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History of Uranium-233 (^{233}U) Processing at the Rocky Flats Plant

In Support of the RFETS Acceptable Knowledge Program

RS-090-056

1 April 1999

ICF KAISER

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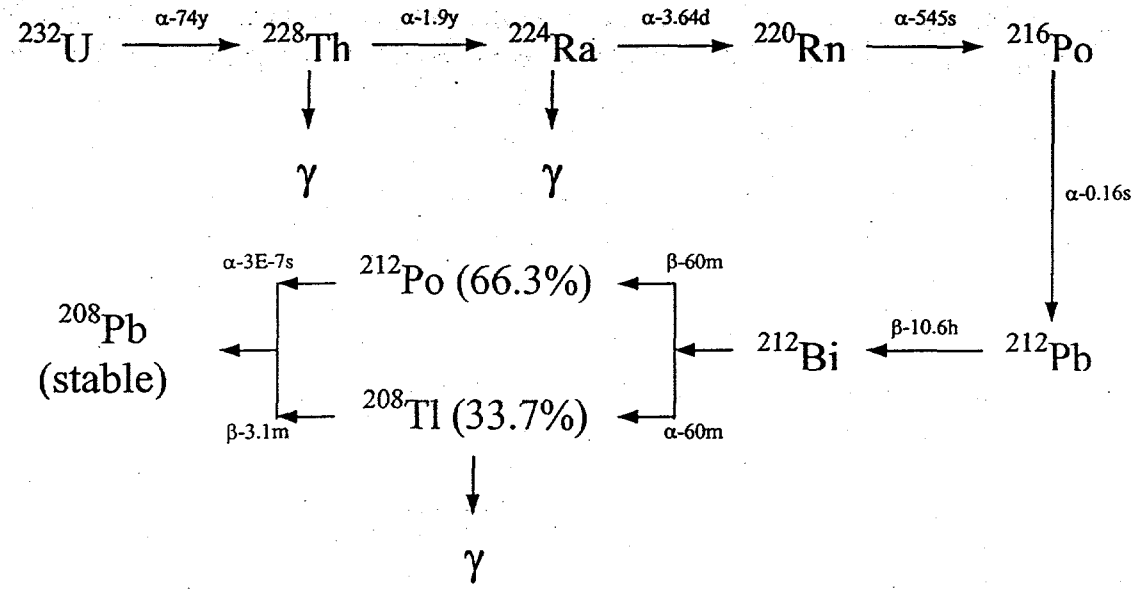
1.0 INTRODUCTION

The purpose of this report is to document the processing of Uranium-233 (^{233}U) at the Rocky Flats Plant (Rocky Flats Environmental Technology Site). Detailed descriptions of the ^{233}U metal processing/component manufacturing, material recovery, and waste handling are provided. ^{233}U inventory data is documented showing the Material Balance Areas (MBAs), timeframes, and specific locations where ^{233}U was processed or where inventory was maintained. This report develops and provides a key reference in documenting Acceptable Knowledge (AK) used to meet Waste Isolation Pilot Plant (WIPP) Waste Acceptance Criteria (WAC). The information provided in this report can be utilized as the starting point for determining potential ^{233}U content in applicable residue waste streams. It can also be utilized to eliminate ^{233}U concerns for various sub-populations of waste streams.

2.0 URANIUM-233 (^{233}U) PROCESSING

Initial Efforts

The first processing operations at Rocky Flats (RF) involving Uranium-233 (^{233}U) occurred in 1965. This project resulted from a special order request to fabricate a number of items out of ^{233}U metal. Feed material arrived at RF from Oak Ridge (OR) in special transport casks as uranyl nitrate solution ($UO_2(NO_3)_2$). Isotopic analysis showed ^{233}U composition at 97.13%, with the balance consisting primarily of other uranium isotopes (^{234}U, ^{235}U, ^{236}U, and ^{238}U). In addition, there was a presence of approximately 50 parts per million (ppm) ^{232}U (as a contaminant) which was important because daughter products from its decay [i.e. Thorium-228 (^{228}Th), Radium-224 (^{224}Ra), and Thallium-208 (^{208}Tl)] give off high-energy gamma radiation[1], which presents a significant external health hazard to processing workers. Figure 1 summarizes the decay scheme for ^{232}U to stable ^{208}Pb, and shows the points where high energy radiation is released.



Half-lives shown in seconds (s), minutes (m), days (d), and years (y). Some high-energy gamma radiation from daughter products is shown.

Note: ^{232}U is present as an isotopic impurity. ^{233}U processed in 1965 contained approximately 50 ppm ^{232}U, while that processed in 1976-77 contained only 7-8 ppm ^{232}U.

Figure 1 – ^{232}U Decay Scheme.

[1] For example, 79.5% of ^{208}Tl gamma is in excess of 100 keV, with 36% emitted as 2.614 MeV gamma. RFP-2817, Chemistry R&D Semiannual Progress Report for January – July 1978, 2/10/79.

