 to ~1,318 Kg of Tc. In general, these estimated constituent quantities appear to be consistent with data from sites receiving Hanford recycled uranium.

6.4.3 Discussion of Pu Data Differences Between K-25 and Hanford

In the K-25 Uranium Mass Balance Preliminary Site Report, analytical data are presented that indicate the Pu concentrations of UO_3 reported by K-25 between January and April 1953 are significantly outside the 10 ppb specification threshold. Hanford and Oak Ridge agree that data generated before and after the January to April 1953 time period are generally well within the 10 ppb threshold. Analysis of source documents are not available from Hanford for shipment composites or lots during this time period. However, monthly and weekly reports for this time period do not indicate that UO_3 shipments to Oak Ridge or Harshaw were made of out-of-specification UO_3 material in the Pu constituent. A reference [Schmidt 1953] recognizes that there was some discrepancy in the plutonium analytical results between Oak Ridge and Hanford. This reference also indicates other constituent data are in agreement. Communications were ongoing at that time to resolve the differences, specifically on shipments 18, 19, and 20. This reference states that Hanford reported less than 10 ppb and Oak Ridge reported concentrations in the range of 15 to 20 ppb on those three shipments. Both laboratories reported 22 ppb on the shipment composite number 43.

In 1952, Hanford was analyzing each lot of material but reduced the analytical workload in November 1952 by only analyzing the carload composite for Pu. The K-25 report apparently includes measurements of each lot. A lot represented eight drums, each drum contained about 300 Kg of UO_3 , and a car shipment included 11 or 12 lots (about 31-32 MTU) but ranged from 5 to 12 lots. Hanford transfer records show 411 MTU of UO_3 and metal scrap were shipped to Oak Ridge during the January to June 1953 time period. On the graphic in the K-25 draft report, approximately 45 -50 data points are shown during the January to April 1953 time frame. Assuming each point represents the results of one lot reported, this indicates four shipments were made and would have contained about 120 to 150 MTU. This is consistent with the weekly and monthly reports for the time period. This represents about three percent of the uranium sent to Oak Ridge. During the same time period Hanford shipped approximately four times more material to Harshaw Chemical Co. than to Oak Ridge. The Harshaw product was shipped on to Oak Ridge after processing to remove non-radionuclide constituents. The Oak Ridge report indicates the Harshaw material met the Pu specification.

The reference, cited above, indicates that Oak Ridge was using an analytical procedure that was not included in the authorized procedure manual of that time. They added an aluminum nitrate "salting agent" that could enhance the extraction of Pu into the organic extractant. Hanford was using the procedure in the manual and this could account for the discrepancy in the constituent concentration. It is evident that the Hanford results

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are biased low compared to the K-25 results. It is interesting to note that during the same time period that Oak Ridge reported receiving high Pu concentrations from Hanford, they also reported receiving slightly higher Pu levels from Harshaw than in preceding or succeeding periods. It seems that there would be a delay of a month or more in the receipt at Oak Ridge of Hanford produced UO_3 while processing was performed at Harshaw. The potential of the Harshaw process to reduce the Pu concentration in UO_3 product from Hanford is not known.

If Hanford data (less than 10 ppb for all shipments except shipment number 43 which Hanford reported as being 22 ppb) are used, about 4.5 grams of Pu would have been included with the UO_3 shipped during the January – April 1953 time period. If Oak Ridge's example data are extrapolated to the material shipped during this period and the average is 22 ppb, the Pu included with the UO_3 is 9 grams. This represents a difference of 4.5 grams of Pu in approximately **120 to 150 MTU** of uranium.

In 1948 and again in 1951 the concentration of Pu allowable in recycled UO_3 was established to limit the increase in potential hazard to personnel to no more than 10 percent of the hazard present from handling aged natural uranium in equilibrium with its decay daughters [Gamertsfelder 1948 and 1951]. The threshold established for Pu was 10 ppb and included a large conservatism factor. In 1985, a task force re-evaluated the 10 ppb limit against the then current air concentration guides and concluded that the 10 ppb value represented 3.5 percent of the concentration guide [DOE/OR-859 1985]. Therefore, with respect to personnel hazards from Pu impurities in recycled uranium, even the Hanford shipments which were above the specification of 10 ppb were well within the concentration guidance as established both in 1951 and again when the guidance was re-evaluated in 1985.

Section 6 Discussion and Conclusions

DOE/RL-2000-43

References:

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Appendix A

Glossary of Acronyms and Terms

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Appendix B

Section 3 - Supplemental Information

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Appendix B

Section 3 – Supplemental Information

Table 3.2.2 (Cont.) 1950 - FY 65 HANFORD (GE Company) URANIUM RECEIPTS FROM OFFSITE

MINOR TIER 1 SITE																MINOR TIER 1 SITE															
St Louis Area Office																Chicago Operations Office															
NY Ops																Argonne (ANL), Battelle Columbus(BML), Westinghouse															
FY	Date		REC	Box #	Doc #	Kg DU	Kg NU	Gm EU	Kg U Subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U Subtotal	FY																
1950	31-Dec-49	31-Dec-50	HGE	38213	FTS-670			0	0	1950		589	0	589	1950																
1951	31-Dec-50	30-Jun-51	HGE	38213	FTS-845		22	0	22	1951		384	4967	388.97	1951																
1952	01-Jul-51	30-Jun-52	HGE	38213	FTS 953		105	0	105	1952		28	88.4	28.09	1952																
1953	01-Jul-52	30-Jun-53	HGE	38213	FTS 1085			0	0	1953		1,571	0	1,571	1953																
1954	01-Jul-53	30-Jun-54	HGE	38213	FTS 1311			0	0	1954		813	2	813	1954																
1955	01-Jul-54	30-Jun-55	HGE	38213	FTS 1481			0	0	1955		702	1,007	703.01	1955																
1956	01-Jul-55	30-Jun-56	HGE	38213	FTS 1644			0	0	1956	21	1,188	1	1,209	1956																
1957	01-Jul-56	30-Jun-57	HGE	38213	FTS 1980			0	0	1957	14	12	5,122	31.12	1957																
1958	01-Jul-57	30-Jun-58	HGE	38213	FTS CLVI 463			0	0	1958		1,362	111,067	1,473.07	1958																
1959	01-Jul-58	30-Jun-59	HGE	38213	HAN 72720			0	0	1959	6	2,629	0	2,635	1959																
1960	01-Jul-59	30-Jun-60	HGE	38213	HAN 75996			0	0	1960			617	0.62	1960																
1961	01-Jul-60	30-Jun-61	HGE	38213	HAN 79125			0	0	1961	11	59	53,926	123.93	1961																
1962	01-Jul-61	30-Jun-62	HGE	38213	HAN 82406			0	0	1962	14	126	58,847	198.85	1962																
1963	01-Jul-62	30-Jun-63	HGE	38213	HAN 85615			0	0	1963		426	160,449	586.45	1963																
1964	01-Jul-63	30-Jun-64	HGE	38213	HAN 88957			0	0	1964	5	27	36,689	68.69	1964																
1965	01-Jul-64	30-Jun-65	HZA	38213	HAN 92119			0	0	1965	346	1	288,894	635.89	1965																
FY 53 thru 65 In-Scope subtotals						0	0	0	0		417	8,916	716,621	10,049.62																	
FY 1950 - FY 65 Total Kgs										127	11,055.68																				
FY 1950 - FY 65 Total MTUs										0.127	11.0557																				

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Appendix B: Table 3.2.4

Hanford Chemical Processing Contractor (HVA) Uranium Receipts FY 1971-March 31, 1999

Based on NMMSS Document SROO-00009 RECEIPTS (classified)

ATLANTIC RICHFIELD CONTRACTOR (HVA)

GM EU										
KG DU										
MAJOR TIER 1: FROM PADUCAH			MAJOR TIER 1: FROM FERNALD		MAJOR TIER 1: FROM OAK RIDGE		MAJOR TIER 1: FROM PADUCAH		MAJOR TIER 1: FROM FERNALD	
Kg DU			Kg DU		Kg DU		Kg DU		Gm EU	
FY	CKY, FYA	FVA, FVB, FVC	FZA, FZE, FZB, FZF	Hanford	External to	Hanford	CKY, FYA	FVA, FVB, FVC	Gm EU	FVA, FVB, FVC
71	0	0	0	171	5,863	0	0	0	0	42,320
72	0	0	0	0	43	0	0	0	0	0
73	0	0	0	0	106	0	0	0	0	0
74	0	0	0	0	1,165	0	0	0	0	0
75	0	1,165	0	1,165	1,162	0	0	0	0	0
76	0	1,162	0	1,162	0	0	0	0	0	0
76T	0	0	0	0	0	0	0	0	0	0
77	0	0	0	0	0	0	0	0	0	0
78	0	0	0	0	0	0	0	0	0	0
AFTER FY 1977 - HVA CONTRACTOR TURNOVER TO ROCKWELL HANFORD (HRA)										
79	0	0	0	0	0	0	0	0	0	0
80	0	0	0	0	0	0	0	0	0	0
81	0	0	0	0	0	0	0	0	0	0
82	0	0	0	0	0	0	0	0	0	0
83	0	0	0	0	0	0	0	0	0	0
84	0	0	0	0	0	0	0	0	0	0
85	0	0	0	0	0	0	0	0	0	0
86	0	0	0	0	0	0	0	0	0	0
87	0	0	0	0	0	0	0	0	0	0
88	0	0	0	0	0	0	0	0	0	0
89	0	0	0	0	0	0	0	0	0	0
90	0	0	0	0	0	0	0	0	0	0
91	0	0	0	0	0	0	0	0	0	0
92	0	0	0	0	0	0	0	0	0	0
93	0	0	0	0	0	0	0	0	0	0
94	0	0	0	0	0	0	0	0	0	0
95	0	0	0	0	0	0	0	0	0	0
96	0	0	0	0	0	0	0	0	0	0
97	0	0	0	0	0	0	0	0	0	0
98	0	0	0	0	0	0	0	0	0	0
99	0	0	0	0	0	0	0	0	0	0
KG TOTAL	0	2,327	0	8,510	0	8,510	0	0	0	42
MTU TOTAL	0.00	2.33	0.00	8.51	0.00	8.51	0.00	0.00	0.00	0.04

[illegible]

Appendix B:

Table 3.2.5

DOE/RL-2000-43

Hanford Chemical Processing Contractor (HRA) Uranium Receipts FY 1971-March 31, 1999

Based on NMMSS Document SROO-00009 RECEIPTS (classified) ROCKWELL HANFORD OPERATIONS (RHO)

KG DU				GM EU			
FY	MAJOR TIER 1:		MAJOR TIER 1:		MAJOR TIER 1:		MAJOR TIER 1:
	FROM PADUCAH	FROM FERNALD	FROM OAK RIDGE	Total Kg DU	FROM PADUCAH	FROM FERNALD	
	Kg DU	Kg DU	Kg DU	External to	Gm EU	Gm EU	
	CKY, FYA	FVA, FVB, FVC	FZA, FZE, FZB, FZF	Hanford	CKY, FYA	FVA, FVB, FVC	
71	PRIOR TO FY 1977 - HVA CONTRACTOR						
72							
73							
74							
75							
76							
77							
78							
79							
80							
81							
82							
83							
84							
85							
86							
87							
88							
89	AFTER FY 1988 - HRA CONTRACTOR TURNOVER TO WESTINGHOUSE HANFORD (HUD)						
90							
91							
92							
93							
94							
95							
96							
97							
98							
KG TOTAL	0	0	0	25,991	0	6,654	
MTU TOTAL	0.00	0.00	0.00	25.99	0.00	6.65	

[illegible]

Appendix B:

Table 3.2.6

Hanford Chemical Processing Contractors (HUD & HTA) Uranium Receipts FY 1971-March 31, 1999

DOE/RL-2000-43

Based on NMSS Document SROO-00009 RECEIPTS (classified)									
WESTINGHOUSE HANFORD (HUD) & FLUOR (HTA)									
DU					EU				
FY	MAJOR TIER 1:		MAJOR TIER 1:		Total Kg DU External to Hanford	MAJOR TIER 1:		MAJOR TIER 1:	
	FROM PADUCAH Kg DU	FROM FERNALD Kg DU	FROM OAK RIDGE Kg DU	FROM FERNALD Kg DU		FROM PADUCAH Gm EU	FROM FERNALD Gm EU	FROM FERNALD Gm EU	FROM FERNALD Gm EU
	CKY, FYA	FVA, FVB, FVC	FZA, FZE, FZF	FVA, FVB, FVC		CKY, FYA	FVA, FVB, FVC	FVA, FVB, FVC	
71									
72									
73									
74									
75									
76									
77									
78									
79									
80									
81									
82									
83									
84									
85									
86									
87	0	0	0	0	0	0	0	0	0
88	0	0	0	0	0	0	0	183,387	0
89	0	0	0	0	0	0	0	112,537	0
90	0	0	0	0	0	0	0	313,608	0
91	0	0	0	0	0	0	0	0	0
92	0	0	0	0	0	0	0	0	0
93	0	0	0	0	0	0	0	0	0
94	0	0	0	0	0	0	0	0	0
95	0	0	0	0	0	0	0	0	0
HUD UNTIL 96	0	0	0	0	0	0	0	0	0
HTA IN 97	0	0	0	0	7,812	0	0	0	0
98	0	0	0	0	62	0	0	0	0
99	0	0	0	0	0	0	0	0	0
KG TOTAL	0	0	0	0	7,876	0	0	610	0
MTU TOTAL	0.00	0.00	0.00	0.00	7.88	0.00	0.00	0.61	0.00

Table 3.2.6 HUD & HTA

[illegible]

Appendix B:

Table 3.2.7

DOE/RL-2000-43

Hanford Reactor/Fuel Fab Contractor (HXA) Uranium Receipts FY 1971-March 31, 1999

Based on NMMSS Document SROO-00009 RECEIPTS (classified)

UNITED NUCLEAR INC. (UNI)

DU		EU									
FY	MAJOR TIER 1: FROM PADUCAH		MAJOR TIER 1: FROM FERNALD		MAJOR TIER 1: FROM OAK RIDGE		Total Kg DU External to Hanford	MAJOR TIER 1: FROM PADUCAH		MAJOR TIER 1: FROM FERNALD	
	Kg DU CKY, FYA	Kg DU FVA, FVB, FVC	Kg DU FZA, FZE, FZB, FZF	Kg DU FZA, FZE, FZB, FZF	Gm EU CKY, FYA	Gm EU FVA, FVB, FVC					
71	0	51	0	0	51			0		332,107,792	
72	0	0	0	0	0			0		28,210,576	
73	0	0	0	0	0			0			
74	0	0	0	0	0			0			
75	0	0	0	0	167			0			
76	0	0	0	0	0			0			
76T	0	0	0	0	0			0			
77	0	0	0	0	0			0			
78	0	0	0	0	0			0			
79	0	0	0	0	0			0			
80	0	0	0	0	0			0			
81	0	0	0	0	0			0			
82	0	0	0	0	0			0			
83	0	0	0	0	0			0			
84	0	0	0	0	0			0			
85	0	0	915	11,085	0			0			
86	0	0	0	0	0			0			
87	0	0	0	0	0			0			
AFTER FY 1987 - HXA CONTRACTOR TURNOVER TO WESTINGHOUSE (HUE)											
KG TOTAL	0	51	915	11,303	0			0		360,318	
MTU TOTAL	0.00	0.05	0.92	11.30	0.00			0.00		360.32	

