4.0 Contaminants in Recycled Uranium

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

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 X 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 1.10E-2 ppm on a ^{235}U basis [Judson 1962]. In

Table 4-1 UO_3 Process Specifications

Document Number	Title	Date	Pu Spec. Ppb	Laboratory Analysis Ref
HW-35938	UO_3 Product Specifications	1955	10	
HW-30654, Rev	Product Specifications Uranium Trioxide (Depleted) for Off-Site Shipment	1959	10	HW-24403 660.22
HW-59136	Product Specifications Uranium Trioxide (Depleted E Metal) for Off-Site Shipment	1959	10	HW-24403 660.22
HW-79219, Rev	Product Specifications Uranium Trioxide (Depleted E Metal) for Off-Site Shipment	1964	<10	HW-65402, Rev
Sloat 1964	Product Specifications Uranium Trioxide (Depleted) for On-Site Storage	1964	<10	HW-65402, Rev
HW-79219, Rev 2	Product Specifications Uranium Trioxide (Depleted E Metal) for Off-Site Shipment	1965	<10	HW-65402, Rev
ISO 528	Product Specifications Uranium Trioxide (Enriched to < 1% U^{235}) for Off-Site Shipment	1967	<10	HW-65402, Rev
ARH 896	Product Specifications Uranium Trioxide Depleted Normal (72) Metal	1969	<10	HW-65402, Rev
ARH 1396	Product Specifications Uranium Trioxide Depleted Normal (72) For Off-Site Shipment	1969	<10	ARH-85
ARH 1396, Rev	Product Specifications Uranium Trioxide Depleted Normal (72) For Off-Site Shipment	1970	<10	ARH-85
ARH 1493	Specification for PUREX UNH Product	1970	10	ARH-85
ARH 1763	Product Specifications Uranium Trioxide (Enriched to < 1% U^{235}) for Off-Site Shipment	1970	<10	ARH-85
AEC-2202	Product Specifications Hanford Uranium Trioxide	1971	<10	ARH-85
OSD-U-185-0001	Uranium Oxide Plant Operating Specifications	1983	<10	
OSD-U-185-0001	Uranium Oxide Plant Operating Specifications	1986	<10	
OSD-U-185-0002	Uranium Oxide Plant Operating Specifications	1992	<10	

1971, a ^{237}Np specification of <1 ppm for a lot composite and <0.3 on 10 lot composites [Corlew 1971] was discussed but not adopted. In 1982, a ^{99}Tc specification of 0.4 ppm was discussed [Miskho 1982, McClusky 1982]. It was implied that there was a 400 ppm limit for ^{99}Tc, but it has never been part of the Hanford product specifications for UO_3 product.

4.1.4 Non-Radiological Contaminants

In addition to primary (radiological) contaminants of concern, the concentration of other constituents were also analyzed and determined to be below specification limits. Since these constituents were based on uranium concentration and the UO_3 process did not significantly reduce the concentration of impurities received in the UNH feed, the same specifications were applied to the separations plants. Infrequently, UNH was

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.

HW-59136
Page -5-

Analysis	Method Ref. HW-24403
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 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

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}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}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.

- 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].

- 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

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

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 (1.30E11) and ^{237}Np (1.56E9) 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 uCi / lb. U for $^{95}ZrNb$, 50 uCi /lb. for the combination of $^{103}Ru^{106}RuRh$, and 2 uCi /lb. for all other isotopes excluding ^{99}Tc. Ten lot average values were also established with the values for $^{95}ZrNb$, ^{103}Ru, and $^{106}RuRh$ and others limited to 10, 25, and 0.5, respectively [Knights 1966].

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 ^{99}Tc from all other beta emitters before quantitation. This was not requested until the restart of the UO_3 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 UO_3 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 UO_3 product. This methodology was originally procured for quantitation of Pu product in the PFP and when the need for analyses of UO_3 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 UO_3 was not recycled and the ^{237}Np concentration was small compared to the concentration

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 ± 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.

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 $\pm$ 10 percent at the 95 percent confidence interval. Neptunium quantitation was held to 95 $\pm$ 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

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

operating in the late 1950s through the mid-1960s, both slightly depleted and low enriched uranium was processed simultaneously in the UO_3 Plant.