Because of this hazard, plans for processing of ^{233}U focussed on isolating it from other RF plutonium (Pu) and uranium (U) process streams. The first step involved precipitating Th (and a small amount of Tl) from the feed material in order to minimize their associated radiation hazard. Subsequent processing steps were then expedited in order to minimize the build-up of ^{232}U decay products over the time that processing occurred. The flow path in Figure 2 summarizes the ^{233}U processing steps.

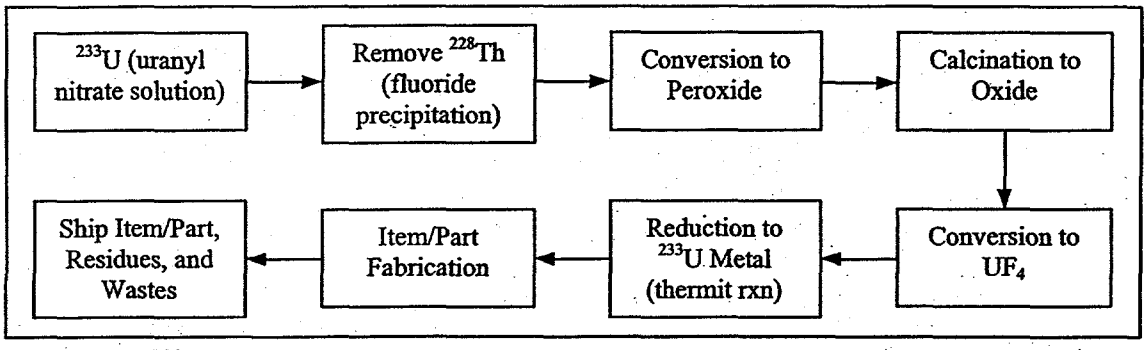


Figure 2 – ^{233}U Processing Steps.

^{233}U processing[2] began in Building 771[3] where the uranyl nitrate solution was transferred to receiving tanks. Fluoride precipitation was then used to remove the "hot" (highly radioactive) daughter products (primarily ^{228}Th), and the uranium was converted to peroxide. The peroxide was shipped to Building 881 where it was calcined to an oxide, which in turn was converted to uranium tetrafluoride (UF_4), and reduced to ^{233}U metal using a thermit reaction. The metal was then cast into feed ingots, which were in turn recast into pieces from which the final parts were fabricated. Casting and machining operations took place in Building 881, while other fabrication steps were handled in Building 883. Final component assembly and inspection occurred in Building 777.

All wastes and residues were collected, treated, packaged, and shipped to various locations off-site. Uranium oxides and green salt residues were converted to uranyl nitrate solution in Building 771 and returned to OR in the original shipping casks[4]. Some casting skulls and machining chips were burned to oxide in Building 881 and subsequently converted to a nitrate solution along with the other oxides[5]. Aqueous wastes went to Building 774 for disposal, while low level wastes were placed in drums and shipped to the Atlantic Richfield Company (ARCO)[6] in Idaho for burial.

Records show a residual inventory of 2 grams ^{233}U at the conclusion of this project (June 1965).

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[2] All processing steps are described in detail in CD65-3184, Investigation of Uranium 233-235 Crossover Incident, June 11, 1965.

[3] Initially two-digit numbers were used to identify buildings. When the three-digit system was introduced the first digit usually was a duplicate of the second. Thus, Building 71 became 771, 81 became 881, etc. The three-digit system of identification in use today will be used throughout this report.

[4] CD65-3184, Investigation of Uranium 233-235 Crossover Incident, June 11, 1965.

[5] CD73-5096, Non-Pu Physical Inventories, Inventory Differences, and NOL (through FY-1988).

[6] AEC/ERDA/DOE facility operated by Atlantic Richfield Company (ARCO). This site is located in Idaho and received many radioactive wastes that were disposed of by burial.

Subsequent ^{233}U Operations

Inventory records indicate that kilogram (kg) quantities of ^{233}U were also received, processed, and shipped at RF over the next three years, 1966-1968[7]. Smaller quantities, typically less than 1 kg, show up on inventories during the period 1969-1973, where projects included casting small metal ^{233}U disks[8]. During 1974-1977, kg quantities of ^{233}U again arrived and were processed at RF, however by 1974, uranium processing had been stopped in Building 881. Two special order projects worked on between 1974 and 1977 again involved manufacturing a number of components from ^{233}U metal. Feed material arrived at RF from OR as uranium oxide, was converted to UF_4, and reduced to ^{233}U metal by a thermit reaction. The metal product was broken into chunks, re-cast into feed ingots, and then cast into the final components using a vacuum induction furnace. By comparison to the 1965 project, the feed material was oxide, not uranyl nitrate solution, and all chemical processing, metal reduction, and casting operations took place in Building 771. It is likely that machining steps were handled in Building 779A, and that final component inspection was done in Building 777.