[illegible]

Table 3.2.8

Pacific Northwest National Laboratory (PNNL) Uranium Receipts FY 1965-March 31, 1999

Based on NMMSS Document SROO-00009 RECEIPTS (classified)

FY	MAJOR TIER 1			MAJOR TIER 1			DU			MAJOR TIER 1			MAJOR TIER 1		
	FROM PADUCAH			FROM FERNALD			FROM OAK RIDGE			FROM PADUCAH			FROM FERNALD		
	Kg DU	CKY, FYA		Kg DU	FVA		Kg DU	FZA, FZE, FZB, FZF	Total Kg DU (Excludes Hanford)	Gm EU	CKY, FYA		Gm EU	FVA	Gm EU
FY 65-FY 70		0	143	152	1,183						0		371,489		49,291
71	0	0	0	5	83					0	0		0	0	622,819
72	0	0	0	31	165					0	0		0	0	0
73	0	0	0	0	641					0	0		0	0	12
74	0	0	0	669	2,317					0	0		0	0	0
75	0	0	0	18	3,950					0	0		0	0	13,068
76	0	0	0	0	3,810					0	0		0	0	3,843
76T	0	0	209	0	1,742					0	0		0	0	0
77	0	0	797	87	19,431					0	0		0	0	52,779
78	0	0	548	84	6,205					0	0		0	0	0
79	0	0	265	13	16,694					0	0		0	0	10
80	0	0	1,256	0	5,936					0	0		0	0	17,451
81	0	0	957	13	13,750					0	0		0	0	2,060
82	0	0	0	0	35,912					0	0		0	0	0
83	0	0	35	0	8,063					0	0		0	0	0
84	0	0	0	8	13,355					0	0		0	0	0
85	0	0	0	29	12,909					0	0		0	0	0
86	0	0	0	0	18,492					0	0		0	0	0
87	0	0	0	0	11,764					0	0		0	0	0
88	0	0	0	0	8,223					0	0		0	0	0
89	0	0	0	42	20,043					0	0		0	0	0
90	0	0	0	1,033	20,574					0	0		0	0	0
91	0	0	0	8,956	12,354					0	0		0	0	0
92	0	0	0	1,316	7,627					0	0		0	0	0
93	0	0	0	182	182					0	0		0	0	0
94	0	0	0	0	216					0	0		0	0	0
95	0	0	0	0	924					0	0		0	0	0
96	0	0	0	0	0					0	0		0	0	0
97	0	0	0	0	1,618					0	0		0	0	0
98	0	0	0	0	0					0	0		0	0	0
99	0	0	0	0	0					0	0		0	0	0
KG TOTAL	0	0	4,210	12,638	248,155					0	0		371		761
MTU TOTAL	0.00	0.00	4.21	12.64	248.16					0.00	0.00		0.37		0.76

Table 3.2.8

Appendix B:

EU Total Gm EU PNNL External (Excludes Hanford)	MAJOR TIER 1 FROM PADUCAH		MAJOR TIER 1 FROM FERNALD		MAJOR TIER 1 FROM OAK RIDGE		NU Kg NU PNNL External (Excludes Hanford)		ALL U Total Kg U External Receipts	
	Kg NU CKY, FYA		Kg NU FVA		Kg NU FZA, FZE, FZB, FZF					
8,367,607	0	0	36	0	0	0	4060			
658,595	0	0	0	0	0	0	9			
725,784	0	0	0	0	0	0	107			
-11,993	0	0	0	0	0	0	0			
90,360	0	0	0	0	0	0	0			
392,115	0	0	0	0	0	0	17			
961,971	0	0	0	0	0	0	777			
3,942,113	0	0	0	0	0	0	10			
4,030,959	0	0	0	0	0	0	0			
3,597,476	0	0	0	0	0	0	0			
48,615	0	0	0	0	0	0	1			
534,954	0	0	0	0	0	0	0			
-10,549	0	0	0	0	0	0	0			
364	0	0	0	0	0	0	0			
4,016	0	0	0	0	0	0	0			
38,118	0	0	0	0	0	0	0			
11,369	0	0	0	0	0	0	0			
1,151,358	0	0	0	0	0	0	0			
75	0	0	0	0	0	0	0			
1,698	0	0	0	0	0	0	0			
56,819	0	0	0	0	0	0	0			
34,320	0	0	0	0	0	0	0			
0	0	0	0	0	0	0	0			
188	0	0	0	0	0	0	6			
560	0	0	0	0	0	0	0			
0	0	0	0	0	0	0	0			
2,280	0	0	0	0	0	0	0			
491,522	0	0	0	0	0	0	0			
6	0	0	0	0	0	0	0			
0	0	0	0	0	0	0	0			
27	0	0	0	0	0	0	0			
25,121	0	0	36	0	0	0	4,987		278,263	
25.12	0.00	0.00	0.04	0.00	0.00	0.00	4.99		278.26	

Appendix B: Table 3.3.1

DECEMBER 1947 THROUGH JUNE 51 HANFORD (GE Company)
URANIUM SHIPMENTS OFFSITE

All Shipments are Out-of-Scope (for reference use)

Time Frame	Date From	Date To	RIS From	Box #	Doc #	GE- Hanford Works Beginning Inventory		IN WASTE TANKS		Mallinckrodt Cher New York Operat
						BEG. INV. Kg NU	IN PROCESS Kg NU	IN WASTE TANKS Kg DU	SEE RECEIPTS	
1947	31-Dec-47	BEG. INV.	HGE	38213	HW-15-701	SEE RECEIPTS	SEE RECEIPTS	SEE RECEIPTS		
1947	31-Dec-47	31-Dec-48	HGE	38213	HW-15-701					
1948	31-Dec-48	31-Dec-49	HGE	38213	FTS-668					
1949	31-Dec-49	31-Dec-50	HGE	38213	FTS-670					
FY 50										
FY 1951	01-Jan-51	30-Jun-51	HGE	38213	FTS 845					
OUT OF SCOPE						Total Kg Shipped				
OUT OF SCOPE						Total MTU Shipped				

Appendix B:

Table 3.3.1

DOE/RL-2000-43

Simonds Saw & Steel Co.				Electro Metallurgical Chem Wks				Metal Hydri			
Simonds Saw & Steel Co.				Electro Metallurgical Chem Wks				Metal Hydri			
NY Ops				NY Ops				NY Ops			
Kg NU	Gm EU	Total Kg	Kg DU	Kg NU	Gm EU	Total Kg	Kg DU	Kg NU	Gm EU	Total Kg	Kg DU
		0.0				35,633.3				0.0	
77,868.0		77,868.0		35,633.3		170,963.0				0.0	
61668		61,668.0		212369		212,369.0				0.0	
		0.0				0.0				0.0	
12606		12,606.0		93045		93,045.0				0.0	
		0.0				0.0				0.0	
		0.0				0.0				0.0	
152,142.0				512,010.3				0.0			
152.142				512.01				0.00			

[illegible]

Table 3.3.1

[illegible]

Appendix B:

Table 3.3.1

DOE/RL-2000-43

Carbide & Carbon Chem Co. ORO Y-12 & K-25				Oak Ridge Natl Lab ORO				Clinton Laboratory									
Cyt		Kg DU		Kg NU		Gm EU		Total Kg		Kg DU		Kg NU		Gm EU		Total Kg	
Total Kg																	
0.0				273.8				273.8				478.4		165.0		478.6	0.0
22,048.0								0.0				191.0		9,505.0		200.5	0.0
0.0				5447		32,224		5,479.2				1499		803		1,499.8	0.0
0.0								0.0								0.0	0.0
0.0				447		64,008		511.0								0.0	0.0
0.0								0.0								0.0	0.0
0.0								0.0								0.0	0.0
22,048.0								6,264.0								2,178.9	0.0
22.05								6.26								2.18	0.00

Appendix B:

Table 3.3.1

DOE/RL-2000-43

GE Research Laboratory Schenectady Ops				Joslyn Manufacturing & Supply Co.				New Brunswick Lab				Sante Fe O			
SGE															
Kg DU	Kg NU	Gm EU	Total Kg	Kg DU	Kg NU	Gm EU	Total Kg	Kg DU	Kg NU	Gm EU	Total Kg	Kg DU	Kg NU	Gm EU	Total Kg
	1,102.2	0.4	1,102.2		201,914.0		201,914.0								0.0
	77.0		77.0				0.0								0.0
	1062		1,062.0				0.0								0.0
			0.0				0.0								0.0
	142		142.0				0.0								0.0
			0.0				0.0								0.0
			0.0				0.0								0.0
2,383.2				201,914.0				0.0							
2.38				201.91				0.00							

Appendix B:

Table 3.3.1

DOE/RL-2000-43

Los Alamos			Hanford Operations-Crane Company			Schenectady Operations Office			Savannah River El DuPont			
Kg NU	Gm EU	Total Kg	Kg DU	Kg NU	Gm EU	Total Kg	Kg DU	Kg NU	Gm EU	Total Kg	Kg DU	Kg NU
30,681.8		30,681.8				0.0				0.0		
6,804.0		6,804.0				0.0				0.0		
24941		24,941.0				0.0				0.0		
27280		27,280.0				0.0				0.0		315
		0.0				0.0				0.0		
		0.0				0.0				0.0		
		89,706.8				0.0				0.0		
		89.71				0.00				0.00		

Appendix B:

Table 3.3.1

DOE/RL-2000-43

Chapman Valve Manuf. Co.

Total Kg	Kg DU	Kg NU	Gm EU	Total Kg
428.5		24,129.7		24,129.7
0.0				0.0
0.0				0.0
0.0				0.0
0.0				0.0
0.0				0.0
0.0				0.0
428.5				24,129.7
0.43				24.13
				1,601.56

Total



1,601,562

Appendix B:

Table 3.3.2

DOE/RL-2000-43

FY 1952-65 HANFORD (GE Company) URANIUM SHIPMENTS TO OFFSITE LOCATIONS

MINOR TIER 1 SITE										MINOR TIER 1 SITE			
										Mallinckrodt Chem Works (MCW)			
										NY Ops - ORO Ops (IN FY55 Weldon Spring)			
FY	Date From	Date To	From RIS	Box #	Doc #	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg DU	Kg NU
1950	31-Dec-49	31-Dec-50	HGE	38213	FTS-670								
1951	31-Dec-50	30-Jun-51	HGE	38213	FTS-845								
1952	01-Jul-51	31-Dec-51	HGE	38213	FTS-864			0.00	0.00	1952			130,377.00
1952	01-Jan-52	30-Jun-52	HGE	38213	FTS 953		4,719.00	0.00	4,719.00	1952			326,377.00
1953	01-Jul-52	30-Jun-53	HGE	38213	FTS 1085		60,627.00	0.00	60,627.00	1953			438,066.00
1954	01-Jul-53	30-Jun-54	HGE	38213	FTS 1311		86,317.00	0.00	86,317.00	1954			
1955	01-Jul-54	30-Jun-55	HGE	38213	FTS 1481	3.00	33,536.00	0.00	33,539.00	1955			
1956	01-Jul-55	30-Jun-56	HGE	38213	FTS 1644		318.00	0.00	318.00	1956			
1957	01-Jul-56	30-Jun-57	HGE	38213	FTS 1980		154.00	0.00	154.00	1957			
1958	01-Jul-57	30-Jun-58	HGE	38213	FTS CLVI 463			0.00	0.00	1958			
1959	01-Jul-58	30-Jun-59	HGE	38213	HAN 72720	4.00	1,554.00	0.00	1,558.00	1959			
1960	01-Jul-59	30-Jun-60	HGE	38213	HAN 75996		138.00	0.00	138.00	1960			
1961	01-Jul-60	30-Jun-61	HGE	38213	HAN 79125		118.00	0.00	118.00	1961			
1962	01-Jul-61	30-Jun-62	HGE	38213	HAN 82406	5.00	611.00	0.00	616.00	1962			
1963	01-Jul-62	30-Jun-63	HGE	38213	HAN 85615			0.00	0.00	1963			
1964	01-Jul-63	30-Jun-64	HGE	38213	HAN 88957			0.00	0.00	1964			
1965	01-Jul-64	30-Jun-65	HZA	38213	HAN 92119				0.00	1965			
FY 52 thru FY 65 Kiloqram totals						12.00	188,092.00	0.00	188,104.00		0.00	0.00	894,820.00
FY 52 thru FY 65 MTU totals										188.104			
IN-SCOPE KG TOTALS						12.0	183,373.0	0.0	183,385.0		0.0	0.0	438,066.0

Appendix B: Table 3.3.2

MINOR TIER 1 SITE											
New York Operations Harshaw Chemical Co., Cleveland						New York Operations Massachusetts Institute of Technology					
MHR											
Gm EU	KgU subtotal	FY	Kg DU	Kg NU	Gm EU	subtotal	FY	Kg DU	Kg NU	Gm EU	subtotal
0.00	130,377.00	1952			0.00	0.00	1952			0.00	0.00
0.00	326,377.00	1952		11.00	0.00	11.00	1952			0.00	0.00
0.00	438,066.00	1953	1,674,970.00	0.00	0.00	1,674,970.00	1953		3,920.00	0.00	3,920.00
0.00	0.00	1954	238,736.00		0.00	238,736.00	1954		267.00	0.00	267.00
0.00	0.00	1955			0.00	0.00	1955			0.00	0.00
0.00	0.00	1956			0.00	0.00	1956			0.00	0.00
0.00	0.00	1957			0.00	0.00	1957			0.00	0.00
0.00	0.00	1958			0.00	0.00	1958			0.00	0.00
0.00	0.00	1959			0.00	0.00	1959			0.00	0.00
0.00	0.00	1960			0.00	0.00	1960			0.00	0.00
0.00	0.00	1961			0.00	0.00	1961			0.00	0.00
0.00	0.00	1962			0.00	0.00	1962			0.00	0.00
0.00	0.00	1963			0.00	0.00	1963			0.00	0.00
0.00	0.00	1964			0.00	0.00	1964			0.00	0.00
0.00	0.00	1965			0.00	0.00	1965			0.00	0.00
0.00	894,820.00		1,913,706.00	11.00	0.00	1,913,717.00		0.00	4,187.00	0.00	4,187.00
						1,913.717					
4.187											
0.00	438,066.00		1,913,706.00	0.00	0.00	1,913,706.00		0.00	4,187.00	0.00	4,187.00