4.4.3 Storage/Shipment of UO_3

The UO_3 product was shipped as rapidly as possible when uranium fuel was in short supply nationally until 1964. Table 4-2 summarizes plutonium concentrations in uranium shipped in 1952 and low enriched uranium shipped from 1962 to 1972. Source documents have not been located to complete all time periods and therefore there are discontinuities in the car/lot numbers. The reason for this distinction is that after 1964, LEU UNH was separated from irradiated fuel in the REDOX process while depleted UNH was separated in the PUREX process. The segregation of data provides a measure of REDOX and PUREX capability to purify UNH independently. (Detailed information is shown in Appendix C, Tables 4-1 and 4-2.)

Table 4-2 Summary of UO_3 Shipments In 1952 and LEU 1967 - 1972

Car/Lot #	Month Shipped	Pu (ppb U)		References
		Minimum	Maximum	
007 to 009[(1)]	Mar - 52	<5	<5	[Richards 1952]
010 to 013[(1)]	Mar - 52	<1	<5	[Richards 1952a]
026 to 035	May - 52	<5	<5	[Richards 1952e]
036 to 045 *	Jun - 52	<5	<5	[Richards 1952b]
046 to 057	Jun - 52	<5	<5	[Richards 1952f]
77 to 88 (composite)	Aug - 52		9	[Richards 1952g]
197 to 200 *	Nov - 52	3	3	[Richards 1952c]
E-58 and E-59 *	May - 62	2	4	[Gifford 1964]
E-122 to E-221 and E-223	Jan - 65 to Aug - 67	<1	10	[Madeen 1967]
E1-8-1 to E1-8-12	Sep - 71 to Oct - 71	<1	9	Analytical Report
E1-9-1 to E1-9-15	Oct - 71 to Nov-71	<1	4	Analytical Report
E1-10-1 to E1-10-8	Nov - 71	<1	5	Analytical Report
E2-2-1 to E2-2-12	Feb - 72 to Mar - 72	2	4	Analytical Report

(1) Trial Lots

* **Note**: Weekly/Monthly Reports available for the period 1952-1962 were examined and except for the five lots identified in Section 4.2.1, Process Issues, statements were made that the UO_3 product met the Pu specification. Reports for the years 1954 and 1958 have not been located.

Weekly Summary Reports for the period June 29, 1956 through August 31, 1956, reported plutonium concentrations in UNH produced in the U-Plant averaged 2.6 ppb

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 ($^{95}ZrNb$, ^{103}Ru and $^{106}RuRh$) were documented for the last 52 lots in the data package. The average $^{95}ZrNb$ activity was 4.58 μCi/lb of uranium with a range of 1.23 to 38.76 μCi/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 μCi/lb of uranium and the range is 1.23 to 7.04 μCi/lb. The average ^{103}Ru activity was 0.29 μCi/lb and the range was from unmeasurable to 2.79 μCi/lb. For $^{106}RuRh$, the average activity was 0.44 μCi/lb of uranium and the range was from unmeasurable to 1.78μCi/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

from low enriched UNH continued to be shipped to the National Lead Company of Ohio, Fernald Plant, for blending and recycling, according to specifications presented by HW-79219 Rev. 1964.