The ^{232}U contamination in this metal was substantially lower, running around 7-8 ppm, compared with 50 ppm in 1965. Still, gamma radiation levels were fairly high and a fluoride precipitation step was used on feed material, and from time to time on in-process material, to remove the "hot" daughter products and reduce the external radiation hazard to workers. It was also observed that surface gamma radiation readings from processed material dropped with each successive processing step: fluorination, casting, and recasting. Concurrently, radiation levels from the casting skulls increased, suggesting segregation of high-radiation daughter products from the ^{233}U was taking place.

Because of the health safety concerns over radiation from isotopic impurities and decay/daughter products within the ^{233}U stream (as well as possible contamination of production Pu and U process streams), the 1974-1977 waste materials generated during processing of ^{233}U on these special order projects were carefully isolated from "normal" RF process streams. The isolated waste material was treated, packaged and disposed of as before (in 1965). These waste materials included:

- Metal reduction residues
- Casting skulls
- Machine turnings
- Oily machine filters and towels
- Contaminated glove box materials

The process of carefully segregating the waste is mentioned in a series of Research and Development (R&D) progress reports. Typical statements included: "Machining scrap

[7] CD73-5096, Non-Pu Physical Inventories, Inventory Differences, and NOL (through FY-1988).
[8] RFP-1848, Casting of Multiple U-233 Metal Target Disks, W. V. Conner and D. L. Baaso, May 1972 (unclassified report).

will be processed for return to LLL[9], and all contaminated waste will be packaged for disposal"[10], and "All metal scrap and residues generated during fabrication were to be recovered and returned to LLL or shipped to the Idaho waste storage site....."[11] Metal reduction residues were shipped to LLL. Casting skulls and machine turnings were burned to oxide, processed again to remove ^{232}U daughter products, and then converted to a stable oxide for shipping to LLL. Contaminated glove box materials were segregated into combustibles and non-combustibles, packed into 55-gallon drums, and shipped to the Idaho waste storage site[12]. The final disposition of oily machine filters and oily towels is somewhat uncertain. In RFP-2680 the statement is made: "The oily machine filters and towels were disposed of in the Rocky Flats incinerator." Interviews with a number of personnel involved with Building 771 operations, including the incinerator, gave conflicting stories as to whether ^{233}U ever went through this incinerator. However, it does appear likely that some oily, low level ^{233}U wastes were incinerated, as they could not have been shipped "as-is" because of the combustion hazard they presented.

Inventory data indicates the presence of kg quantities of ^{233}U into Fiscal Year (FY) 1982, which suggests other project activities continued with this material. In May 1982 the inventory dropped to 267 grams, and in December 1983, was recorded as 0 grams. From August 1984 through March 1988, the last date in the inventory summary document[13], ^{233}U inventory is listed as 13 or 14 grams from month to month. This suggests that all operations with ^{233}U essentially ceased after 1982.

Summary

Processing operations involving ^{233}U were carried out at RF starting in 1965 and ending in 1982. Activities included chemical processing of various uranium compounds, conversion to metal, casting, metal fabrication, and waste and residue disposal.

All processing reports state that ^{233}U materials and wastes were carefully monitored and segregated from other RF processing streams. Residues were shipped to either LLL or OR, and wastes were sent to Idaho for burial.

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[9] Called Lawrence Livermore Laboratory (LLL) and Lawrence Livermore Research Laboratory (LLRL) in earlier years, this facility is now known as Lawrence Livermore National Laboratory (LLNL).

[10] RFP-2546, Chemistry Research and Development Semiannual Progress Report for the period January – June 1976, dated February 14, 1977.

[11] RFP-2745, Chemistry Research and Development Semiannual Progress Report for the period July through December 1977, dated May 29, 1978.

[12] Information on treatment and disposal of these items is described in Report RFP-2680, Chemistry Research and Development Semiannual Progress for the period January through June 1977, dated October 12, 1977.

[13] CD73-5096, Non-Plutonium Physical Inventories, Inventory differences, and NOL — operations to date. Though apparently created in 1973, this document was updated through 1988.

3.0 ^{233}U INVENTORY

Records

Though ^{233}U inventory data records exist, there are some questions that affect interpretation of the numbers. One is whether the data describes only ^{233}U or alternatively, all uranium isotopes in an item, of which ^{233}U is the prime constituent. The following discussion will outline some of the basis for uncertainty.

The Inventory Process

It is important to understand the process used in arriving at inventory numbers. Each building is divided into Material Balance Areas (MBAs). An inventory of accountable nuclear materials is maintained for each MBA. The sum of individual nuclear material/isotope inventory numbers for each MBA provides the plant total inventory. Inventories were conducted monthly, and the starting value for each MBA was the ending inventory number from the month before. All material received into an MBA was added to this starting value, and all material shipped out of the MBA was subtracted. Material transactions were accompanied by Material Transfer Reports, which contained isotope assay data, as well as quantities received or shipped. An estimate was made for measured discarded waste, also called normal operating loss (NOL), and this also was subtracted from the starting value. Finally, all material on hand was weighed and the total compared with a calculated value of what the inventory should be (using the various record data just described). The inventory calculations are summarized below.