Appendix B:

Table 3.3.2

DOE/RL-2000-43

MINOR TIER 1 SITE										MINOR TIER 1 SITE										MINOR TIER 1 SIT									
New York Ops Office										St Louis Area Office										Chicago Operations Of									
National Lead Company (MNL)										SubOffice to New York Ops										GE, Battelle Columbus									
Aeroprojects, Nuclear Metals Inc. (MIO)																													
FY	Kg DU	Kg NU	Gm EU	subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU												
1952			0.00	0.00	1952		14,355.00	0.00	14,355.00	1952					1952														
1952		90.00	0.00	90.00	1952		35,617.00	0.00	35,617.00	1952					1952														
1953		111,637.00	0.00	111,637.00	1953					1953					1953														
1954		372,669.00	0.00	372,669.00	1954					1954					1954														
1955			0.00	0.00	1955					1955					1955														
1956			0.00	0.00	1956					1956					1956														
1957			0.00	0.00	1957					1957					1957														
1958			0.00	0.00	1958					1958					1958														
1959	4,614.00	1,515.00	1,012,413.00	7,141.41	1959					1959					1959														
1960		1,172.00	0.00	1,172.00	1960					1960					1960														
1961		2,558.00	85,672.00	2,643.67	1961					1961					1961														
1962		2,161.00	87,116.00	2,248.12	1962					1962					1962														
1963			0.00	0.00	1963					1963					1963														
1964			0.00	0.00	1964					1964					1964														
1965		24.00	45,454.00	69.45	1965					1965					1965														
4,614.00 491,826.00 1,230,655.00 497,670.66										0.00 49,972.00 0.00 49,972.00										43.00 1,839.00									
497.671										49.972																			
4,614.0	491,736.0	1,230,655.0	497,580.7			0.0	0.0	0.0	0.0																				

MINOR TIER 1 SITE												
MINOR TIER 1 SITE												
Chicago Operations Office Iowa State College (sent uran slugs)												
Chicago Operations Office Argonne Nat Lab ANL												
Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	Kg U subtotal
2.00	28.00	1952		15.00	5,166.00	20.17	1952			0.00	0.00	0.00
0.00	119.00	1952		864.00	1,675.00	865.68	1952			0.00	0.00	0.00
0.00	267.00	1953		242.00	0.00	242.00	1953		196.00	0.00	196.00	196.00
2.00	0.00	1954	65.00	50.00	192,000.00	307.00	1954		95.00	0.00	95.00	95.00
1,007.00	237.01	1955	2.00		1,729.00	3.73	1955		155.00	0.00	155.00	155.00
287.00	3.29	1956	14.00	1,173.00	11.00	1,187.01	1956		170.00	0.00	170.00	170.00
1,187.00	85.19	1957	14.00		13.00	14.01	1957			0.00	0.00	0.00
1,222.00	303.22	1958			0.00	0.00	1958			0.00	0.00	0.00
47,096.00	115.10	1959			0.00	0.00	1959			0.00	0.00	0.00
0.00	33.00	1960			0.00	0.00	1960			0.00	0.00	0.00
2,894.00	78.89	1961		1.00	0.00	1.00	1961			0.00	0.00	0.00
60,160.00	196.16	1962			0.00	0.00	1962			0.00	0.00	0.00
179,309.00	689.31	1963			0.00	0.00	1963			0.00	0.00	0.00
18.00	20.02	1964	19.00	8.00	0.00	27.00	1964			0.00	0.00	0.00
677.00	0.68	1965	970.00		0.00	970.00	1965			0.00	0.00	0.00
293,861.00	2,175.86		1,084.00	2,353.00	200,594.00	3,637.59		0.00	616.00	0.00	616.00	616.00
2.176 3.638 0.616												
293,859.0	2,028.9		1,084.0	1,474.0	193,753.0	2,751.8		0.0	616.0	0.0	616.0	616.0
0.616												

MINOR TIER 1 SITE					MINOR TIER 1 SITE					MAJOR TIER 1 SITE				
Miscellaneous Site 2					New Brunswick Lab Oak Ridge Operations					National Lead of Ohio/Fernald Oak Ridge Operations				
Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	NBL Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	NLO Kg DU	Kg NU	Gm EU		
	121.00	377.00	121.38	1952		1.00	0.00	1.00	1952			0.00		0.00
	623.00	0.00	623.00	1952		312.00	0.00	312.00	1952			0.00		0.00
	697.00	0.00	697.00	1953			0.00	0.00	1953	124.00		0.00		0.00
392.00	1,037.00	1,433.00	1,430.43	1954			0.00	0.00	1954			0.00		0.00
	179.00	29.00	179.03	1955			0.00	0.00	1955		266,206.00	0.00		0.00
21.00	260.00	89.00	281.09	1956			0.00	0.00	1956		411,533.00	0.00		0.00
		67.00	0.07	1957	22.00		0.00	22.00	1957		348,388.00	462,208.00		
		7.00	0.01	1958	28.00	1.00	0.00	29.00	1958		359,711.00	5,497,057.00		
		0.00	0.00	1959	27.00		0.00	27.00	1959	1,363.00	489,893.00	17,736,122.00		
		0.00	0.00	1960	18.00	1.00	0.00	19.00	1960	18.00	362,114.00	20,546,711.00		
2.00		0.00	2.00	1961			0.00	0.00	1961		283,949.00	49,937,914.00		
		0.00	0.00	1962			0.00	0.00	1962		144,415.00	284,990,701.00		
		0.00	0.00	1963			136.00	0.14	1963		227,797.00	1,215,954,820.00		
		0.00	0.00	1964	1.00		109.00	1.11	1964		241,931.00	1,269,135,177.00		
		0.00	0.00	1965	1.00		979.00	1.98	1965		89,293.00	1,946,833,080.00		
415.00	2,917.00	2,002.00	3,334.00		97.00	315.00	1,224.00	413.22		1,505.00	3,225,230.00	4,811,093,790.00		
					3.334					0.413				
415.0	2,173.0	1,625.0	2,589.6		97.0	2.0	1,224.0	100.2		1,505.0	3,225,230.0	4,811,093,790.0		

Appendix B:

Table 3.3.2

DOE/RL-2000-43

MAJOR TIER 1 SITE

MAJOR TIER 1 SITE

Carbide & Carbon Chem Co. Oak Ridge Y-12 & K-25 GDP						Union Carbide of Kentucky-Paducah Oak Ridge Operations					
Kg U subtotal	FY	CYT & CCC		Kg NU	Gm EU	Kg U subtotal	FY	CKY		Kg NU	Gm EU
		Kg DU						Kg DU			
0.00	1952		70	0			1952				0.00
0.00	1952	154,394		16,238	0	170,632.0	1952				0.00
124.00	1953	557,145		46,405	21,356	603,571.4	1953				0.00
0.00	1954	1,147,291		28,508	289,605	1,176,088.6	1954	2,233,024.00			0.00
266,206.00	1955	498,888			519,689	499,407.7	1955	2,586,213.00			496,031.00
411,533.00	1956	289,124			69,259	289,193.3	1956	4,105,038.00			0.00
348,850.21	1957	98,083			680,000	98,763.0	1957	5,385,900.00			11,564.00
365,208.06	1958	8,598		14	489,870	9,101.9	1958	6,056,423.00			0.00
508,992.12	1959	82			288,203,113	288,285.1	1959	5,202,424.00			0.00
382,678.71	1960	15			610,585,416	610,600.4	1960	5,148,061.00			0.00
333,886.91	1961				614,923,511	614,923.5	1961	6,093,776.00			0.00
429,405.70	1962				46,799,845	46,799.8	1962	4,576,354.00			915,461,646.00
1,443,751.82	1963				1,571,464	1,571.5	1963	5,771,936.00			0.00
1,511,066.18	1964				5,230	5.2	1964	4,087,367.00			0.00
2,036,126.08	1965				3,283	3.3	1965				0.00
8,037,828.79		2,753,690		91,165	1,564,161,641	4,409,016.5		51,246,516.00		0.00	915,969,241.00

4,409.017

8,037.829

8,037,828.8	2,753,620.0	74,927.0	1,564,161,641.0	4,392,708.6	51,246,516.0	0.0	915,969,241.0
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Table 3.3.2

35

TIER 1 SITE					MINOR TIER 1 SITE					MINOR TIER 1 SITE				
lec. Products Ops					San Francisco Operations North Amer Aviation (DNA), Oregon Metallurgical					Bridgeport Brass Co. NY Ops then under ORO				
Kg NU	Gm EU	Kg U subtotal	FY		Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY
16.00	0.00	16.00	1952					0.00	1952			0.00	0.00	1952
234.00	0.00	234.00	1952					0.00	1952			0.00	0.00	1952
774.00	0.00	774.00	1953		14.00	37.00		51.00	1953			0.00	0.00	1953
196.00	0.00	196.00	1954				3.00	0.00	1954			0.00	0.00	1954
	0.00	0.00	1955			54.00		54.00	1955			0.00	0.00	1955
	0.00	0.00	1956		22.00	35.00		57.00	1956		87.00	0.00	87.00	1956
	0.00	0.00	1957			54.00		54.00	1957			0.00	0.00	1957
	0.00	0.00	1958		21.00	228.00		249.00	1958			0.00	0.00	1958
	0.00	0.00	1959		1.00	1,183.00	11,362.00	1,195.36	1959		2,096.00	0.00	2,096.00	1959
	0.00	0.00	1960		2.00	5,025.00	290,491.00	5,317.49	1960		1,542.00	0.00	1,542.00	1960
	0.00	0.00	1961			806.00	10,864.00	816.86	1961			0.00	0.00	1961
	0.00	0.00	1962			4.00	1,173.00	5.17	1962			0.00	0.00	1962
	0.00	0.00	1963			13.00	3,983.00	16.98	1963			0.00	0.00	1963
	0.00	0.00	1964					0.00	1964			0.00	0.00	1964
	0.00	0.00	1965				1.00	0.00	1965			0.00	0.00	1965
1,220.00	0.00	1,220.00			60.00	7,439.00	317,877.00	7,816.88		0.00	3,725.00	0.00	3,725.00	
1,220					7,817					3,725				
970.0	0.0	970.0			60.0	7,439.0	317,877.0	7,816.9		0.0	3,725.0	0.0	3,725.0	

Appendix B:

Table 3.3.2

DOE/RL-2000-43

MINOR TIER 1 SITE					MINOR TIER 1 SITE					MINOR TIER 1 SITE				
Savannah River (SR)					Idaho Operations					Idaho Operations				
DuPont					Phillips Petroleum Co. (MTI),					American Cyanamid Co.				
Sylvania Elec, Sylvania Coming Nuc Co.					AEC Health Physics Div									
Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	
					OUT-OF SCOPE					OUT-OF SCOPE				
					IRRADIATED FUEL					IRRADIATED FUEL				
	185.00	0.00	185.00	1952										
	11.00	0.00	11.00	1952										
	21.00	0.00	21.00	1953										
		0.00	0.00	1954										
1,207.00	0.00	0.00	1,207.00	1955										
104.00	0.00	0.00	104.00	1956										
297.00	0.00	0.00	297.00	1957										
943.00	0.00	0.00	943.00	1958										
318.00	0.00	0.00	318.00	1959										
59.00	0.00	0.00	59.00	1960										
4,248.00	0.00	0.00	4,248.00	1961										
46.00	0.00	0.00	46.00	1962										
34.00	20,226.00		54.23	1963										
	0.00	0.00	0.00	1964										
	0.00	0.00	0.00	1965										
0.00	7,473.00	20,226.00	7,493.23											
					</									

Appendix B:

[illegible]

MINOR TIER 1 SITE										MINOR TIER 1 SITE										MINOR TIER 1 SIT									
Materials Leasing Office Leases to Licensees										United Nuclear Corp Chem Division Oak Ridge										Kerr-McGee Oil Indust Oak Ridge									
Kg U subtotal		FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Kg EU	Kg U subtotal	FY	Kg DU	Kg NU	Kg EU	Kg U subtotal	FY	Kg DU	Kg NU	Kg DU		Kg DU		Kg DU		Kg DU		Kg DU	
0.00		1952			0.00	0.00	1952					1952			0.00	0.00	1952												
0.00		1952			0.00	0.00	1952					1952			0.00	0.00	1952												
0.00		1953			0.00	0.00	1953					1953			0.00	0.00	1953												
0.00		1954			0.00	0.00	1954					1954			0.00	0.00	1954												
0.00		1955			0.00	0.00	1955					1955			0.00	0.00	1955												
0.00		1956			0.00	0.00	1956					1956			0.00	0.00	1956												
0.00		1957			0.00	0.00	1957					1957			0.00	0.00	1957												
0.00		1958			4,586.00	7,373.59	1958					1958			0.00	0.00	1958												
10.14		1959			4,586.00	2,514.41	1959					1959			0.00	0.00	1959												
6.46		1960			0.00	0.00	1960					1960			0.00	0.00	1960												
3.40		1961			0.00	0.00	1961					1961			0.00	0.00	1961												
0.02		1962			0.00	0.00	1962					1962			0.00	0.00	1962												
0.00		1963			2,194,696.00	2,194.70	1963					1963			0.00	0.00	1963												
0.00		1964	6.00		0.00	6.00	1964					1964			0.00	0.00	1964		564.00										
0.00		1965			0.00	0.00	1965					1965			0.00	0.00	1965												
20.02			6.00	9,888.00	2,194,696.00	12,088.70		0.00	564.00	0.00	564.00		0.00	564.00	0.00	564.00													
0.020						12.089					0.564																		
20.0			6.0	9,888.0	2,194,696.0	12,088.7		0.0	564.0	0.0	564.0		0.0	564.0	0.0	564.0													

Appendix B:

Table 3.3.2

DOE/RL-2000-43

TE	MINOR TIER 1 SITE						MINOR TIER 1 SITE						MINOR TIER 1 SITE					
	Brookhaven Nat Lab New York Operations						Vitro Manufacturing Co. New York Operations						Hanford Operations US Bureau of Mines Oregon Metallurgical					
	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY
	0.00	0.00	1952		22.00	0.00	22.00	1952		284.00	0.00	284.00	1952		830.00			
	0.00	0.00	1952		313.00	0.00	313.00	1952		3,328.00	0.00	3,328.00	1952		1,637.00			
	0.00	0.00	1953		76.00	0.00	76.00	1953		1,629.00	0.00	1,629.00	1953		484.00			
	0.00	0.00	1954			0.00	0.00	1954		2.00	0.00	2.00	1954					
	0.00	0.00	1955			0.00	0.00	1955			0.00	0.00	1955					
	0.00	0.00	1956			0.00	0.00	1956			0.00	0.00	1956					
	0.00	0.00	1957		229.00	0.00	229.00	1957			0.00	0.00	1957					
	0.00	0.00	1958			0.00	0.00	1958			0.00	0.00	1958					
	0.00	0.00	1959			0.00	0.00	1959			0.00	0.00	1959					
	0.00	0.00	1960			0.00	0.00	1960			0.00	0.00	1960		8,326.00			
	0.00	0.00	1961			0.00	0.00	1961			0.00	0.00	1961		23,015.00			
	0.00	0.00	1962			0.00	0.00	1962			0.00	0.00	1962					
	0.00	0.00	1963	4.00		0.00	4.00	1963			0.00	0.00	1963					
	0.00	2,273.00	1964			0.00	0.00	1964			0.00	0.00	1964					
	0.00	0.00	1965			0.00	0.00	1965			0.00	0.00	1965					
	0.00	2,273.00		4.00	327.00	313.00	331.31		0.00	5,243.00	0.00	5,243.00		0.00	34,292.00			
	2.273						0.331						5.243					
	0.0	2,273.0		4.0	305.0	0.0	309.0		0.0	1,631.0	0.0	1,631.0		0.0	31,825.0			

Error

An error occurred while processing this page. See the system log for more details.