Table 4-3 Summary of Drummed UO_3 Product

Date Lot No.	Pu Conc. Low/High (ppb)	Beta % Low/High	Gamma % Low/High	^{103}Ru	$^{106}RuRh$	$^{95}ZrNb$
May 1964 45-1 – 45-15	1 / 5	3 / 5	26 / 42			
Jun 1964 46-1 – 46-16	2 / 8	4 / 6	40 / 69			
Jul 1964 47-1 – 47-5	3 / 5	6	44 / 49			
Aug 1964 48-1 – 49-15	2 / 5	4 / 6	39 / 54			
Sep 1964 49-1 – 49-16	2 / 4	5 / 8	45 / 91			
Oct 1964 410-1 – 410-15	1 / 5	5 / 9	61 / 106			
Nov 1964 411-1 – 411-12	1 / 6	5 / 10	58 / 115			
Dec 1964 412-1 – 412-12	1 / 2	9 / 11	107 / 136			
Jan 1965 51-1	2	9	99			
Feb 1965 52-1 – 52-2	2	12 / 13	176 / 183			
Mar 1965 53-1 – 53-13	1 / 3	10 / 12	30 / 168			
Apr 1965 54-1 – 54-19	1 / 3	9 / 36	111 / 163			
May 1965 55-1 – 55-20	1 / 2	10 / 13	116 / 166			
Jun 1965 56-1 – 56-17	1 / 5	5 / 19	64 / 212			
Jul 1965 57-1 – 57-6	1 / 2	7 / 10	74 / 90			
Aug 1965 58-1 – 58-11	1 / 2	7 / 12	67 / 158			
Sep 1965 59-1 – 59-12	1 / 4	5 / 7	63 / 80			
Oct 1965 510-1 – 510-10	1 / 4	3 / 7	25 / 86			
Nov 1965 511-1 – 511-4	1 / 2	8 / 12	98 / 149			
Dec 1965 512-1 – 512-6	1 / 2	8 / 9	101 / 124			

Table 4-3 (Cont'd) Summary of Drummed UO_3 Product

Lot No.	Pu Conc. Low/High (ppb)	Beta % Low/High	Gamma % Low/High	^{103}Ru UCi/lb U Low/High	$^{106}RuRh$ Uci/lb U Low/High	$^{95}ZrNb$ UCi / lb U Low/High
Jan 1966 61-1 – 61-12	1 / 3	5 / 11	67 / 121			
Feb 1966 62-1 – 62-6	1 / 4	4 / 6	52 / 81			
Mar 1966 63-1 – 63-7	1 / 2	2 / 4	23 / 52			
Apr 1966 64-1 – 64-8	1 / 2	2 / 3	23 / 45			
Sep 1966 69-1 – 69-11	1 / 3	0.3 / 2	4 / 24			
Oct 1966 610-1 – 610-15	1 / 6	2 / 4	21 / 55			
Nov 1966 611-1 – 611-15	1 / 3	3 / 4	34 / 69			
Jan 1967 71-1 – 71-11	1 / 7	2 / 3	33 / 44	0.02 / 0.7	0.3 / 1.8	3 / 4
Feb 1967 72-1 – 72-10	2 / 3	2	31 / 41	0.04 / 0.7	0.03 / 2	3 / 4
Apr 1967 74-1 – 74-16	2 / 4			1E-3 / 0.4	0.05 / 0.6	2 / 39
May 1967 75-1 – 75-12	1 / 5			0.09 / 0.4	1E-3 / 1	1 / 7
Jun 1967 76-1 – 76-2	2			0.08 / 0.4	0.2 / 0.6	6 / 8
5/71 – 7/71 15-1 - 15-13	<1 / <9					
Aug 1971 16-13 - 16-15	<1 / <3					
8/71 – 10/71 17-1 - 17-12	<1 / 6					
8/71 – 10/71 18-1 - 18-12	<1 / 2 (lot 18-8=9)					
10/71 – 11/71 19-1 – 19-11	<1 / 1					
2/72 – 3/72 22-1 – 22-11 Inclusive	2 / 4					
Apr 72 23-1 – 23-2	3 / 4					
4/72 – 6/72 24-1 – 24-8	2					

Analytical data has not yet been located on UO_3 that was produced between 1952 and 1964. It appears that since the maximum plutonium concentration did not change in the specifications and that no evidence was found that any shipments were made without

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. $^{238}U\ (n,2n) \rightarrow\ ^{237}U\ (\beta) \rightarrow\ ^{237}Np$