Current MBA Inventory	=	**Previous MBA Inventory**	+	**Material Received In MBA**	-	**Material Shipped From MBA**	-	**Normal Operating Loss (NOL)**

Weights were taken for the total mass of items and it was implicitly understood that other isotopes were present (i.e. a piece of ^{233}U was weighed and the value recorded, but this weight included all other uranium isotopes present as well). Nuclear Material Safeguards was expected to use analytical information to calculate actual constituent isotope amounts[14] (for example, in the case of Pu streams, averages for Pu isotope percentages were used). Any difference between expected/calculated inventory on hand and the inventory measured became material unaccounted for, or MUF. This calculation is summarized below.

Material Unaccounted For (MUF)	=	**Expected and/or Calculated Inventory**	-	**Measured Inventory**

The MUF values could vary from month to month, especially since many of the adjustment numbers related to ongoing operations were calculated based on assumed process and operations efficiencies (based on historical data). For example, oxide sent

[14] Interview with Bill Conner.

for isotopic assay would have an estimated U (or Pu) value, which would be confirmed after the assay was completed. If this occurred during the following inventory period, an adjustment of data would have to be made for the previous month in which an estimate had been recorded. It appears that MUF data for ^{233}U suffered from many unknowns and this is reflected in large swings and subsequent corrections.

Data

As previously discussed, initial ^{233}U activities at RF occurred in 1965. Unfortunately, an incident involving cross-contamination with ^{235}U occurred which resulted in preparation of a detailed summary report[15] on this particular project and event. Exhibit N, U-233 Material Balance Report (MBR), of this incident report lists the initial total quantity of U received as uranyl nitrate solution on April 25, 1965. The ^{233}U assay is listed in a separate column, and the difference between these two values was supposedly made up of ^{234}U, 0.19% ^{235}U, ^{236}U, and ^{238}U, plus any additional impurities, such as ^{232}U which was present at a level around 50 ppm. Taking the ratio of ^{233}U grams to total U grams gives 97.14% ^{233}U. This MBR inventory of ^{233}U as of the end of May 1965 lists an excess of total uranium but a shortfall (MUF) of ^{233}U. The incident report mentions that all of the received uranyl nitrate solution may not have been transferred from the shipping cask. This was not of concern since the same cask would be used to return all left-over material to OR at the end of the project, so it would ultimately be accounted for. It is possible that inventory discrepancies, especially for the ^{233}U, may have resulted from the manner in which calculations (using assay percentages) were made.

Two additional reports containing ^{233}U inventory data are also available. One is a Confidential document that contains a number of inventory notation sheets for a few specific years as well as an overall table of non-Pu inventory summary data for the period 1965 through April 1977. Numbers shown are for the end of each fiscal year (June 30 for years through FY-1976 and September 30 starting with FY-1977), and the table is labeled U-233. Entries are provided for "Beginning Inventory", "Receipts", "Shipments", "NOL", MUF, and "Ending Inventory" categories. The listed amount of ^{233}U received in 1965 is 2.7% higher than the total U value, or 5.7% higher than the ^{233}U value given in the MBR table mentioned above. The other inventory report is the Non-Pu Physical Inventories, Inventory Differences and NOL document, CD73-5096. It contains inventory data by month for Code 70 material, which is total ^{233}U. The value given for material received in April 1965 is 0.8% higher than that listed in the MBR table for total uranium, or 3.8% higher than the ^{233}U number. The relative values of these inventory numbers in these three documents are shown in Table 1.

[15] CD65-3184, Investigation of Uranium 233-235 Crossover Incident, Dated June 11, 1965.

Table 1 – 1965 ^{233}U Inventory Record Data Comparison.[16]

	^{233}U	Code-70 Material "Total ^{233}U"	Total Uranium Material	"Corrected" Numbers (Red)
Material Balance Report (Exhibit N to Incident Report CD65-3184	$V_2 = 97.13\% V_1$		V_1	
^{233}U Inventory Summary for 1965 – 4/77 (C/RD)		102.76% V_1 (105.7 % V_2)		97.4% V_1 (100.2% V_2)
Non-Pu Physical Inventories, Inventory Differences, and NOL (CD73-5096)		100.8% V_1 (103.8% V_2)		

Note: V_1 represents the MBR inventory for total uranium received (as reported in the MBR) while V_2 is the ^{233}U assay in this amount. Inventory entries in the other reports differed and their relative value to these two reference numbers are listed in this table.

These numbers, therefore, call into question whether values listed in various non-Pu inventory reports represent just the ^{233}U or, alternatively, all uranium isotopes in lots of which ^{233}U was the major constituent.