Appendix B: Table 3.3.3

FY 1966-70 Hanford Uranium Offsite Shipments

BASED ON HANFORD MBR REPORTS

MAJOR TIE																					
MINOR TIER 1 SITE											MINOR TIER 1 SITE										
New Brunswick Lab Oak Ridge Ops FBE											New York Ops Office Babcock & Wilcox										
FY	Date From	Date To	RIS From	Shipped From Hanford Contractor	Box #	Doc #	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	
1966	01-Jul-65	30-Jun-66	HZA	General Electric	38213	HAN 95170	1		776	1.78	1966				0.00	1966				0.00	
1966	01-Jul-65	30-Jun-66	HWA	Iscochem Inc.	38213	HAN 95136	1.00	0.00	2,148.00	3.15	1966	0.00	0.00	0.00	0.00	1966	0.00	0.00	0.00	0.00	
1966	01-Jul-65	30-Jun-66	HXA	Hanford Chem Processing Contractor subtotals			1.00	0.00	2,148.00	3.15	1966	0.00	0.00	0.00	0.00	1966	0.00	0.00	0.00	0.00	
				Douglas United Nuc	38214	HAN 95171	1.00	0.00	2,148.00	3.15	1966	0.00	0.00	0.00	0.00	1966	0.00	0.00	0.00	0.00	
				Hanford FY Aggregate subtotals			1.00	0.00	2,148.00	3.15	1966	0.00	0.00	0.00	0.00	1966	0.00	0.00	0.00	0.00	
1967	01-Jul-66	31-Dec-66	HZA	General Electric	38213	HAN 96413				0.00	1967				0.00	1967				0.00	
1967	01-Jan-67	30-Jun-67	HZA	General Electric	38213	HAN 96196				0.42	1967				0.00	1967				0.00	
1967	01-Jul-66	31-Dec-67	HWA	Iscochem Inc.	38213	HAN 96400	0.5		645	1.15	1967	0.00	0.00	0.00	0.00	1967	0.00	0.00	0.00	0.00	
1967	01-Jan-67	30-Jun-67	HWA	Iscochem Inc.	38213	HAN 96196	0.50	0.00	1,067.00	1.57	1967	0.00	0.00	0.00	0.00	1967	0.00	0.00	0.00	0.00	
1967	01-Jul-66	31-Dec-66	HXA	Hanford Chem Processing Contractor subtotals			0.50	0.00	1,067.00	1.57	1967	0.00	0.00	0.00	0.00	1967	0.00	0.00	0.00	0.00	
1967	01-Jul-66	31-Dec-66	HXA	Douglas United Nuc	38214	DUN 1916				0.00	1967				0.00	1967				0.00	
1967	01-Jan-67	30-Jun-67	HXA	Douglas United Nuc	38214	HAN 96194				0.00	1967	0.00	0.00	0.00	0.00	1967	0.00	0.00	0.00	0.00	
				Hanford FY Aggregate subtotals			0.50	0.00	1,067.00	1.57	1967	0.00	0.00	0.00	0.00	1967	0.00	0.00	0.00	0.00	
1968	01-Jul-67	31-Dec-67	HVA	Atlantic Richfield Han	46425	HAN 99439			495	0.50	1968				0.00	1968				0.00	
1968	01-Jan-68	30-Jun-68	HVA	Atlantic Richfield Han	46425	ARH 589			472	0.47	1968				0.00	1968				0.00	
1968	01-Jul-67	31-Dec-67	HXA	Hanford Chem Processing Contractor subtotals			0.00	0.00	967.00	0.97	1968	0.00	0.00	0.00	0.00	1968	0.00	0.00	0.00	0.00	
1968	01-Jan-68	30-Jun-68	HXA	Douglas United Nuc	38214	DUN 3524				0.00	1968				0.00	1968				0.00	
1968	01-Jul-67	31-Dec-67	HXA	Douglas United Nuc	38214	DUN 4436				0.00	1968	0.00	0.00	0.00	0.00	1968	0.00	0.00	0.00	0.00	
				Hanford FY Aggregate subtotals			0.00	0.00	967.00	0.97	1968	0.00	0.00	0.00	0.00	1968	0.00	0.00	0.00	0.00	
1969	01-Jul-68	31-Dec-68	HVA	Atlantic Richfield Han	46425	ARH 1036			36	0.04	1969				0.00	1969				0.00	
1969	01-Jan-69	30-Jun-69	HVA	Atlantic Richfield Han	46425	ARH 1099-6			104	0.10	1969				361	1969				361.00	
1969	01-Jul-68	31-Dec-68	HXA	Hanford Chem Processing Contractor subtotals			0.00	0.00	142.00	0.14	1969	0.00	0.00	0.00	0.00	1969	0.00	0.00	0.00	0.00	
1969	01-Jan-69	30-Jun-69	HXA	Douglas United Nuc	38214	DUN 5250				0.00	1969				0.00	1969				0.00	
1969	01-Jul-68	31-Dec-68	HXA	Douglas United Nuc	38214	DUN 5942				0.00	1969	0.00	0.00	0.00	0.00	1969	0.00	0.00	0.00	0.00	
				Hanford FY Aggregate subtotals			0.00	0.00	142.00	0.14	1969	0.00	0.00	0.00	0.00	1969	0.00	0.00	0.00	0.00	
1970	01-Jul-69	31-Dec-69	HVA	Atlantic Richfield Han	46425	ARH 1099-12			125	0.13	1970				0.00	1970				0.00	
1970	01-Jan-70	30-Jun-70	HVA	Atlantic Richfield Han	46425	ARH 1540-6			220	0.22	1970				0.00	1970				0.00	
1970	01-Jul-69	31-Dec-69	HXA	Hanford Chem Processing Contractor subtotals			0.00	0.00	345.00	0.35	1970	0.00	0.00	0.00	0.00	1970	0.00	0.00	0.00	0.00	
1970	01-Jan-70	30-Jun-70	HXA	Douglas United Nuc	38214	DUN 6557				0.00	1970				0.00	1970				0.00	
1970	01-Jul-69	31-Dec-69	HXA	Douglas United Nuc	38214	DUN 7049				0.00	1970	0.00	0.00	0.00	0.00	1970	0.00	0.00	0.00	0.00	
				Hanford FY Aggregate subtotals			0.00	0.00	345.00	0.35	1970	0.00	0.00	0.00	0.00	1970	0.00	0.00	0.00	0.00	
KG TOTAL: ALL U TYPES																361.00					
MTU TOTAL: ALL U TYPES:																0.36					
IN-SCOPE RECYCLE TOTAL: ALL U TYPES MTU																0.36					

ER 1 SITE																MAJOR TIER 1 SITE																MINOR TIER 1 SITE																MAJOR TIER 1 SITE															
Ohio (NLO), Fernald, FEMP																Carbide & Carbon Chem Co. ORO Y-12 & K-25 GDP (CYT, FZB), (FZA, FZE, FZF)																Oak Ridge Natl Lab Oak Ridge Operations FZC																Union Carbide Co.-Fernald Fernald Oak Ridge Operations FYA															
Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU	Kg U subtotal	FY	Kg DU	Kg NU	Gm EU																																					
122166	895631472	1,017,797.47	1966				0.00	1966				0.00	1966				0.00	1966				0.00	1966																																								
122,166.00	1126064945	1,126,064.85	1966				0.00	1966				0.00	1966				0.00	1966				0.00	1966																																								
82,398.00	2,023,696,317.00	2,145,862.32		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00																																					
284,964.00	14,038,208.00	96,436.21	1966		84		84.00	1966				84.00	1966				84.00	1966				84.00	1966																																								
	2,637,734,523.00	2,342,296.52		0.00	84.00	0.00	84.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00																																					
2,454	56,738,050	59,192.05	1967				0.00	1967				0.00	1967				0.00	1967				0.00	1967																																								
1,413	117,584,456	118,987.46																																																													
	550,001,735	550,001.74																																																													
	735,171,140	735,171.14	1967				0.00	1967				0.00	1967				0.00	1967				0.00	1967			14,432,875.00	0.00																																				
3,867.00	1,469,495,387	1,469,362.36		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00																																					
32174	10,790,511	42,064.51					0.00					0.00					0.00					0.00																																									
40443	14,413,920	54,356.92	1967				0.00	1967				0.00	1967				0.00	1967				0.00	1967																																								
76,484.00	1,404,698,812.00	1,561,163.87		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00																																					
	552,206.88	552,206.88	1968				0.00	1968				0.00	1968				0.00	1968				0.00	1968																																								
0.00	1001717324	1,001,717.32	1968				0.00	1968				0.00	1968				0.00	1968				0.00	1968																																								
	1,563,924,205.00	1,563,924.21		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00																																					
58158	88359837	146,517.84					0.00					0.00					0.00					0.00																																									
20421	173,619,791	200,040.78	1968				0.00	1968				0.00	1968				0.00	1968				0.00	1968																																								
84,978.00	1,815,983,833.00	1,908,482.83		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00																																					
	835,030.06	835,030.06	1969				0.00	1969				0.00	1969				0.00	1969				0.00	1969																																								
	1034973941	1,034,973.84	1969				0.00	1969				0.00	1969				0.00	1969				0.00	1969			3,537,144.00	0.00																																				
0.00	1,870,012,897.00	1,870,012.90		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00																																					
46447	112,223,945	158,870.95					0.00					0.00					0.00					0.00																																									
27208	83,035,338	110,243.34	1969				0.00	1969				0.00	1969				0.00	1969				0.00	1969																																								
73,855.00	2,045,272,788.00	2,138,827.18		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00																																					
	1149680201	1,149,680.20	1970				0.00	1970				0.00	1970				0.00	1970				0.00	1970																																								
13	618946796	1,087,516.80	1970				0.00	1970				0.00	1970				0.00	1970				0.00	1970																																								
13.00	1,789,006,997.00	2,234,877.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00																																					
36861	130,535,155	175,894.16					0.00					0.00					0.00					0.00																																									
14541	49,398,234	63,910.23	1970				0.00	1970				0.00	1970				0.00	1970				0.00	1970																																								
81,216.00	1,943,911,398.00	2,472,897.29		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00	0.00		0.00	0.00	0.00																																					
				10,319,573.7				84.00				0.00				0.00				0.00				0.00				0.00																																			
				10,319.57				0.08				0.08				0.00				0.00				0.00				0.00																																			
				10,319.57				0.08				0.08				0.00				0.00				0.00				0.00																																			

DOE/RL-2000-43

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Appendix B:

Table 3.3.4

DOE/RL-2000-43

Hanford Chemical Processing Contractor (HVA) Uranium Shipments FY 1971-March 31, 1999

Based on NMMSS Document RL2000-I-00002 SHIPMENTS (classified)

ATLANTIC RICHFIELD HANFORD

FY	DU				EU			
	MAJOR TIER 1:		MAJOR TIER 1:		MAJOR TIER 1:		MAJOR TIER 1:	
	To PADUCAH Kg DU CKY, FYA	To FERNALD Kg DU FVA, FVB, FVC	To OAK RIDGE Kg DU FZA, FZE, FZB, FZF	Total Kg DU To External From Hanford	To PADUCAH Gm EU CKY, FYA	To FERNALD Gm EU FVA, FVB, FVC		
71	624,855	0	0	1,206,462	96,721,586	130,928		
72	1,292,075	0	0	1,292,201	1,786,421,765	0		
73	208,144	0	0	208,144	349,974,776	0		
74	0	0	0	1,017,222	0	0		
75	0	0	0	4	0	0		
76	0	0	0	0	0	0		
76T	0	0	0	0	0	0		
77	0	0	0	0	0	0		
78	0	0	0	0	0	0		
79	0	0	0	0	0	0		
80	0	0	0	0	0	0		
81	0	0	0	0	0	0		
82	0	0	0	0	0	0		
83	0	0	0	0	0	0		
84	0	0	0	0	0	0		
85	0	0	0	0	0	0		
86	0	0	0	0	0	0		
87	0	0	0	0	0	0		
88	0	0	0	0	0	0		
89	0	0	0	0	0	0		
90	0	0	0	0	0	0		
91	0	0	0	0	0	0		
92	0	0	0	0	0	0		
93	0	0	0	0	0	0		
94	0	0	0	0	0	0		
95	0	0	0	0	0	0		
96	0	0	0	0	0	0		
97	0	0	0	0	0	0		
98	0	0	0	0	0	0		
99	0	0	0	0	0	0		
KG TOTAL	2,125,074	0	0	3,724,033	2,233,118	131		
MTU TOTAL	2,125.07	0.00	0.00	3,724.03	2,233.12	0.13		

AFTER FY 1977 - HVA CONTRACTOR TURNOVER TO
ROCKWELL HANFORD (HRA)

Appendix B:

Table 3.3.5

DOE/RL-2000-43

Hanford Chemical Processing Contractor (HRA) Uranium Shipments FY 1971-March 31, 1999

Based on NMMSS Document RL2000-1-00002 SHIPMENTS (classified)

ROCKWELL HANFORD OPERATIONS (RHO)

DU						EU					
FY	MAJOR TIER 1:		MAJOR TIER 1:		MAJOR TIER 1:		MAJOR TIER 1:		MAJOR TIER 1:		
	To PADUCAH	To FERNALD	To OAK RIDGE	Total Kg DU	To PADUCAH	To FERNALD	To OAK RIDGE	Total Kg DU	To PADUCAH	To FERNALD	
	Kg DU	Kg DU	Kg DU	To External	Gm EU	Gm EU	Gm EU	From Hanford	Gm EU	Gm EU	
	CKY, FYA	FVA, FVB,FVC	FZA,FZE,FZF,FZF		CKY, FYA	FVA, FVB,FVC			CKY, FYA	FVA, FVB,FVC	
71	PRIOR TO FY 1977 - HVA CONTRACTOR										
72											
73											
74											
75											
76											
77											
78											
79											
80											
81											
82											
83											
84											
85											
86											
87											
88											
89											
90											
91											
92											
93											
94											
95											
96											
97											
98											
99											
00											
KG TOTAL	0	0	0	1,134	0	0	0	1,134	0	3,088,285	
MTU TOTAL	0.00	0.00	0.00	1.13	0.00	0.00	0.00	1.13	0.00	3,088.29	