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

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

Table 4-4 Analyses of UO_3 Produced In/After 1984 at Hanford

Lot No.	Date	Pu ppb	Np * ppb	Tc ppm	^{103}Ru + $^{106}RuRh$ uCi/lb U	$^{95}ZrNb$ uCi/lb U	$^{234}Th/Pa$ uCi/lb U	^{234}U wt %	^{235}U wt %	^{236}U wt%	^{238}U wt %
84-08	6/11/84	2	N/A	N/A	<5	<3	<10	0.008	0.884	0.060	99.088
85-11	6/21/85	<0.5			<8	<6	<10	0.008	0.845	0.065	99.082
85-12	6/25/85	<5			<6	<8	<10	0.010	0.849	0.068	99.073
85-13	6/26/85	<5			<6	<8	<10	0.011	0.852	0.070	99.067
85-14	7/16/85	<5			<6	<8	<10	0.009	0.846	0.068	99.077
85-15	7/16/85	<5			<6	<8	<10	0.009	0.849	0.071	99.071
85-16	7/19/85	<5			<4	<6	<10	0.008	0.848	0.066	99.078
85-17	7/19/85	<5			<4	<6	<10	0.009	0.848	0.067	99.076
85-18	9/30/85	<5	<1000	7	<8	<6	<1	0.009	0.924	0.076	98.991
85-19	9/30/85	<5	<1000	7	<8	<6	<1	0.010	0.942	0.074	98.974
85-20	9/30/85	<5	<1000	7	<8	<6	<2	0.010	0.940	0.072	98.978
86-05	5/6/86	<2	490	12	<6	<4	<31	0.011	0.807	0.080	99.102
86-16	9/22/86	1	400	10	<6	<4	6	0.010	0.873	0.073	99.044
86-23	11/17/86	1	300	8	<6	<4	6	0.011	0.957	0.075	98.957
88-1	3/17/88	2	40	4	<6	<4	9	0.008	0.819	0.074	99.099
88-2	3/17/88	2	120	4	<6	<4	8	0.008	0.950	0.074	99.068
88-3	3/17/88	<1	160	3	<6	<4	10	0.009	0.818	0.073	99.100

Data retrieved from Analytical Data Sheets

* Limited additional ^{237}Np data preceding 1985 are provided in Section 4.5.4, 4.5.5, and Table 4-7.

Table 4-5 Special Process Batch

Lot No.	Date	Pu ppb	^{237}Np ppb	^{99}Tc ppm	^{235}U wt %
93-1	8/30/85	<50 *	8	8	0.298
93-2	8/30/85	<50*	140	8	0.267
93-3	8/30/85	<50*	140	8	0.276

4.5.3 Predicted vs. Actual ^{237}Np Production

The early reports predicted that the ^{236}U would build-up in recycled uranium at a rate of 80-180 ppm per cycle for the single pass reactors and 400 ppm for N Reactor depending on the enrichment level of the blend material. The actual ^{237}Np production was a function of the type of uranium (natural or enriched), the per cent ^{240}Pu in the irradiated fuel and the concentration of ^{236}U in the feed [Schneller 1968]. The predicted ^{237}Np production (starting with fuel of 100 ppm ^{236}U in N Reactor) per ton of uranium is: 2.15 g for 0.947% enriched ^{235}U and 2.88 g for 1.25% enriched ^{235}U. For the reactors with 0.947% enriched ^{235}U, the production is 0.5 g. The relationship of parameters is summarized in Table 4-6 [Schneller 1968].

Table 4-6, Calculated Production of Neptunium in Hanford Reactors

Fuel Enrichment	% ^{240}Pu	^{236}U ppm	g ^{237}Np/MTU
0.71	6	0	1.9
0.71	12	0	6.1
0.94	6	260-340	4.9- 5.4
.94- 1.25	9	60	9.5
.94- 1.25	12	60	18.9
.94- 1.25	20	60	47
2.10	6	-	16.4

4.5.4 Enhancing Production of Neptunium

Enhancing the production of ^{237}Np became of interest because it is the primary precursor for the production of ^{238}Pu, an important isotopic heat source. Therefore, there were efforts to enhance the separation of ^{237}Np from the uranium stream in the REDOX and PUREX processes. Neptunium was isolated in REDOX starting on November 30, 1959 [Weekly Report 1959] and on a semi-continuous basis in PUREX in January 1963.

In PUREX, the inventory of ^{237}Np was allowed to accumulate by reflux between the Backcycle Waste System, the First Decontamination and Partition, and the Final Uranium Cycles. During scheduled plant shutdowns, the ^{237}Np was recovered on a campaign basis [Schmittroh 1995]. In 1959, modifications were made to the PUREX flowsheet to improve the efficiency of the Np recovery operation [Weekly Report February 1959]. In 1963, ^{237}Np was recovered on a semi-continuous basis with the

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 ±1ppb. From 1969-1976 the average Np concentration varies from 11 ppb to

89 ppb. These values all fall within the limited measurement data available from Hanford and Fernald. These data also cover the period that all three uranium recovery processes at Hanford operated.