The Confidential non-Pu inventory summary sheet mentioned above also contains a second set of entries in red. These are slightly lower values that seem to be corrections to ^{233}U (assuming the original entries were actually for total U). For example, the red entry for material received in 1965 is 94.78% of the originally listed value. While in the right direction, this correction is less than the 97.13% U-233 assay reported in CD65-3184. "Corrected"/red entries are listed for most inventory categories ("Beginning Inventory", "Receipts", "Shipments", "NOL", and "Ending Inventory") in each of the years. However, the ratio of corrected to original entry values varies considerably, from 94.7% up to around 98%. No mention is made on the table of the basis for the red entries and it is surmised that they represent an attempt to track actual ^{233}U values. To add to the confusion, the initial (non-red) table entries from the Confidential non-Pu inventory summary agree with FY year-end inventory numbers listed in CD73-5096, which are for Code 70 material, or total ^{233}U. These discrepancies between numbers suggest there may have been some confusion in labeling the data when inventories were taken, and thus calls into question whether various non-Pu inventory reports correctly track ^{233}U or, alternatively, total uranium isotopes for lots in which ^{233}U was the major constituent.

MUF numbers in the Confidential non-Pu inventory summary table also vary substantially from year to year, typically ranging from 0 to 100 grams, but are significantly higher on four occasions. A few hundred grams MUF in FY-1968 is accompanied by a similar magnitude Shipper/Receiver adjustment that same year. Another year listed a reduction in MUF of some 106 grams.

[16] Specific inventory values can be found in the referenced reports.

Large swings in inventory numbers, along with the confusing situation regarding whether the data is ^{233}U or total U, suggests caution should be exercised when attempting to calculate running inventory from these numbers. Because total mass weights were recorded during inventory each month, it appears possible that corrections for actual ^{233}U isotope might not have been made, which is why inventory data (as in report CD73-5096), particularly for 1965, appears high.

According to CD73-5096, Non-Plutonium Physical Inventories, Inventory Differences, and NOL, most of any remaining ^{233}U inventory was shipped from RF in 1983. However, the validity of current RF ^{233}U amounts is not significantly affected by the inventory/assay issue discussed above since the error would be around 5% and for the 8 grams currently listed this amounts to only 0.4 grams.

Inventory data does not include explanations for changes in either total amounts or MUF, so the increase in year-end ^{233}U inventory from 0 grams (the value from December 1982 through June 1984) to 13 grams in July 1984 is not understood. Furthermore it is listed at 13 grams through May 1991, except for February – October 1986 when it was 14grams.

In June 1991 the ^{233}U inventory changed downward to 8 grams, where it remains today (March 1999). Searching inventory records turns up the following information as to present locations of ^{233}U contained in drums.

Table 2 – ^{233}U Inventory at Rocky Flats, January 1999.

Building	Room	Bin	IDC	Drum #	Description	^{233}U Amount
371	3189	16	480	D44501 42	Metal	3 grams
371	2306	N/A	480	D21552 42	Metal	1 gram
371	3420	N/A	330	D30664 42	Dry Combustibles	2 grams
371	3420	N/A	330	D21548 42	Dry Combustibles	1 gram
371	3420	N/A	337	D36455 42	Plastic	1 gram

Waste

Waste containing ^{233}U could have been generated wherever this material was processed. This includes both known wastes from specific operations at the time, as well as potential residual contamination in waste generated from these gloveboxes during subsequent clean-up long after processing was over. Waste drums were marked with the MBA of the source. Table 3 lists those buildings where ^{233}U operations took place over the years, and thus identifies where waste drums containing or being in contact with residual ^{233}U might have originated. The assignment of buildings in any given year is based on interpretation of report information and interviews conducted with personnel involved with or who had knowledge of ^{233}U operations at RF. It is possible that operations were conducted in additional or fewer years.

Table 3 – Likely Locations of ^{233}U Processing at RF for 1965 - 1983.

	559	771	774	777	777A	779A	881	883
1965		X	X	X	X		X	X
1966		X	X	X	X		X	X
1967		X	X	X	X		X	X
1968		X	X	X	X		X	X
1969		X					X	
1970	X	X					X	
1971	X	X					X	
1972	X	X					X	
1973	X	X	X				X	
1974		X	X					
1975		X						
1976	X	X	X			X		
1977	X	X	X	X	X	X		
1978	X	X	X	X	X	X		
1979	X	X	X	X	X	X		
1980	X	X	X	X	X	X		
1981	X	X	X	X	X	X		
1982	X	X	X	X	X	X		
1983		X	X					

Building 559 – Analytical Laboratories
Building 771 – Chemical processing, analytical laboratories, waste processing
Building 774 – Waste Disposal
Building 777 – Non-Destructive Testing, inspection, manufacturing processing
Building 777A – Assembly
Building 779A – Machining
Building 881 – Chemical processing, casting, machining
Building 883 –Fabrication

Non-Pu Workbooks, containing monthly ^{233}U inventory records, were used to identify specific MBAs that handled this material, as well as the time periods when this occurred. This information is summarized in Table 4. Waste drums generated by these MBAs, especially during that time when they were actively working with ^{233}U, potentially could contain this material as a contaminant. Table 5 Identifies the building and title of the MBA's listed in Table 4.

Table 4 – Reported ^{233}U Inventory by Calendar Year.