AFTER FY 1988 - HRA CONTRACTOR TURNOVER TO
WESTINGHOUSE HANFORD (HUD)

[illegible]

Appendix B: Table 3.3.6

Hanford Chem. Processing Contrs (HUD & HTA) Uranium Shipments FY 1971-March 31, 1999

Based on NMMSS Document RL2000-1-00002 SHIPMENTS (classified)

WESTINGHOUSE HANFORD (HUD) & FLUOR (HTA)

FY	DU				EU			
	MAJOR TIER 1: To PADUCAH Kg DU		MAJOR TIER 1: To FERNALD Kg DU		MAJOR TIER 1: To OAK RIDGE Kg DU		MAJOR TIER 1: To PADUCAH Gm EU	
	CKY, FYA	FVA, FVB, FVC	FZA, FZE, FZF	Total Kg DU To External From Hanford	CKY, FYA	FVA, FVB, FVC	FZA, FZE, FZF	MAJOR TIER 1: To FERNALD Gm EU
71								
72								
73								
74								
75								
76								
77								
78								
79								
80								
81								
82								
83								
84								
85								
86								
87	0	0	0	0	0	0	0	0
88	0	0	0	4,792	0	0	0	123,425.579
89	0	0	0	0	0	0	0	212,831
90	0	0	0	0	0	0	0	0
91	0	0	0	0	0	0	0	0
92	0	0	0	1	0	0	0	0
93	0	0	2,937	2,943	0	0	0	0
94	0	0	0	138	0	0	0	0
95	0	0	0	0	0	0	0	0
96	0	0	0	102,463	0	0	0	0
HUD UNTIL 97	0	0	0	0	0	0	0	0
HTA IN 97	0	0	0	29,855	0	0	0	0
98	0	0	0	0	0	0	0	0
99	0	0	0	2	0	0	0	0
KG TOTAL	0	0	2,937	140,194	0	0	0	123,638
MTU TOTAL	0.00	0.00	2.94	140.19	0.00	0.00	0.00	123.64

PRIOR TO FY 1987 - HRA CONTRACTOR

HUD & HTA

[illegible]

Appendix B: Table 3.3.7

Hanford Reactor Contractor (HXA) Uranium Shipments FY 1971-March 31, 1999

Based on NMMSS Document RL2000-I-00002 Shipments (classified)

UNITED NUCLEAR INC. (UNI)

DU										EU									
FY	MAJOR TIER 1:				MAJOR TIER 1:				Total Kg DU To External From Hanford	MAJOR TIER 1:				MAJOR TIER 1:					
	To PADUCAH		To FERNALD		To OAK RIDGE		To FERNALD			To PADUCAH		To FERNALD							
	Kg DU CKY, FYA	FVA, FVB, FVC	Kg DU FVA, FVB, FVC	Kg DU FZA, FZE, FZB, FZF	Kg DU FZA, FZE, FZB, FZF	Kg DU FZA, FZE, FZB, FZF	Kg DU FZA, FZE, FZB, FZF	Gm EU CKY, FYA		Gm EU FVA, FVB, FVC	Gm EU CKY, FYA	Gm EU FVA, FVB, FVC							
71	0	195	0	0	0	0	0	2,807	0	0	0	268,837,731							
72	0	0	0	0	0	0	0	2	0	0	0	80,729,637							
73	0	0	0	0	0	0	0	0	0	0	0	366,733,111							
74	0	0	0	0	0	0	0	0	0	0	0	122,947,745							
75	0	167	0	0	0	0	0	167	0	0	0	55,173,776							
76	0	0	0	0	0	0	0	0	0	0	0	16,293,460							
76T	0	0	0	0	0	0	0	0	0	0	0	1,531,320							
77	0	0	0	0	0	0	0	0	0	0	0	121,614,441							
78	0	0	0	0	0	0	0	0	0	0	0	92,030,675							
79	0	0	0	0	0	0	0	0	0	0	0	56,744,559							
80	0	0	0	0	0	0	0	17	0	0	0	56,647,925							
81	0	0	0	0	0	0	0	0	0	0	0	91,358,407							
82	0	0	0	0	0	0	0	0	0	0	0	77,508,844							
83	0	0	0	0	0	0	0	17	0	0	0	159,046,418							
84	0	0	0	0	0	0	0	45	0	0	0	236,609,566							
85	0	0	0	0	0	0	0	36	0	0	0	170,342,951							
86	0	0	0	0	0	0	0	1	0	0	0	158,122,879							
87	0	5,075	0	0	0	0	5,075	5,075	0	0	0	54,070,469							
AFTER FY 1987 - HXA CONTRACTOR TURNOVER TO WESTINGHOUSE																			
KG TOTAL	0	0	5,437	0	0	0	5,437	8,167	0	0	0	2,186,344							
MTU TOTAL	0.00	0.00	5.44	0.00	0.00	0.00	5.44	8.17	0.00	0.00	0.00	2,186.34							

HXA

NU									
MAJOR TIER 1:		MAJOR TIER 1:		MAJOR TIER 1:		MAJOR TIER 1:		MAJOR TIER 1:	
To OAK RIDGE	Total Gm EU	To PADUCAH	To FERNALD	To OAK RIDGE	Total Kg NU	To OAK RIDGE	Total Kg NU	To External	Total
Gm EU	To External	Kg NU	Kg NU	Kg NU	To External	FZA,FZE,FZB,FZF	FZA,FZE,FZB,FZF	From Hanford	
FZA,FZE,FZB,FZF	From Hanford	CKY, FYA	FVA, FVB, FVC	FZA,FZE,FZB,FZF	From Hanford				
4,473	271,052,547	0	294,782	0	294,787				
0	80,816,881	0	32	0	35				
0	366,733,111	0	2	0	2				
0	124,128,581	0	1,079,051	0	1,079,051				
0	55,294,056	0	273	0	273				
0	16,376,285	0	9,570	0	9,576				
0	1,544,483	0	0	0	0				
0	121,673,126	0	25,132	0	25,132				
0	184,032,236	0	258	0	258				
0	56,801,590	0	4,831	0	4,831				
0	56,683,550	0	16	0	16				
0	106,767,550	0	0	0	0				
0	77,882,087	0	11	0	11				
0	159,578,973	0	0	0	0				
0	237,020,621	0	328	0	328				
0	170,518,638	0	4,760	0	4,774				
0	164,284,960	0	10,574	0	10,717				
0	54,302,961	0	1,716	0	1,716				
4	2,305,594	0	1,431,336	0	1,431,507				3,745,268
0.00	2,305.59	0.00	1,431.34	0.00	1,431.51				3,745.27

Appendix B:

Table 3.3.8

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Pacific Northwest National Laboratory (PNNL) Offsite Uranium Shipments

Based on NNMSS Document SROO-00009 SHIPMENTS (classified)

FY	TO PADUCAH		TO FERNALD		TO OAK RIDGE		DU		TO PADUCAH		TO FERNALD		TO OAK RIDGE		EU	
	MT 20 Kg DU	CKY, FYA	MT 20 Kg DU	FVA/FVB, FVC	MT 20 Kg DU	FZA, FZE, FZB, FZC	Total Kg DU PNNL External (Excludes Hanford)		MT 20 Gm EU	CKY, FYA	MT 20 Gm EU	FVA, FVB, FVC	MT 20 Gm EU	FZA, FZE, FZB, FZC	Total Gm EU PNNL External (Excludes Hanford)	
FY65 - FY 1970	0	0	2	0	0	0	3,029		0	0	5,350,361	0	1,108	0	5,612,875	
71	0	0	7,608	0	1	0	7,761		0	0	3,347,630	0	515	0	3,491,460	
72	0	0	4,490	0	0	0	5,349		0	0	5,689,782	0	172,707	0	7,989,439	
73	0	0	0	0	0	0	477		0	0	1,700,676	0	760	0	1,705,331	
74	0	0	0	0	66	0	1,467		0	0	4,040,965	0	1,850,671	0	5,998,866	
75	0	0	0	0	4	0	2,260		0	0	6,852	0	0	0	1,127,815	
76	0	0	0	0	0	0	2,246		0	0	1,383,773	0	0	0	1,613,160	
76 1/2	0	0	0	0	0	0	598		0	0	0	0	0	0	193	
77	0	0	360	0	357	0	4,510		0	0	15,365	0	0	0	95,263	
78	0	0	608	0	207	0	5,257		0	0	26,723	0	242	0	155,382	
79	0	0	100	0	274	0	5,008		0	0	1,486	0	18,169	0	2,329,082	
80	0	0	686	0	11	0	8,582		0	0	1,155,465	0	0	0	1,345,290	
81	0	0	0	0	16	0	9,814		0	0	-851	0	0	0	103,413	
82	0	0	210	0	0	0	16,872		0	0	7,784	0	0	0	115,112	
83	0	0	114	0	1	0	4,409		0	0	973,544	0	100	0	1,004,561	
84	0	0	11	0	4	0	1,761		0	0	0	0	0	0	57,779	
85	0	0	0	0	30	0	1,907		0	0	0	0	0	0	39,407	
86	0	0	0	0	0	0	3,389		0	0	0	0	45,472	0	63,501	
87	0	0	0	0	0	0	1,335		0	0	0	0	0	0	17,206	
88	0	0	0	0	16	0	16,645		0	0	382,173	0	0	0	385,442	
89	0	0	0	0	26	0	15,291		0	0	0	0	0	0	28,839	
90	0	0	0	0	0	0	10,544		0	0	0	0	0	0	49,472	
91	0	0	0	0	0	0	16,422		0	0	0	0	0	0	22,540	
92	0	0	0	0	0	0	12,531		0	0	0	0	0	0	5,169	
93	0	0	0	0	5,570	0	6,184		0	0	0	0	0	0	6,267	
94	0	0	0	0	0	0	12,630		0	0	0	0	0	0	0	
95	0	0	0	0	0	0	2,570		0	0	0	0	0	0	145,118	
96	0	0	0	0	0	0	0		0	0	0	0	0	0	1,743,425	
97	0	0	0	0	0	0	0		0	0	0	0	0	0	95	
98	0	0	0	0	0	0	0		0	0	0	0	0	0	0	
98 1/2	0	0	0	0	0	0	0		0	0	0	0	0	0	6,718	
KG TOTAL	0	0	14,169	0	6,583	0	179,819		0	0	24,082	0	2,090	0	34,359	
MTU TOTAL	0	0	14,169	0	6,583	0	179,819		0	0	24,081,828	0	2,089,744	0	34,358,879	

Table 3.3.8

Appendix B:

TO PADUCAH MT 81 Kg NU CKY,FYA	TO FERNALD MT 81 Kg NU FVA,FVB,FVC	TO OAK RIDGE MT 81 Kg NU FZA,FZE,FZB,FZF	NU MT 81 External (Excludes Hanford)	ALL U Total Kg U External Shipments
0	4358	0	10108	18,751
0	1,467	0	1,875	12,927
0	3,918	0	6,518	18,956
0	-40	0	-7	2,175
0	10,288	0	10,296	17,762
0	3	0	20	3,438
0	160	0	936	4,785
0	0	0	10	569
0	0	0	5	4,710
0	0	0	29	5,441
0	0	0	1	8,138
0	0	0	1	9,928
0	0	0	2	9,919
0	0	0	93	17,080
0	0	0	1	5,415
0	0	0	0	1,819
0	0	0	0	1,846
0	0	0	1	3,454
0	0	0	0	1,552
0	0	0	0	17,030
0	0	0	4	15,325
0	0	0	2	10,595
0	0	0	7	16,452
0	0	0	0	12,536
0	0	0	43	6,213
0	0	0	0	12,630
0	0	0	0	2,715
0	0	0	3	1,746
0	0	0	0	0
0	0	0	0	0
0	0	0	0	7
0	20,154	0	29,749	243,927
0	20,154	0	29,749	243.93

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Appendix C

Section 4 - Supplemental Information

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APPENDIX D
Section 5 Supplemental Information

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Appendix D
Section 5 Supplemental Information

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Appendix E

Description of Research Strategies Used in Study

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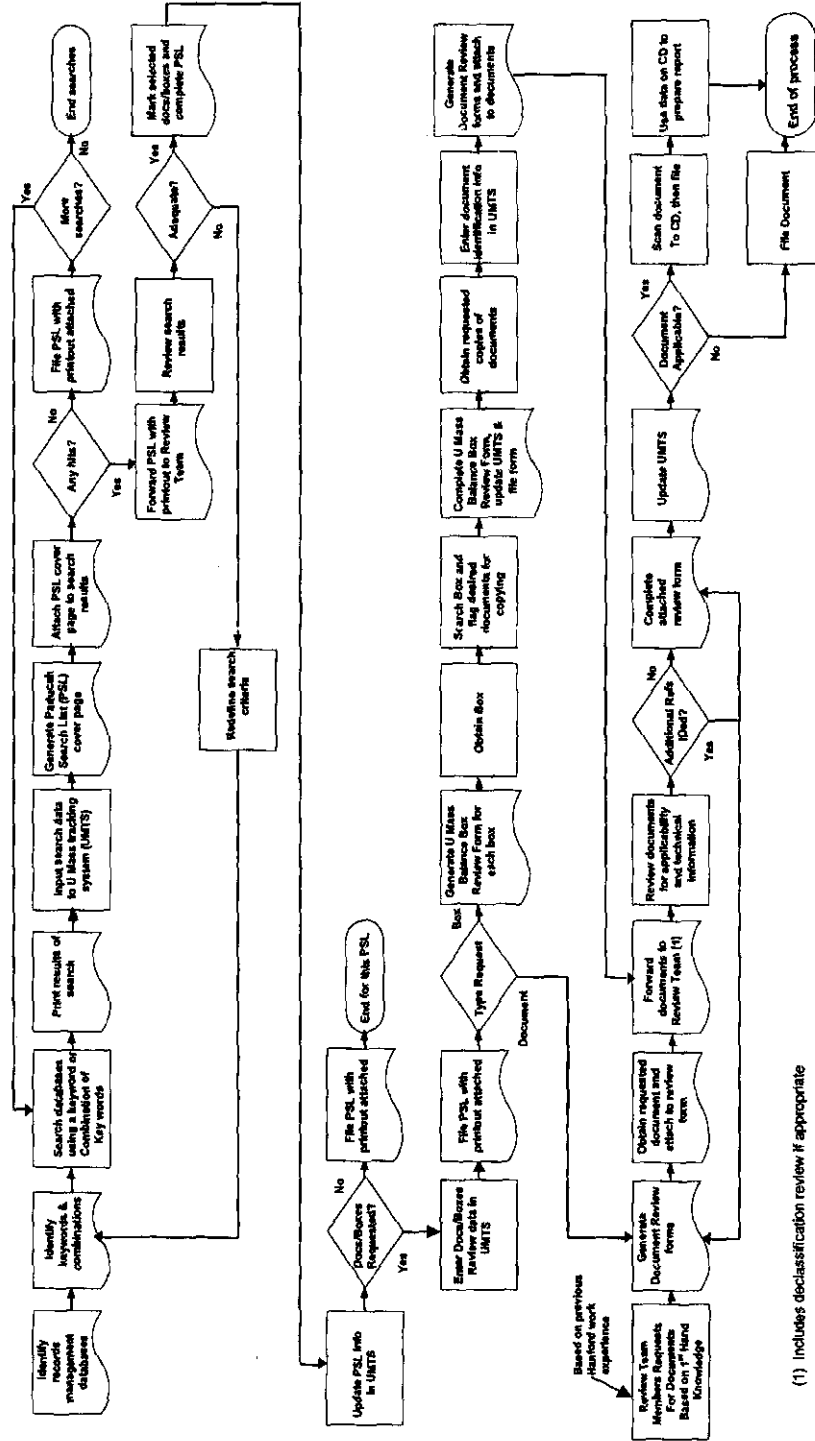


