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,

recovering both plutonium and uranium. At about this same time, Hanford initiated recovery of uranium from B-Plant and T-Plant tank waste in U-Plant, and operation of the UO_3 Plant was started for conversion of the recovered uranyl nitrate hexahydrate (UNH) to UO_3 powder for shipment offsite. Subsequently, in 1956, the Plutonium-Uranium Extraction Plant (PUREX) facility initiated operation to also process irradiated fuel and recover both Pu and uranium. PUREX became the sole irradiated fuel processing plant at Hanford after 1967 and ceased operation in early 1990. PUREX production eventually accounted for approximately 69 percent of all uranium recovered for recycle at Hanford. All recovered uranium at Hanford, which was in the form of UNH, was calcined to a shippable powder in the UO_3 Plant. The UO_3 powder was sampled and packaged into either drums or specially designed "T-Hoppers" for shipment.

Data Examination

This study involved searching, reviewing, and analyzing documents which have been prepared throughout the history of the site. These searches and document reviews involved personnel on the RL Team with varied technical expertise and knowledge of past Hanford operations and previous studies. Due to the sheer volume of the records to potentially be searched and the allocated time and available resources, automated keyword-based data searches were conducted in parallel with the searches by the field experts. The computerized screening of records and documents produced since 1943 was then screened for data to supplement the records located by the field experts.

Significant Information

The following list is a summary of significant findings of this study:

Hanford Uranium Summary	
Total uranium received at Hanford	~119,271 MTU
Total uranium shipped from Hanford	~112,287 MTU
Total uranium in inventory	~4,006 MTU
Uranium sent to waste (all forms)	~2,174 MTU
Uranium lost to fission & transmutation	~140 MTU*
Total difference (~0.56% by weight)	**~664 MTU**

* Estimated uranium consumption in reactors

- Hanford received, fabricated as fuel, irradiated, and processed **~119,271** metric tons of uranium (MTU) to produce **~67** MT of Pu for defense and non-defense purposes.

- Hanford has shipped **~112,287** MTU (which includes **~109,792** MT of recycled uranium as UO_3 product oxide and fuel fabrication scrap and **2,495** MT of unirradiated uranium in various forms). Approximately **74,491** MT of the recycled

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

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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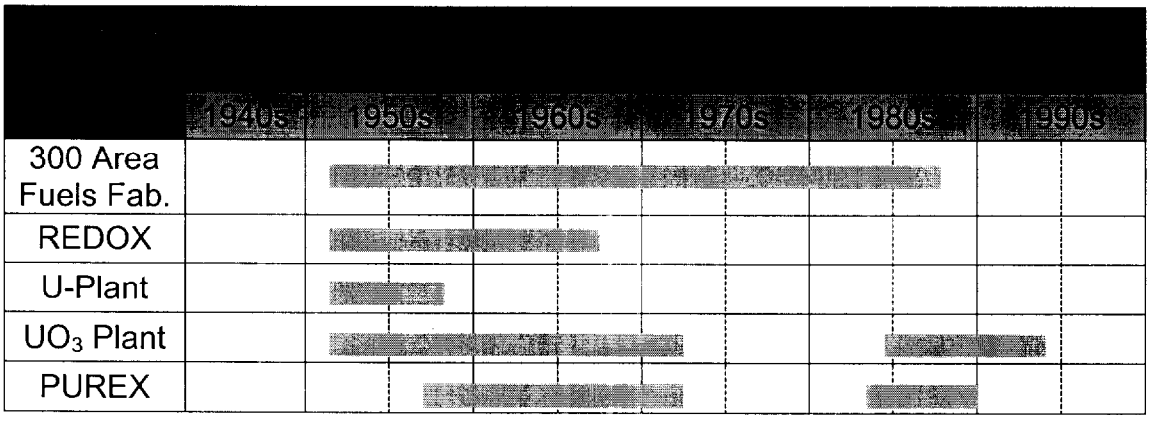
- Limited comparisons which could be done on the level of constituents in UO_3 product containing recycled uranium from Hanford, with analytical results reported in available historical documents from the receiving sites, indicates that the analytical results have been reasonably consistent between sites.

- During this study, documentation reviewed has not indicated that any process within those facilities which handled recycled uranium concentrated the impurities in the recycled uranium. 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 in the recycled uranium could have volatilized to the off-gas system and then could have been either plated out on equipment surfaces, accumulated in off-gas scrubber solutions, or released to the environment. Documentation has been found to indicate that some of the ruthenium volatilized during UNH calcination, with a decrease in concentration ranging from a factor of <1 to 6.

- The primary facilities which handled recycle uranium at Hanford were the UO_3 Plant and the fuel fabrication facilities. The UO_3 Plant operated intermittently from 1952 to 1994, and reactor fuel fabrication operations occurred during 1942 to 1987. Some of the facilities used for fuel fabrication operations continue to be



used for the storage of unirradiated uranium fuel and fuel fabrication material. The separation facilities (REDOX, and PUREX), which processed irradiated fuel, and the U-Plant, which recovered uranium from high-level waste, only involved handling recycle uranium during concentration and loadout of the recovered UNH. This UNH was subsequently transferred to the UO_3 Plant for conversion to an oxide powder for shipment. The U-Plant operated from 1952 to 1958, REDOX Plant operated 1952 to 1967, and the PUREX Plant operated 1956 to 1990.

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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.