Bldg.	MBA	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86	87	88	89	90	91	92	93	94	95	96
559	361-31		X	X		X	X	X	X	X	X	O		X	X											
707	1576-07							X																		
750	0374-32		(NOL Write-off)					X																		
771	0217-80														O	O	O	O	O	O	O					
	0223-71				O			X													X	O				
	0233-71		X	X	O	X	X	X	X	O	X	X														
	0233-74								O																	
	0367-71			X		X	X	X	X	O	X	O														
	0383-33						X	X																		
	1371-30																									O
	1371-31																				O	O	O	O	O	
	1371-43																								O	
	1374-31						X	X		X	X															
	1374-34						O	O																		
776	0233-76					X																				
777	0233-77		O			O	O	X																		
	1177-53		O					O	O		O	O														
779	0233-80					X	X	X																		
779A	0233-79		X	X		X	X	X																		
881	0216-71		O	O																						
	0362-31		X																							
991	1144-91											O														
UNK	0218-71		X																							
	0233-81		O																							

X = MBA's reporting inventory and MUF losses and gains in those years. O = MBA's reporting inventory and no MUF in those years.

UNK = Unknown Building

Table 5 – MBA Identification and Building Location.

MBA	Material Balance Area Title	Building
0216-71	R&D Chemistry Instrumentation	881
0217-80	Aqueous Process Chemistry Development	771
0218-71	UNK	UNK
0223-71	R&D Plutonium Metallurgy	771
0233-71	R&D Chem. Tech. – U-233 Account	771
0233-74	Special Recovery – U-233 Account	771
0233-76	Quality Acceptance – U-233 Account	776
0233-77	Assembly – U-233 Account	777
0233-79	R&D Machining – U-233 Account	779A
0233-80	R&D Lab. Chem. Tech – U-233 Account	779
0233-81	UNK	UNK
0361-31	Analytical Labs. Production Support	559
0362-31	Lab. Service Uranium Analysis	881
0367-71	Analytical Labs. Building 771	771
0374-32	Solid Waste	750
0383-33	Chem. Standards Lab.	771
1144-91	Product Warehouse Shipping	991
1177-53	Metal Fabrication Disassembly	777
1371-30	Drum & Crate Storage	771
1371-31	Nonspec. In-Process	771
1371-43	Building 771 Drum Holding	771
1374-31	Special Recovery Operations – Aqueous	771
1374-34	Special Recovery – Special Alloy	771
1576-07	Metal Fab. Machining	707

UNK = Unknown Material Balance Area Title and Building

Even though ^{233}U operations occurred over limited periods of time, residual contamination and hold-up material within process lines could inadvertently be part of discarded waste generated from these lines as they underwent subsequent periodic cleaning over the years. This is particularly true in areas where monthly inventories at some time showed MUF losses and gains for ^{233}U, indicated by an "X" in the table. Waste drums from these MBAs are suspect as possibly containing ^{233}U in small amounts. If no MUF was reported by an MBA in a given year, this is indicated by an "O" in the table. A number of MBAs never reported ^{233}U MUF in any year, and thus it would be unlikely that waste drums coming from them in years subsequent to their reporting any ^{233}U inventory would contain this contamination. These include 0217-80, 1371-30, 1371-31, 1371-43, 1177-53, 0216-71, 1144-91, and 0233-81. For example, there are MBAs where packaged material only passed through, and no processing activities occurred that might have generated contaminated waste, or opportunities for material hold-up that could result in reported MUF.

Only Non-Pu Workbooks for fiscal years FY-1973 through FY-1996 were located, so MBA-specific information is not listed during the first eight years when ^{233}U was processed at RF. However, the previous table identifies those buildings where it was handled, and this may suffice to flag drums from this period that could potentially contain ^{233}U contaminated waste.

Since drum counters were set up to look for Pu isotopes and ^{235}U, ^{233}U was not detected and its presence in waste drums would be unknown. Presence of ^{233}U in current waste drums would have to be verified by counting.

Summary

Variations between ^{233}U inventory values, as compared between different documents, probably should not cause concern. The most notable variance occurs between the Material Balance Report (Exhibit N in the 1965 incident report) and the non-Pu inventory record (CD73-5096) for material received in 1965. The listed 2-5% "adjustment" downward of ^{233}U values, represented by the red numbers in the Confidential summary inventory document, could indicate a potential, systematic error. However, since there was a concerted effort to ship all ^{233}U material and wastes from RF over the years, any such error would have been applied equally to both shipments and receipts, and today it would only matter as to now much ^{233}U remains in the RF inventory. Since this appears to be on the order of only 8 grams ^{233}U, a correction of 2 - 5% would result in lowering the amount by only 0.1 – 0.4 grams, which is less than the recorded data sensitivity. Therefore, it would be more relevant to focus on whether the listed inventory is all-inclusive of ^{233}U on site or not, and identifying any other waste drums that might possibly contain ^{233}U, but are currently not labeled as such.