Figure E-2 Uranium Mass Balance Project Document Identification/Review Process

APPENDIX E

Description of Research Strategies Used in Study

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E.1 Databases Used

The Technical Review Team members were selected based on their previous experience at Hanford, in most cases including experience directly related to one or more of the facilities involved in the uranium recovery program. Once the team was assembled, the members, based on their past experience, identified the types of records that would be needed to respond to the four specific points in the Deputy Secretary of Energy's letter of direction dated September 15, 1999 [Glauthier, 1999]. Several existing databases were identified that catalogued various types of documentation that might be applicable to this project were searched. These were:

- *Human Radiation Experiments Information Management System (HREX)* – This publicly accessible database was created in 1994 and contains approximately 392,000 pages of historical documents related to radiation experiments on human subjects performed by or for the Departments of Defense, Energy, Health and Human Services, and Veterans Affairs, as well as the Nuclear Regulatory Commission, the National Aeronautical and Space Administration, the Central Intelligence Agency, and their predecessor organizations.
- *OpenNet* – This database includes references to Department of Energy (DOE) documents that were declassified and made publicly available after October 1, 1994, in compliance with former Secretary Hazel O'Leary's Openness Initiative. It presently contains approximately 400,000 documents; however, the declassification process is still in progress.
- *Records Holding Task Group* – This collection of approximately 102,000 classified documents is held in the records holding area of the DOE's Oak Ridge Operations Office. In the summer of 1998, the Hanford "Downwinders" Litigation staff reviewed the entire collection and indexed all Hanford-related documents, which resulted in a listing of approximately 14,000 Hanford related documents. This Hanford-related listing was searched as part of the U-Mass study.
- *Hanford Site Historian Database* – This database contains documents gathered by the Site Historian in generating several publicly available publications related to various aspects of the Hanford site history.
- *Document Declassification Tracking System (DDTS)* – This is a site database created in 1995 with approximately 61,000 classified and formerly classified Hanford documents.
- *Record Holding Area Management Information System (RHA-MIS)* – This database was developed in the mid-1970s as the Hanford Site master database for managing RL and its contractors' archived boxes of records in storage. The system currently tracks the location and general content of approximately 75,000 boxes of records

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from the field to various storage locations, including the Federal Records Center in Seattle, WA.

- *Document Tracking Application (DTA)* – Created in the mid-1970s, this system contains approximately 41,000 classified documents.
- *Master* – This system was created in the mid-1970s and contains approximately 750,000 engineering documents related to the Fast Flux Test Facility. Other contractor documents were added to the system between 1982 and 1993, when this system was replaced with the Records Management Information System (RMIS).
- *Records Management Information System* – This system was created in 1993 and consists of 14 different subsystems with over 1,700,000 documents. The system was not retrofitted to capture documents issued prior to its startup; however, some older documents have been added to the system from time-to-time in special cases. Only the following subsystems were of interest to the Project:
 - *Records* – This subsystem contains over 640,000 documents, including ALARA records, correspondence, declassified documents, engineering drawings, legacy/historical information, media highlights, management system documents, radiological survey records, technical documents, publications, and work packages.
 - *Tank Farms Information Center (TFIC)* – This subsystem contains primarily older Westinghouse Hanford Company and Rockwell Hanford Company documents related to the tank farms.
 - *Solid Waste* – This subsystem contains hazardous waste manifests.
 - *RL* – This subsystem contains RL correspondence.
 - *Administrative Record (AR)* – This subsystem contains documents relied on or considered in order to arrive at a final decision for remedial action for hazardous waste management.

Although, three additional RMIS subsystems were identified and searched, later analyses verified these files could have been eliminated because the information they contained was not germane to the Project.

E.2 Database Keyword Searches

The Technical Review Team identified keywords or combinations of keywords that related to the facilities, materials, and types of documents that would be used in the database searches in an attempt to identify documents relevant to the Team's

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evaluation. Table E-1 shows the primary keywords that were used, normally in combination, in the searches. The team also identified variations in the way in which the keywords might appear in the titles or documents identified in the databases. Combining the keywords into meaningful logical expressions resulted in identifying several hundred queries/combinations of queries to be run on each of the various databases.

Each specific query was assigned a unique identification number (Paducah Search List or PSL number) and was logged into a Project database along with:

- Keywords and logic used in the search
- Name of the database searched
- Date of the search
- Name of the researcher
- Number of documents matching the search criteria (or "hits")
- Date the search results were provided to the Technical Review Team

The above information was reflected on a database generated cover sheet that was then attached to the search results printout. The printout identified the documents or boxes of records that satisfied the search criteria. The cover sheet and printout were then delivered to the Technical Review Team for review.

If the number of hits on a particular query exceeded 200, only the first two pages of the results were printed and provided for review. The problem of a high number of hits was generally caused by one of two things:

- 1) The search criterion was too general.
- 2) In some databases documents have been scanned and digitized and full text searches using optical character recognition (OCR) software is available; however both the lack of quality of the original documents and the numerous different fonts used in them caused the OCR software to register many false hits.

In those cases, a Technical Review Team member would evaluate the first two pages of hits and determine whether to ask for the rest of the results or to narrow the search criteria and have the search performed again.

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titles of the documents listed, determined whether copies of any of the documents should be obtained for Team review. Where appropriate, document titles were flagged to indicate that they were being requested for review. In the case of the RHA-MIS database, the computer printouts provided box numbers, storage locations, and a general description of the types of documents in each box. Again, one or more Review Team members reviewed the list of boxes and flagged those that might contain records related to the Project. The reviewer(s) completed the PSL cover sheet to include the reviewer's name, date of review, whether any documents or boxes were selected and, if so, the number of documents or boxes selected, along with any comments the reviewer wished to make.

E.4 Obtaining Documents and Boxes of Records

After review by the Technical Review Team the PSL cover sheet and its associated list were used to update the Project database with the information provided from the reviewers. For each document requested by the Technical Review Team, the following information was entered into the Project database:

- A unique document tracking number (for the request)
- The unique PSL number that resulted in the request
- Document identification number (if any)
- Document revision
- Document alias number(s) (if any)
- Document date
- Document Title
- Document source
- Box number
- File folder title (if any)
- Name of requestor

The requested document was then either printed out the document (if it was available online) or a copy was ordered from the appropriate source. When the document became available, the database generated a Document Review/Production Order containing the above information as well as the date the document was delivered to the requestor and the number of pages in the document. In those cases where the requests were for entire boxes of records, arrangements were made to retrieve the boxes from offsite storage if necessary and made available for review in the RL Records Holding Area (RL-RHA). Once the boxes were available in the RL-RHA, the Technical Review Team members who had requested them were notified via Box Review Tracking forms which identified the box number, the box owner, security classification of the box content, and whether the box had been completely scanned. The form also provided the box tracking history.

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E.5 Additional Document Searches

Because of their previous work experience at Hanford, many of the Technical Review Team members were aware of the existence and location of specific documents that would be relevant to the Team's efforts. In such cases the Team member would obtain a copy of a document. A Document Review/Production Order form was initiated for each document obtained in this manner and it was entered into and tracked in the Project database.

E.6 Technical Review of Documents

One or more members of the Technical Review Team reviewed each document requested to determine whether it contained information relevant to the Team's efforts. If a document contained no relevant information, the reviewer marked the Document Review/Production Order form to indicate that the document was not relevant and then signed and dated the form and returned both to to be used to update the Project database. If a document did contain relevant information, the reviewer also indicated the security classification status, and whether declassification in whole or part was needed. The reviewer also marked the form to identify keywords to be associated with the document in the Project database to assist in future searches by the Project Team or others.

In the case of boxes of records, a Review Team member (normally, the requestor, but not always) examined the contents of a box and flagged any relevant documents for copying. The RL-RHA staff made copies upon request and a Document Review/Production Order was initiated for each document copied. The information was then entered into the previously described Project database.

E.7 Project Records

The Project records for this project include:

- Documentation (completed Paducah Search List or PSL) for each query run on each database and the marked up computer printouts indicating the documents requested.
- A completed Document Review/Production Order for each of the documents requested showing the results of the Teams review.
- A completed Box Review Tracking form for each box requested showing the results of the box review.

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- A computer database that cross-links the queries to the PSLs, the PSLs to the document requests, and the keywords assigned to a document by the Technical Review Team.

In preparing its report, the Technical Review Team cited references to the sources of information used in the report. Copies of the source documents will be retained by RL until it is determined they are no longer needed. It is the Project's intent to scan these documents onto CD-ROMs and catalog them in a database to minimize future search and duplicating time and expense.

E.8 References

Glauthier 1999

Memorandum, T. J. Glauthier, Deputy Secretary of Energy, to All Departmental Elements, *Paducah Gaseous Diffusion Plant – Follow-Up Activities*, U.S. Department of Energy, Washington, DC, September 15, 1999

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F.1 Single Pass Reactor Fuel Fabrication

F.1.1 Fuel Canning

The original fuel canning process implemented at Hanford involved the use of an electric heater press, known by workers as the "whiz-bang," to heat and bond the uranium fuel cores to their aluminum jackets. However, the heaters burned out frequently, did not heat the elements and cans to consistent temperatures, and did not produce a uniform bonding. This problem was serious because non-uniform bonding caused thin places in the jacketing which were subject to localized heating during irradiation. These "hot spots" could cause fuel element ruptures in the reactors. By August 1944, the uranium fuel cores were being jacketed in a triple-dip method that consisted of bathing them in molten bronze, tin, and then a molten aluminum-silicon mixture. The bronze used in this process at Hanford was relatively high in tin content (53% tin and 47% copper), and the bronze bath itself had a flux cover composed of barium chloride, potassium chloride, and sodium chloride. As fuel cores were dipped into this mixture, they acquired trace coverings of all of these substances.

Initially, the bare uranium cores were cleaned by passing them through a trichloroethylene vapor degreaser. They were then passed through a nitric acid pickling tank, two rinse tanks, and a hot air dryer. Meanwhile, a steel "sleeve" that would surround each can during the dipping process was cleaned in sodium hydroxide; and aluminum end caps and cans were cleaned in a sodium dichromate solution followed by a methanol rinse. The bare uranium cores were dipped in a bronze bath to heat them to a uniform temperature within the uranium beta phase (660 °C to 770 °C). Then they were placed in a tin bath to: (1) cool them into the uranium alpha phase (less than 660 °C) and (2) to remove any excess bronze. Next they were centrifuged to throw off excess tin. Then the cores were immersed quickly in an aluminum-silicon brazing bath while in the uranium alpha phase, and water quenched. The various heating and cooling procedures were done to randomize the uranium grains, thus inhibiting the uranium "growth" (expansion under irradiation) problem. After water quenching, the steel sleeve was pulled away and cleaned with sodium hydroxide and soap to remove any remaining aluminum-silicon. The sleeve then could be reused many times. The thickness of the residual end cap on the element was then measured with a fluoroscope and marked with a punch to indicate the amount that needed to be removed in subsequent end machining. Identification numbers were stamped on the can base end, and the braze line on the end cap was tungsten inert gas welded to seal the porous braze to the end cap and can. A final etching in nitric acid completed the procedures.

Three tests followed the canning process. The first, was the frost test, which consisted of spraying the can with acenaphthene mixed with carbon tetrachloride (CCl_4). The canned element was then placed into an induction coil to heat its surface. If there was a gas bubble or a non-bonded spot, this spot would become shiny, and the element then would be rejected and sent back through a recycling process. If the bond was good, the acenaphthene was removed with trichloroethylene, and the element was heated in one

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of several autoclaves located in the 314 Building. In that step, the canned element was placed into a steam autoclave, which operated at about 100 pounds per square inch gauge (psig) and 175 °C for more than 20 hours, to reveal any pinholes or incomplete welds. Water from the steam would be conducted through any such openings, and the uranium core would expand rapidly, resulting from the formation of a uranium oxide compound known as U_3O_8 , and split the aluminum can. If an element passed the autoclave test it then underwent a final radiograph (X-ray) test in the 314 Building, to detect porosity in the end weld bead. Any porosity could have become a pathway for water to contact the uranium fuel and cause the element to rupture during irradiation.

F.1.2 Single Pass Fuel Fabrication Changes

In 1948, the extrusion press in the 314 Building was excessed, and Hanford began receiving rolled uranium rods from an offsite commercial mill. The rolling process seemed to offer metallurgical advantages, because the uranium could be processed at lower temperatures, which induced less oxidization and produced smaller and more random grains within the metal. From 1950 to 1951, a rolling mill was procured and installed in the 314 Building, to save the costs of shipment to offsite mills. However, this mill was relatively small, and the rolling operation was transferred to a large facility constructed at the Feed Materials Production Center (FMPC), an Atomic Energy Commission (AEC) site in Fernald, Ohio, in 1952. Thereafter, no extruding or rolling operations were conducted at Hanford in connection with the fabrication of fuel elements for single-pass reactors. The 314 Building process continued to operate for the purposes of straightening uranium rods, providing autoclave and radiograph testing of canned elements, and providing uranium scrap processing operations.