Table 4-7 Neptunium Analyses on UNH

Reference *Date*	Date of Transfer (1)	Neptunium *Gram/gallon*	Calculated (2) Neptunium (ppb)	
McIntosh 1969	6/3/69 – 7/31/69	<4.50E-5 – 1.78E-4	<20 – 90	
McIntosh 1969a	8/1/69 – 8/25/69	3.01E-5 – 5.18E-4	20 – 270	3 samples 5.17E-4
McIntosh 1969b	10/2/69 –10/25/69	4.81E-5 – 2.71E-4	20 – 140	
McIntosh 1970	12/5/69 – 12/31/69	3.3E-5 – 1.6E-4	20 – 80	
McIntosh 1970a	1/5/70 – 1/30/70	4.2E-5 – 8.15E-4	20 – 430	
McIntosh 1970b	2/3/70 – 2/11/70	6.51E-5 – 3.02E-4	30 – 160	
McIntosh 1970c	4/15/70 – 5/1/70	4.11E-5 – 5.56E-4	20 – 290	

(1) It is recognized that these values are all from near the end of production operations at Hanford. (Concentrations of neptunium measured in the product UO_3 were given in Table 4-4)

(2) Uranium concentration assumed to be 504 g/L , based upon the minimum U concentration defined in the UNH specification for the PUREX Plant.

4.6 Technetium-99

4.6.1 Technetium-99 Concentration in Recycled Uranium

Technetium-99 was produced in Hanford reactors during the production of plutonium from the fission of ^{235}U. ^{99}Tc is formed at the rate of 30.0 mg ^{99}Tc per gram ^{235}U fissioned. Most recent ORIGEN2 calculations representing the production history of all the single-pass reactors and N-reactor indicate that 1960 kg ^{99}Tc were produced at the Hanford Site [Watrous 1997]. While the uncertainties of the ORIGEN2 results vary for different nuclides, for ^{99}Tc the uncertainties are expected to be less than 10%. The quantity of ^{99}Tc co-processed with the recovered UO_3 is a function of the solvent extraction process used to recover uranium and the distribution coefficients (organic/aqueous phases) for ^{99}Tc. Based on analyses and reported distributions coefficients, between 20% to 30% of the ^{99}Tc was co-processed with the UO_3 and shipped offsite [Roberts 1971, Schmitroth 1995].

4.6.2 Hanford Technetium Measurements

Analytical measurements for ^{99}Tc in the UO_3 product were not routinely performed prior to 1985 at Hanford. Most of the uranium shipped in the 1980s was from N-Reactor weapons-grade production. The measured ^{99}Tc concentrations in UO_3 recovered from PUREX were in the range of 7-8 ppm (Table 4-4). These values are well below the 400

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 1.74E5 d/m/gram in PUREX-produced UNH and 2.9E4 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 +/-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

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.

Table 4-8 Typical Uranium Isotopic Ratio of LEU Produced In/After 1970

Lot	Year	^{234}U wt %	^{235}U wt %	^{236}U wt %
329A - C	May 1970		0.813	
330A - C	June 1970		0.806	
331A - C	June 1970		0.808	
332A -C	June 1970		0.813	
333A & B	June 1970		0.817	
E - 335	1971		0.865	
E - 336	1971		0.865	
18-1 - 18-12	1971		0.846	
19-1 - 19-16	1971		0.847	
110-1 - 110-8	1971		0.844	
22-1 - 22-12	1972		0.748	
84-08	1984	0.008	0.884	0.06
84-1 to 84-21	1984		0.85	0.06
85-11	1985	0.008	0.845	0.065
85-012	1985	0.01	0.849	0.068
85-13	1985	0.011	0.852	0.07
85-014	1985	0.009	0.846	0.068
85-015	1985	0.009	0.849	0.071
85-016	1985	0.008	0.848	0.066
85-017	1985	0.009	0.848	0.067
85-018	1985	0.009	0.924	0.076
85-019	1985	0.01	0.924	0.074
85-020	1985	0.01	0.94	0.072
86-05	1986	0.011	0.807	0.08
86-16	1986	0.01	0.873	0.073
86-23	1986	0.011	0.957	0.075
88-1	1988	0.008	0.819	0.074
88-2	1988	0.008	0.85	0.074
88-3	1988	0.009	0.818	0.073
Average		**0.009**	**0.86**	**0.071**