4.0 CONCLUSIONS

Based on process knowledge, obtained through interviewing individuals who either worked directly with ^{233}U, or in areas where process stream wastes and residues were handled, it appears likely that little ^{233}U material waste remains on plant site, as past and present inventory records indicate. There had been a concerted effort to ship not only special order items but all associated residues and wastes from RF as quickly as possible. This action was motivated by two goals: (1) to minimize radiation exposure to RF workers from any stored materials, as high gamma radiation was associated with ^{233}U through decomposition daughter products, primarily from ^{232}U impurities, and (2) to minimize contamination of normal RF process streams by ^{233}U. Shipments could be verified by examining associated records.

It is possible that ^{233}U exists as a contaminant in waste drums generated by MBAs where it was once processed or handled, and this "contaminated" waste could have been generated any time subsequent to actual processing operations. Knowing both building and MBA where ^{233}U processing occurred, as well as those years when there was a reported inventory is data key to the determination of whether a residue is potentially contaminated with ^{233}U. This allows waste drums to be identified for further analysis, such as drum counting using instruments set to detect the types of radiation and energy levels expected from $^{233}U/^{232}U$ daughter products.

Inventory data recorded over the years reflects uncertainties that were common but typically within tolerance limits accepted at the time. The existence of relatively small quantities of ^{233}U in the present inventory provides an opportunity to verify this data and perform calibration measurements that would allow easy validation of ^{233}U in other suspected containers. The five drums listed in inventory should be counted using scanners set to detect radiation of the type and energy levels expected from $^{233}U/^{232}U$ and related radioactive isotopes and daughter products. Scanning these drums will show (1) whether radiation from these small quantities can be detected and any effect other packing materials in the drums might have on shielding, and (2) allow radiation measurements to be calibrated against the quantities present. These results would then allow a procedure to be developed for scanning unknown drums with the purpose of identifying whether they also contain any ^{233}U.

5.0 REFERENCES

1. CD65-3184, "Investigation of Uranium 233-235 Crossover Incident," June 11, 1965.

2. RFP-2546, Chemistry Research & Development Semiannual Progress Report for the period January – June 1976, February 14, 1977, S/RD.

3. RFP-2680, Chemistry Research & Development Semiannual Progress Report for the period January – June 1977, October 12, 1977, S/RD.

4. RFP-2745, Chemistry Research & Development Semiannual Progress Report for the period July – December 1977, May 29, 1978, S/RD.

5. RFP-2817, Chemistry Research & Development Semiannual Progress Report for the period January – July 1978, February 10, 1979, S/RD.

6. CD73-5096, Non-Pu Physical Inventories, Inventory Differences, and NOL – operations to date (information through FY-1988), S/RD.

7. Inventory summary data on Pu-238 and U-233 covering the period 1957 through April 1977, C/RD.

8. RFP-2983, Chemistry Research & Development Semiannual Progress Report for the period January – June 1979, June 3, 1980, S/RD.

9. RFP-1848, "Casting of Multiple U-233 Metal Target Disks," W. V. Conner and D. L. Baaso, May 1972 (U).

10. "Preparation of Actinide Metal Targets Using Special Casting Techniques", W. V. Conner, Nuclear Instruments and Methods, v. 102, p. 417, 1972.

11. CD74-4225, Non-Pu Workbook, FY-74 & FY-75, S/RD.

12. CD76-2492, Non-Pu Workbook, FY-76 & FY-76A, S/RD.

13. CD77-648, Non-Pu Workbook, FY-77 & FY-78, S/RD.

14. CD78-4074, Non-Pu Workbook, FY-79, S/RD.

15. CD80-1627, Non-Pu Workbook, FY-80, S/RD.

16. CD81-533, Non-Pu Workbook, FY-81, S/RD.

17. CD84-1550, Non-Pu Workbook, FY-82, S/RD.

18. CD84-1543, Non-Pu Workbook, FY-83, S/RD.

19. CD85-118, Non-Pu Workbook, FY-84, S/RD.

20. CD85-4954, Non-Pu Workbook, FY-85, S/RD.

21. CD86-317, Non-Pu Workbook, FY-86, S/RD.

22. CD86-5665, Non-Pu Workbook, FY-87, S/RD.

23. CD87-5410, Non-Pu Workbook, FY-88, S/RD.

24. CD88-5008, Non-Pu Workbook, FY-89, S/RD.

25. CD89-01361, Non-Pu Workbook, FY-90, S/RD.

26. RFCD90-02563, Non-Pu Workbook, FY-91, S/RD.

27. RFCD91-03823, Non-Pu Workbook, FY-92, S/RD.

28. RFCD92-02257, Non-Pu Workbook, FY-93, S/RD.

29. RFCD93-01653, Non-Pu Workbook, FY-94, S/RD.

30. NMA94-0148, Non-Pu Workbook, FY-95, S/RD.

31. NMA95-0167, Non-Pu Workbook, FY-96, S/RD.

6.0 APPENDICIES

A1. Interviews on ^{233}U Operations

A2. Interviews

A2.1 Ed Vejvoda Interview
A2.2 Don Cox Interview
A2.3 Jack Weaver Interview
A2.4 Rod Hoffman and Larry Wilson Interview
A2.5 Don Baaso Interview
A2.6 Chuck Tesitor Interview
A2.7 Larry Crisler Interview
A2.8 Jerry Haynes Interview
A2.9 Ed Kinneson Interview
A2.10 Dick Cleavenger Interview
A2.11 Jim Deyo Interview
A2.12 Bill Conner Interview
A2.13 Duane Dunn Interview
A2.14 Brenda Douglass and Fred Lyons Interview