In 1954, the 313 Building underwent a major remodeling and expansion, reaching a total size of 182.5 feet by 486 feet, with a total area of 76,633 square feet. At that time, much contaminated equipment and other solid wastes from this building and its immediate surrounding area and from the 303 fresh fuel warehouses were buried. The remodeling occurred at the time that fuel canning technology in the 313 Building switched from the triple-dip process to the new lead-dip process. Lead-dip consisted of immersing the uranium fuel cores in a duplex bath (molten lead covered with molten aluminum-silicon) to preheat the cores in the uranium alpha phase. This step formed an intermetallic compound of uranium and lead (UPb or UPb_3) on the core. It was followed by a molten aluminum-silicon bath (also in the uranium alpha phase) to braze and bond the cores to the aluminum cans and caps. This process allowed the first canning bath to occur at a lower temperature (lower than 660 °C) because the uranium cores already had been beta heat treated in a molten salt bath at the FMPC. However, the new method introduced a great deal more lead and other heavy metals into 313 Building waste streams. At about the same time that the lead-dip process replaced the triple-dip method, an ultrasonic test replaced the frost test, which eliminated the use of acenaphthene and CCl_4 . Concurrently, the majority of testing autoclaves were removed from the 314 Building and placed in the north end of the 313 Building.

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In the early 1960's, just before the eight single-pass reactors at Hanford began to close, experiments were under way in the 304, 3716, and 313 Buildings with a new canning procedure known as the Hot Die Size Process. Also termed the "nickel-plate" procedure, this operation plated uranium fuel cores with nickel, using nickel sulfate, nickel chloride, and boric acid. It included standard fuel fabrication cleaning, degreasing, etching, and testing chemicals and processes. Although the Hot Die Size method was tested successfully, it was not implemented on a large scale because of the impending closures of Hanford's eight original reactors.

F.2 N-Reactor Fuel Fabrication Processing

Fuel element preparation activities for the single-pass reactors ended in the 313 and 314 Buildings in 1971, when the last of these reactors closed. The 314 Building was modified in the 1970's and was used for a variety of research projects and crafts services. The majority of the fabrication equipment for single-pass reactor fuel elements was removed from the 313 Building between the mid-1970's and the mid-1980's. However, the south end of the 313 Building continued to house major functions in support of N-Reactor fuel production. Among these functions were: (1) the receiving and inspection of uranium billets and other components used to make N Reactor fuel elements, (2) the chemical passivation of spacers from N-Reactor, (3) the casting and machining of copper-silicon pre-shape components used in N-Reactor fuel elements (beginning in 1973), and, (4) the neutralization and handling of non-uranium-bearing acid wastes from N-Reactor fuel fabrication processes in the 333 Building. Finished N-Reactor fuels and fabrication components, tools, and miscellaneous supplies were stored in the north end of the 313 Building from 1971 to 1987, and an Engineering Development Laboratory, including facilities for working with uranium, was established in this structure in the 1970's.

F.2.1 N-Reactor Fuel Fabrication

The fuel-making process for the New Production Reactor (N-Reactor) was very different from that used to make fuel for single-pass reactors. Soon after funding was secured for N-Reactor in 1958, a high-pressure heat transfer apparatus was emplaced in the 189/190-D Building, a converted World War II pumphouse in the Site's 100-D Area. Its purpose was to test a new, N-Reactor fuel concept being developed in the 306 Metallurgical Pilot Plant, a 300 Area building dedicated to fuel manufacturing experimentation. The concept first tried for N-Reactor fuel was a wire-wrapped, seven-element cluster of long, thin fuel rods spaced together in a horizontal flow tube. Each individual element was only 0.625 to 0.704 inches in diameter, and was 35 to 45 inches long. However, attention soon turned to yet another new concept developed in the 306 Building. This idea, of a co-extruded tube-in-tube fuel element design, eventually was adopted for N-Reactor.

Supplemental Information on Hanford Fuel Fabrication**F.2.2 Co-Extrusion Process**

The co-extrusion process began with inspection and cleaning of copper and copper-silicon pre-shapes and backing plates used in the process. The cleansing agents were nitric acid, nitric hydrofluoric, and chromic nitric sulfuric acid. Next, cladding components made from Zircaloy-2TM were degreased, rinsed in nitric and hydrofluoric acid, and dried with forced-air heating. In the meantime, uranium billets were degreased with perchloroethylene, etched with nitric acid, rinsed, dried and inspected. Next, the copper, copper-silicon, Zircaloy-2TM, and uranium components were assembled and welded into a billet assembly. This assembly was evacuated of air, leak tested, sealed preheated, and then co-extruded (squeezed together) in the Loewry Press. As the process specifications for this step emphasized: "The quality of the extruded tube is dependent upon many things, not the least of which is skill, care, effort, and precision that are put into the co-extrusion operation."

The process of cleaning, degreasing, etching and drying components, then assembling and pressing them, was repeated for both the outer (larger) and inner (smaller) tubes that made up the tube-in-tube configuration. The extruded tubes then exited the press to a roll-out table where they were rolled continuously for at least six minutes to prevent tube deformation and non-uniform cooling. Next they were sectioned to the specified length, and the ends were machined to create fuel sections or elements. Nitric acid was used to remove copper silicon residues, and nitric sulfuric acid was used to chemically mill (i.e., dissolve away) excess uranium on fuel element ends. Elements then were etched with nitric hydrofluoric and nitric acid, and brazed with an etched braze ring material consisting of Zircaloy-2TM alloyed with about five percent beryllium. (This braze material previously had been degreased and etched.) The brazed elements were heat-treated in a molten salt bath to randomize the uranium grain structure to prevent preferential grain growth that could rupture the elements in the reactor.

The next step in the process was to weld projections or supports onto the fuel elements. Eight lengthwise protrusions were attached to the outer surface of each fuel element, evenly spaced around its diameter. This configuration allowed cooling water to circulate optimally around the elements, without creating hot spots where the sides of elements rested too close to the inner walls of the process tubes. After projections were welded onto the elements, the two tubes (inner and outer) had to be attached together. Support hardware was attached to the outer surface of the inner tube, and locking hardware was affixed to the inner surface of the outer tube. The two tubes were then given a final nitric hydrofluoric acid etch, separately tested in autoclaves, inspected, assembled and interlocked, and stored as finished fuel. The co-extrusion process was carried out continuously in the 333 Building from 1960 until December 1986, reaching a peak volume of approximately 250 finished fuel elements per week in the mid-1980s.

Worker exposure to uranium was controlled. Machining and other fines-producing fabrication activities were conducted within ventilated enclosures. Air exhausted from these enclosures was filtered. Uranium scrap recovery activities such as burning in a controlled atmosphere incinerator, while providing a higher potential for worker uranium

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uptake, were limited in scope. Additionally, due to transportation issues with untreated scrap, much of the scrap was processed to a recoverable wet sodium diuranate cake before shipment offsite. Some low concentration liquid wastes were discharged to ponds and cribs for disposal. Dry wastes were packaged and sent to the Hanford waste burial sites for disposal.

F.3 Material Sources

A number of private contractors were involved with the early fuel fabrication operations. B&T Metals of Columbus, Ohio, extruded a large quantity of uranium metal rods for Hanford from April through August 1943. Copperweld Steel Company of Warren, Ohio out-gassed and straightened a large quantity of uranium rods for the reactors between May and August 1943. Revere Copper & Brass also out-gassed and straightened rods in Detroit. Hanford began out-gassing and straightening its own uranium fuel rods in September, 1944. Baker Brothers of Toledo, Ohio, manufactured unbonded uranium slugs for Hanford from early 1944 through July, 1944. The William E. Pratt Manufacturing Company turned and ground unbonded slugs in the spring of 1944. Between May and August of 1944, McKinney Tool and Manufacturing in Cleveland, Ohio turned and ground unbonded slugs. During the late 1940s and early 1950s, uranium rods were rolled or extruded by Vulcan Crucible Steel Company in Aliquippa, PA, Revere Copper and Brass, and the Brush Beryllium Company in Detroit, Joslyn Manufacturing & Supply Company in Fort Wayne, Indiana, Allegheny-Ludlum Steel Corporation in Waukegan, New York, and Simonds Saw & Steel Co. of Lockport, NY.

Hanford stopped extruding uranium rods in 1948, switching to rolled rods on site. The AEC shifted the rolling work to the Fernald, Ohio Feed Materials Production Center and its supporting contractors in 1952.

Uranium slug machining was taken over by FMPC at Fernald Ohio, which opened in 1952, and the Weldon Spring plant which opened in 1956. FMPC and Weldon Spring produced ingots of natural, low enriched and depleted uranium to be extruded offsite into tubes and billets for further machining into uranium cores. The cores were then shipped to Hanford for cladding and assembly. The ingots were extruded into tubular billets by Bridgeport Brass Company in Adrian, MI from 1954 to 1961 and later by its corporate successor, Reactive Metals, Inc., in Ashtabula, Ohio. Fernald then shipped the billets to Hanford.

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F.4 References (This entire Appendix is supported by one or more of the following general references.)

DOE/EM-0319 1997

DOE/EM-0319, *Linking Legacies*, January 1997

Gerber 1992

WHC-MR-0388, M. S. Gerber, *Past Practices Technical Characterization Study*, 300 Area, December 1992

Gerber 1996

WHC-MR-0521, Rev 0, M. S. Gerber, *The Plutonium Production Story at the Hanford Site, Process and Facilities History*, June 1996

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G.1 Operating History Uranium Trioxide (UO₃) Plant [Gerber 1993]

The original Uranium Trioxide (UO₃) Plant was located in a World War II facility known as the 224-U Bulk Reduction Building. This three-story, frame and concrete block structure had just under 12,000 square feet, and had been constructed to house a part of the plutonium separation and concentration process used at Hanford in the 1940s. However, process improvements as early as 1945 demonstrated that the 224-U Building was not needed. Rather than contaminate it with plutonium, Hanford management decided to utilize the building as a training facility until another use was found. The 224-U Building was modified in 1951 to accomplish conversion of Uranyl Nitrate Hexahydrate (UNH) to UO₃. This facility subsequently became known as the UO₃ Plant.

The facility began test runs in January 1952, and full operations the following month. The first UO₃ calciners simply were large pots that heated the UNH in batches. An early difficulty concerned the UNH feed from REDOX, which contained impurities sufficient that much of Hanford's UO₃ product "proved to be inadequate for successful processing in the continuous UF₆ conversion process at Oak Ridge." Process improvements in the REDOX Plant were made during 1952-1953, to correct this situation. The first feed supplies of UNH received from the U-Plant in 1953 also proved to be problematic. Metallic impurities, as well as the nitric acid content and the presence of Tri-butyl Phosphate (TBP) and its decomposition products required that the UO₃ produced from initial U-Plant product UNH had to be sent to an intermediate metallurgy facility in Cleveland, Ohio further purification before shipment to Oak Ridge. Additionally, the U-Plant UNH foamed, caked and stuck in the pots during the calcining process, so that the UO₃ would have to be vacuumed and even chopped out. This condition was undesirable from the standpoints of efficiency, operator safety, and contamination control. However, changes made in the U-Plant rendered the UNH so pure that the UO₃ Plant product, by the end of 1953, did not have to be sent offsite for further purification, and was suitable to be shipped and used directly at Oak Ridge. That year, the UO₃ Plant processed more than 1.5 times the total fuel inventory of uranium that was received at Hanford.

During 1954, UO₃ Plant capacity increased to 18 tons per day with the addition of two gas-fired calciners. The unit cost per ton also dropped to 79 percent that of 1953. Overall production in 1954 was limited, however, by the quantity of feed available from the REDOX and U plants. REDOX experienced many shutdowns caused by equipment failures that year, and the U-Plant was closed for much of the autumn to allow for system modifications that would allow it to process wastes aged only one year (instead of usual three years). Several problems were encountered and overcome during 1954, including high radiation fields around the calciner pots caused by the processing of newer UNH (higher in zirconium, niobium, and ruthenium) from the two Hanford feed plants. This problem was solved by the addition of special shielding to protect operators during pot unloading. Also, sulfamic acid added to the UNH prior to calcination caused some caking in the calciner pots, and renewed foaming occurred due to the presence of organic decomposition products in the UNH. Silicon anti-foaming agents, as well as

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other sulfur-based additives were used to control these situations. A key improvement that year was the use of vacuum cleaning techniques that enabled the plant to recover 90-95 percent of the oxide powder associated with failed filter bags. A large backlog of such bags that had accumulated since startup, were cleaned and discarded. By year's end, plans were underway to expand the UO₃ Plant.

During 1955, monthly UO₃ production averaged 106 percent that of 1954, while the unit costs per metric ton dropped to 92 percent of that for the previous year. The plant had to shut down for portions of the summer due to contamination problems resulting from high gamma feed from the U-Plant. More persistent problems resulted from the gradual corrosion and mechanical wear of equipment and calciner seals and gaskets to the point where a vacuum could not be maintained in the pots. Airborne contamination around the pots increased until operators had to wear masks for respiratory protection. In 1956, equipment modifications, including new pot agitators, shafts, seals, bearings and supports, as well as new pot cover assemblies, off-gas systems and gasketing, were installed to address these problems.

In the meantime, plans went forward to construct the 224-UA Building, a major addition to the UO₃ Plant. The plant was equipped with six large, new, continuous action calciners. These calciners were developed and designed at Hanford, and had large troughs with paddle agitators that essentially turned and mixed the UNH/UO₃ bed throughout the process. The caking and clogging problems that had plagued the pots was obviated by the below-grade valving of the continuous calciners. The new valves used air as an agitator, and maintained a seal between the calciner and the powder pickup bin, while passing a continuous stream of UO₃. The new calciners also produced a pebbly product that consisted of spheres with an average diameter of 200 microns (about 1/100th of an inch), as opposed to the granular oxide product of the pot.

The first three of the continuous calciners began operations in the last quarter of 1956, and, despite mechanical difficulties with their powder handling systems during the earliest months, they soon demonstrated their production potential. That December, UO₃ Plant throughput exceeded that of any previous month by 27 percent. Overall, the 1956 plant production exceeded that of 1955 by 57 percent.

Another salient improvement made during 1956 was the replacement of the old, 900-pound drums used for offsite shipping of the UO₃ powder with new T-Hoppers. These containers left the site on specially fitted rail flat cars and served as feed hoppers for the Oak Ridge plant customer.

Installation of the last three of the continuous calciners was completed at the UO₃ Plant in early 1957. At that time, the 18 pot units were retired from service, and all processing was done through the continuous calciners. Late that year, design scoping was begun for new facilities needed to segregate regular UNH feed from that generated by the processing of E-Metal in the REDOX Plant. The UO₃ powder that resulted from processing the two types of UNH streams had significantly different nuclear reactivity levels, and separation was needed by the customer at Oak Ridge.

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During 1958, two new tanks with associated piping and instrumentation were installed and fitted to receive the enriched UNH from the stainless steel, overhead pipeline that brought the UNH from the REDOX Plant. Routine transfers of enriched UO₃ from Hanford to Oak Ridge began in early 1959. Meanwhile, in 1958, the UO₃ Plant established another high production record that was 5.5 percent higher than that of 1957, and almost double that of 1954. Metallurgical examination of the two-year old continuous calciners showed their condition to be excellent.