Table 4-9 Deeply Depleted UO_3 Isotopic Data

Lot	Year	Wt % of ^{234}U	Wt % of ^{235}U	Wt % of ^{236}U
93-1	1985	0.004	0.298	0.016
93-2	1985	0.002	0.267	0.015
93-3	1985	0.004	0.276	0.017
Average		**0.003**	**0.280**	**0.016**

Table 4-10 ^{235}U Isotopic Data of Pre-1972 Product or Depleted UO_3

Lot*	Year	^{235}U wt %	Lot *	Year	^{235}U wt %
7	1952	0.68	56-1 to 56-17	1965	0.650
8	1952	0.68	57-1 to 57-6	1965	0.647
9	1952	0.67	58-1 to 58-11	1965	0.638
036 to 045	1952	0.64	59-1 to 59-12	1965	0.652
007 to 009	1952	0.68	510-1 to 510-10	1965	0.65
010 to 013	1952	0.66	511-1 to 51-4	1965	0.654
197-200	1952	0.64	512-1 to 512-6	1965	0.63
9T	1952	0.64	Adjustments	1966	0.647
45-1	1964	0.63	Adjustments	1966	0.638
45-2	1964	0.632	Adjustments	1966	0.651
45-3	1964	0.633	Adjustments	1966	0.65
45-4	1964	0.636	Adjustments	1966	0.654
45-5	1964	0.635	Adjustments	1966	0.633
45-6	1964	0.634	Adjustments	1966	0.63
45-7	1964	0.641	Adjustments	1966	0.648
45-8	1964	0.637	Adjustments	1966	0.644
45-9	1964	0.644	Adjustments	1966	0.647
45-10	1964	0.642	Adjustments	1966	0.65
45-11to 45-15	1964	0.644	Adjustments	1966	0.622
46-1 to 46-16	1964	0.646	61-1 to 61-12	1966	0.63
47-1 to 47-5	1964	0.620	62-1 to 62-6	1966	0.648
48-1 to 48-15	1964	0.634	63-1 to 63-7	1966	0.644
49-1 to 49-16	1964	0.640	64-1 to 64-8	1966	0.647
410-1 to 410-15	1964	0.636	69-1 to 69-11	1966	0.65
411-1 to 411-12	1964	0.642	610-1 to 610-15	1966	0.643
412-1 to 412-12	1964	0.640	75-1 to 72-12	1967	0.633
Adjustments	1965	0.648	76-1	1967	0.633
Adjustments	1965	0.647	71-1 to 71-11	1967	0.654
Adjustments	1965	0.642	72-1 to 72-7	1967	0.658
Adjustments	1965	0.650	72-8 to 72-10	1967	0.653
51-1	1965	0.644	74-2 to 74-16	1967	0.629
52-1 to 52-2	1965	0.644	1-5-2 &1-5-7	1971	0.658
53-1 to -53-13	1965	0.648	1-5-1 – 1-5-10 & 1-5-13	5/25/71 - 7/26/71	0.651
54-1 to 54-19	1965	0.647	1-6-2 - 1-6-15	7/1/71 - 8/3/71	0.651
55-1 to 55-20	1965	0.645	1-7-2 - 1-7-12	9/7/71 - 10/12/71	0.66 est.
			Average		**0.644**

* Data collected from Uranium Oxide – Source Data – Revised by Month [Murphy 1971]

4.8 Contaminants in Uranium Metal

Uranium metal was received at the Hanford Site in the form of bare rods or ingots for use as fuel for the reactors. The fabrication operations in the 300 Area processed this metal into fuel elements or "slugs" suitable for use in Hanford reactors. There were several steps in this process including dipping the metal in a tin, lead, or chromium solution; extruding or pressing into aluminum or later zircaloy tubes; and cutting these elements to length and welding end caps to form a complete seal. These operations were labor intensive and required extensive exacting physical and reactivity tests to be met. Prior to the mid-1960 time period, the desired ^{235}U content of uranium metal for fuel was attained by using natural U or processing recycle UO_3 through a GDP. This process reduced the concentration of fission product radionuclides before the metal ingot production. Analyses performed by the supplier on the metal were accepted at Hanford after an initial test program demonstrated the reliability of the measurement system. Products of uranium decay were expected to be present and fuel fabrication operations at Hanford were tailored to be consistent with the guidelines established at that time.