In 1958, the U-Plant finished its mission and was shut down. However, PUREX's UNH feed to the UO₃ Plant far surpassed that which had been coming from U-Plant. Together, the UO₃ and PUREX Plants went on to set and surpass production records almost continually, while the REDOX Plant continued to supply a small stream of enriched UNH until its shutdown in 1967. During 1959, the concentration equipment and the acid recovery system at the UO₃ Plant were automated, and in 1960, for the first time, the calciners were automated so that they could be operated and shut down remotely. Programming of the calciners included preliminary air blowing and steam heating of the feed points, admission and regulation of the feed, and control of the UO₃ bed temperature. Steep production climbs at the plant continued through 1961, but dropped off slightly in 1962 due to mechanical and process difficulties at the feed plants (PUREX and REDOX). By 1963, production again was so high that serious consideration was given to constructing a pipeline, or converting an existing one, to carry UNH from PUREX to the UO₃ Plant. However, the project did not go forward, and transfers by tanker truck continued. That same year, production of UO₃ shipped offsite rose to about 12,000 tons per year, and the capability was added to process UNH derived from "125 Metal." During 1965-1966, an experimental process of conversion of thorium nitrate into thorium oxide powder was carried out in the UO₃ 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. However, for reasons unrelated to the UO₃ Plant, the use of thorium oxide powder was abandoned, in favor of experiments with thorium wafer targets.

Beginning in 1967, UO₃ Plant operations were tied exclusively to those of the PUREX Plant. That year, UNH concentrator modifications in the UO₃ Plant improved heat distribution and allowed the calciner feed to maintain a uniformly higher specific gravity, thus producing more powder in the same operating time. Both the UO₃ and PUREX plants closed in 1972. However, PUREX resumed operations in late 1983 -- closely followed by UO₃ Plant in early 1984. Since that time, there have been 17 startups at the UO₃ Plant, averaging about eight days each, as the plant could calcine UNH at a much faster rate than the PUREX Plant could produce it. Final deactivation orders came for both plants in 1992. In April 1993, the UO₃ Plant resumed operations to convert the final 200,000 gallons of remaining UNH to UO₃ powder. That run was the plant's last.

In general, waste management was handled very simply and efficiently at the UO₃ Plant. Radiation levels in the UNH were relatively low, as compared with levels in the chemical processing feed plants, as there was no high-level waste generated or processed by the UO₃ process. Process condensate was sent to various U-Plant cribs

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over the years, specifically the 216-U-12 crib in the 1980s and the 216-U-17 crib for later operations. Various ditches and swamps received equipment cooling water and the steam condensate, previously the U-Pond and more recently the 214-U Ditch. Airborne UO₃ powder was trapped in primary bag filters, with secondary bag filters and then HEPA (High Efficiency Particulate Air) filters added during the 1972 - 1984 shutdown. That period also witnessed the addition of fire protection improvements, a backup power supply for instrumentation, an open faced hood and associated exhaust and filtration equipment for the UO₃ loadout, and a nitrous oxide (NO_x) monitor for the 296-U-4 stack. Beginning in the late 1950s, the main off-gas from the UO₃ process (NO_x), was routed through "wet scrubbers," which sprayed the gas with nitric acid to capture the NO_x vapors and dissolve any entrained solid oxide particulates. The acid mixture was then routed through an acid absorber (bubble-cap tower) and stored in chemical tanks awaiting transfer to PUREX. In the late 1980s, a Fiber Mist Eliminator was added to scrub acid vapors from the UNH concentrator to minimize entrained solution and particulates.

G.2 Process Operations

The major unit operations performed at the UO₃ Plant were concentration of UNH, calcination of UNH to UO₃, packaging of the UO₃ product, and nitric acid recovery. Uranium containing wastes were generated during routine operation. The primary waste streams were the liquid effluents discharged to the ground and gaseous effluents released to the atmosphere.

The UNH solution was transferred to the UO₃ Plant and stored in Tank C-1 in C-Cell. During operation the UNH solution was accumulated in storage tanks and then transferred to the concentrators in D-Cell. The UNH was concentrated to approximately 100 weight per cent UNH in thermosiphon evaporators. The optimum operating temperature was 125°C to 127°C. The concentrated UNH was stored in a steam heated tank. Off-gas from the concentrator knockout pot was routed through a catch tank to a fiber mist eliminator. The mist eliminator captured UNH droplets entrained from the concentrators and the recycle unit concentrator. The recovered UNH was eventually recycled to PUREX. Part of the condensate, was used as reflux water in the acid absorber tower, and the balance of the condensate was neutralized and pumped to the U-17 crib [Raab 1978; Harmon 1979; Strickland 1993] .

Concentrated UNH was pumped to the 224-U building where the calcination process was conducted and UNH converted to UO₃ powder. Initially calcination was performed using pot calciners. The UO₃ product from the pot calciners was unloaded using a pneumatic unloading system, which conveys uranium oxide from the pots and conveys it to bulk storage. The system consisted of an exhaustor, a bag filter, a cyclone collector, a rotary valve, a hammer mill, and various pipe and fittings. The pot calciners were replaced by 1956 with continuous calciners. The UO₃ particle bed in the calciner was maintained at 270°C. The shell temperature was maintained at about 500°C for optimal efficiency.

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The UO₃ powder was collected from the calciners in ground-level pickup bins and then transferred by the pneumatic transfer system, using air, to the fourth floor of the UA tower to the cyclone separator. From there it flowed by gravity through a hammer mill (not normally used) on the third floor of the tower, then to a storage hopper on the second floor, and was unloaded from the hopper into T-Hopper shipping containers in the powder loadout room. The exhaust air from this operation was routed through primary and secondary bag filters in series, a prefilter, and HEPA filter before discharge to the environment, via a roof exhauster. Powder from the cyclone and primary bag filter was collected in the powder hopper. Powder from the secondary bag filter was also collected.

As the UNH was thermally decomposed, oxides, of nitrogen and water were driven off and drawn through the vent piping to the Acid Recovery Tower where they were converted to nitric acid. The system consisted of calciner off-gas scrubbers that remove fine particles of entrained UO₃, a vapor cooler, an absorber tower and a system of reflux water addition. The acid was collected and pumped to the 211-U Tank Farm for storage and return to the processing plants. Net acid recovery was about 92 percent of that represented by the incoming UNH solution. The nitric acid had a low residual level (0.043 molar) of UNH.

G.3 Waste Generation [Harmon 1979]

The processing of 1,000 MTU at the UO₃ Plant typically resulted in the generation of ~15 million gallons of cooling water, plus steam condensate from the acid recovery and uranium nitrate concentration steps, and chemical sewer waste. Total radionuclide content of this water which totaled 120 million gallons in 1972 was less than 2 Ci of combined fission products and alpha-emitting nuclides. This stream was sent to 216-U-10 Pond.

Process condensate, largely from the UNH concentrators in the 224-U Building, averaged about 300,000 gallons annually, containing about 0.01 Ci of total mixed fission products. It was sent to the 216-U-12 Crib.

Approximately 1.2×10^9 ft³ of gases were discharged to the atmosphere from the UO₃ Plant annually, containing an average (total) of 6×10^{-3} Ci of fission products and 9×10^{-5} of alpha emitting radionuclides. The radionuclides were essentially all ¹⁰⁶Ru and uranium, respectively.

Solid uranium waste was generated from failed and replaced equipment and normal line-generated process waste. About 120 to 140 ft³ (total) was generated during a six week campaign mode of operation. This represented approximately 0.01 Ci of total beta emitters; 0.01 Ci of combined ⁹⁰Sr, ¹³⁷Cs and ¹⁰⁶Ru nuclides; and less than 0.2 kg of total uranium. The solid wastes were buried in the 200 Area waste burial grounds.

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G.4 Radiation Exposure

The early years of UO₃ Plant operations were plagued with dusting and radiation problems around the calcination pots and during bag filter changes. Gustavson mentioned [Gustavson 1950] that "the dust problem in the pot room was one of the greatest obstacles in designing the UO₃ Building." Gerber cited [Gerber 1993] that Annual Reports reported occasions when "airborne contamination around the pots increased until operators had to wear assault masks." Two separate ventilation systems were installed to address the dust problems [Gustavson 1950]. Manual unloading of the calciner pots and high levels of maintenance were also reported. Radiation fields around the pot calciners resulted in the addition of special shielding in 1954 to protect the operators during pot unloading. The installation of continuous calciners greatly reduced exposure and dusting problems associated with the calcination process. Exposure during bag filter changes was also a concern in the early years of operation due to dusting problems. As a result, protective clothing and respiratory protection were used at these locations.

After many modifications and improvements to process operations, the total complement of workers when the UO₃ Plant was in full operation was approximately 30, including operations, maintenance and supervision during the latter years of UO₃ Plant operations. In 1971 to 1972, the average annual whole body occupational radiation dose of the UO₃ workers was approximately 1 rem; the maximum was 2.3 rem and the minimum was 0.09 rem. However, because this plant operated only part time, the process operations workers were assigned elsewhere for more than half of the operating year [Harmon 1979].

G.5 Contaminate Partitioning

The major unit operations performed at the UO₃ Plant were concentration of UNH, calcination of UNH to UO₃, packaging of the UO₃ product, and nitric acid recovery. None of these unit operations would preferentially concentrate or decontaminate the plutonium or neptunium from the uranium. The chemical and physical properties of the actinides are similar under the concentration and calcination operating conditions used at the UO₃ Plant such that there was no separation of one transuranic radionuclide from another. The ratio of transuranic constituents to uranium in the UO₃ product, residual uranium in waste streams, and uranium in any recovery streams, would have been the same as that in the UNH feed to the UO₃ Plant.

There is some evidence, however, of fission product partitioning during calcination. There is a statement in the UO₃ Fission Product Specifications [Knights 1966] that "⁹⁵ZrNb and ⁹⁹Tc are carried almost quantitatively through the concentration and denitration processes, whereas ruthenium concentration is reduced by a factor of 2 to 5. An early report indicated an average decontamination factor (DF) of 6 across the UO₃ Plant from U Plant UNH. This was attributed to volatilization of ruthenium during the calcination operation. The acid specific fission product analyses indicated that 95

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percent was ruthenium. [Abrams 1955]. In contrast, the DF from REDOX UNH was less than one, indicating that the ruthenium concentration was significantly lower.

Some technetium heptoxide (Tc₂O₇ – pertechnatate), which has a boiling point of 310°C, could be expected to be volatilized during the uranium calcination step, which operated at < 300°C. The off-gas from the calciners was driven off and drawn through vent piping to the Acid Recovery Tower. This system consisted of calciner off-gas scrubbers that remove fine particles of entrained UO₃, a vapor cooler, an absorber tower and a system of reflux water addition. Any ⁹⁹Tc that would have been volatilized would have been recovered along with the NO₂, entrained uranium and fission products and been recycled as nitric acid to the reprocessing plants. There is no evidence that any ⁹⁹Tc reached the final exhaust filters. The presence of other beta-gamma fission products would have masked any radiation from the soft beta of the ⁹⁹Tc.

G.6 UO₃ Related Incidents

One incident involving UO₃ powder occurred April 12, 1960 when the flexible hose used to load powder into a large shipping container broke and spilled about 1.5 tons of powder in the loadout room, of which 2 to 5 Kg escaped to the loadout pad. This powder spread over the asphalt loading ramp and to the road around the UO₃ Plant. Most of the powder was swept up and put in drums for recovery, but the rest of the powder was washed off the asphalt and onto the ground surface. Contaminated soil was removed and buried. The contamination was limited to the immediate areas of the spill. There was no general spread in the 200 West Area, no detectable contamination offsite, and no personnel contamination problems of concern were encountered [HW-64898 1960].

There were several incidents involving UNH and liquid effluents. On December 30, 1954 2,000 gallons of UNH were spilled on the main roadway just east of the 200-East Area hill. No external exposure in excess of permissible limits is known to have resulted from this incident. All individuals involved in this incident were surveyed and found to be free of contamination, and an analysis of the bioassay result indicated no detectable deposition of soluble uranium [HW-34494-DEL 1955].

The most serious radiation event identified during this study involving personnel exposure at the UO₃ Plant occurred in December 1960, when the fresh air mask hose being used by an employee was blocked by condensate in the line and the mask was improperly adjusted. The employee inhaled/ingested 7-12 mg of uranium [DOE/AD-0015 1991]. Up to that time, this was the highest recorded deposition at the plant. Within 24 hours, 80% of the uptake had been eliminated, reducing the body burden to 10% of the permissible level. Subsequent whole body counter examinations a week after exposure showed no detectable uranium.

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Appendix H

Timeframe of Events at Hanford

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APPENDIX I

Estimates of Annual Recycled Uranium Constituents Shipped Offsite

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Estimates of Annual Recycled Uranium Constituents Shipped Offsite

I.1 This appendix contains engineering estimates of annual quantities of constituents (Pu, Np, and Tc) contained in shipments of recycled uranium shipped to/from the Hanford site. Tables I-1 and I-2 summarize the annual receipts and shipments of recycled uranium to and from the Hanford site during the last fifty years. These tables contain summaries of annual receipt and shipment based upon data presented in Section 3 and Appendix B of this report. Analytical data on the amount of Pu, Np, and Tc in recycled uranium received at Hanford is not available since there were no requirements for these measurements. Limited analytical data has been identified which indicates that the recycled uranium shipped offsite from Hanford typically contained Pu in the range of one to five ppb, Np in the range of 20 to 500 ppb, and Tc in the range of 3 to 12 ppb. Data supporting these constituent concentration ranges is given in Section 4 and Appendix C of this report. Data on the concentration of Np and Tc in Hanford recycled uranium is minimal, since there were specification requirements for these elements,

Since it is not possible to provide a complete historical assessment of the constituent levels in all recycled uranium shipped each year, rough estimates of annual quantities of constituents in recycled uranium were made assuming that the constituent levels during the years where analytical data is not available are the same as the constituent levels for years when analytical data is available. These estimates are based upon the constituent levels mentioned above and were done for the bounding minimum and maximum values, and the mean value of the constituent concentration range. Tables I-3, I-4, and I-5 illustrate the estimated quantities of Pu contained in the recycled uranium shipped. Tables I-6, I-7, and I-8 illustrate the estimated quantities of neptunium in the recycled uranium shipped. Tables I-9, I-10, and I-11 illustrate the estimated quantities of technetium contained in the recycled uranium shipped. Tables I-12, I-13, and I-14 present estimates of the Pu, Tc, and Np constituents in recycled uranium received at Hanford, shipped from Hanford, contained in waste at Hanford, released to the environment at Hanford, or in the current inventory at Hanford.

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