4.8.1 Non-radiological Composition

Incoming uranium metal was shipped from NLO after being shown to meet specifications of chemical impurities with maximum concentrations given for the elements in Table 4-11 [Gill 1963].

Table 4-11 Chemical Specifications for Uranium Metal Billets

Element	Min / Max Concentration (ppm)	Element	Max Concentration (ppm)
Be	/ 6	Mg	15
B	/ 0.25	Mn	20
Cd	/ 0.2	Ni	90
C	300 / 650	N	50
Cr	/ 20	Si	50
Cu	/ 65	Zr	75
H	/ 2.0	U	99.81% (Minimum)
Fe	115 / 200		

By 1985, aluminum was added to the "routine" N-Reactor fuel specification when FEDC Alloys were processed for irradiation. In addition, uranium ingots were to be analyzed periodically for 20 "incidental" non-radiological impurities [WHC-SP-0056 1987].

These specifications are rather recent updates, but appear to be consistent with earlier requirements in which the importance of maintaining the quality of the incoming uranium metal was recognized. In 1952, chemical analyses were being improved in the 300

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, Hartcorn 1954, Gardner 1956].

4.8.3 Radiological Contaminants

Prior to 1952, no uranium oxide was recycled and as a result, Pu, ^{237}Np, ^{99}Tc and fission product contamination were not present in the metal received for fuel fabrication. Between 1952 and 1962, UO_3 was processed through the gaseous diffusion plants, which significantly reduced the concentration of Pu and ^{237}Np in the enriched product to

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

indicates traces of ^{237}Np entered the product stream, but no evidence was found for the presence of Pu to have been carried over into the product.

Table 4-12 Transuranic Content in Ingot Composites

Composite No.	Pu (ppb)	^{237}Np (ppb)	^{99}Tc (ppm)
85-1	3.0	155	3.2
85-3	2.7	211	3.2
85-8	3.5	179	3.7
85-9	2.4	243	4.0
85-19	2.7	185	5.5
85-25	2.2	192	3.2
85-26	2.0	198	5.8
85-33	2.9	185	4.2
85-42	7.0	204	1.8
85-43	4.3	166	2.9
85-52	8.1	192	2.3
85-53	2.2	134	3.4
85-59	4.3	185	0.7
85-60	5.2	179	0.9
85-61	3.0	153	0.8

In a personal communication with Fernald personnel [J. Neyer and C.W. Lower April 6, 2000], the following information was verified:

1. No specification was present for radionuclides in uranium metal prior to 1986. At that time, a 10 ppb upper threshold for Pu was listed.

2. Prior to 1962, Fernald only made metal using U processed through a GDP. Limited data indicated this material contained approximately 30 parts plutonium per trillion parts uranium. Np was usually in the same order of magnitude.

3. Direct blending of recycle uranium with GDP enriched uranium began in 1962 and the first shipment of metal produced from this blend occurred in 1963. Because of dilution in the process, the Pu averaged about 1 ppb.

4. Uranium metal received at Hanford from 1963 to the end of receipts (1980s) had bounding levels of Pu about 1 ppb for 0.95% ^{235}U and 6 – 7 ppb in 1.25% U. During 1985, GDP tails were blended with the metal products fabricated and the values listed incorporate those tails. The weighted average of Pu in all UO_3 used for direct blending was about 2.6 ppb.

5. The Np concentration is bounded during the same time period at a high of about 211 ppb. They recognized the wide variation in the neptunium concentrations.

6. ^{99}Tc concentrations in metal during the 1980s ranged from 8 to 15 ppm.

Section 4
Contaminants in Recycled Uranium

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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