A1. Interviews on ^{233}U Operations

One 1965 report on ^{233}U processing contains much detail describing where various activities took place at RF on that specific project. Subsequent ^{233}U work is less well documented, with information provided on processing steps but not locations where they occurred. To fill in some of these details, a number of individuals were interviewed who had either worked on ^{233}U projects or in process areas where ^{233}U materials or waste streams are likely to have been present. Most people admitted to uncertain recollections, as the time periods discussed were some 20 or more years ago. However, by cross-checking statements, likely scenarios for a few of the operations could be identified. There were two particular ^{233}U related items in question. One dealt with incineration of some hazardous wastes, specifically oily filters and towels from machining operations. The other concerned where such machining work took place after 1974, the timeframe when processing of U in Building 881 had ceased. The following notes summarize recollections on these topics as well as other notable information on ^{233}U work.

Conclusions from Interviews

Though some contradictory statements were encountered and individual memories appeared far from perfect, a somewhat consistent picture emerges of ^{233}U operations at RF.

- Machining of ^{233}U during the mid-1970s and later most likely took place in Building 779A.

- R&D personnel used hot plates or small muffle furnaces to burn some combustible wastes.

- Some wastes containing ^{233}U were likely burned in the Building 771 production incinerator. It is likely that any ^{233}U contamination was at a low enough level as not to cause any concern over possible RF process stream contamination.

- Wastes and residues were carefully isolated from normal RF process streams, monitored, and shipped off site as soon as possible.

No definitive explanation was found for what happened to oily machine filters and towels.

A2. Interviews

A2.1 **Ed Vejvoda**	**Manager of various groups in chemistry and chemical processing operations during 1960-1980** • Oily filters possibly were shipped to Building 774 and off site • ^{233}U waste went to Idaho. Residues to Oak Ridge. • Waste going to Idaho may not have been marked as containing ^{233}U. Such waste shipments likely contained a variety of constituents and were just marked as "waste". • In the late 1960s some ^{233}U was made into samples for use by radiochemistry as diagnostic tools/standards. • Machining of ^{233}U might have taken place in Building 776 or Building 777. (From others comments on Building 776, this appears very unlikely and Building 777 seems unlikely as well.) • Chemistry analysis probably took place in both Building 559 and Building 771 labs. • Process lines where ^{233}U was handled were cleaned up prior to and after its' handling. Small amounts of ^{233}U may have been disposed of mixed in with general waste generated during subsequent cleanup of glovebox lines.
A2.2 **Don Cox**	**Building 771 incinerator operator and foreman** • NMC maintained run sheets on everything that passed through the incinerator. • No ^{233}U waste was incinerated. • NDA scanned all drums of material submitted for incineration. • Individual packages went through a can counter.
A2.3 **Jack Weaver**	**Building 771 incinerator and chemical operator, and foreman** • No uranium was run through the incinerator. • R&D had small (approximately 8"x8"x8") muffle furnaces where small quantities of items could have been incinerated. • He did not know specifically what R&D might have burned.
A2.4 **Rod Hoffman & Larry Wilson**	**Special Orders Engineering during the 1960s and 1970s** • Machining of ^{233}U in the 1960s took place in the metal-floored room in Building 881. • Small ^{233}U projects were worked on in Building 881 into the early 1970s (prior to 1973 or 1974). • When Th "strikes" were performed to remove highly radioactive daughter products of ^{232}U, there would have been a decrease in the ^{233}U inventory numbers to account for that material which had decayed. (Assay information suggests that these quantities would have been very small and insignificant compared to MUF or NOL values). • Furnaces for processing uranium, which were removed from Building 881 in the mid-1970s probably, contained some ^{233}U contamination.

A2.5 **Don Baaso**	**Technician and engineer in chemistry R&D groups** • He does not believe any uranium was processed through the Building 771 production incinerators (this was his initial statement). • In the 1976-1978 timeframe, casting of ^{233}U metal for special projects took place in Building 771. • Machining of ^{233}U most likely occurred in Building 779A. • Because the oily filters and towels associated with machining (some from cleanup activities) were potentially ignitable, something had to be done with them as they could/should not be shipped in their "as-is" condition. • Towels may have been burned in muffle furnaces or on hot plates • Filters would not fit into small R&D muffle furnaces. Don does not remember what was done with them. • Most ^{233}U projects basically involved processing to remove highly radioactive daughter products from the ^{232}U contaminant/impurity as well as making of metal. • Don believes Building 771 production equipment was "commandeered" for use on ^{233}U projects (during 1976-1978 timeframe). Thus, he "suspects" that the incinerator was also used for treating some waste (contrast to his opening statement). His statement to this effect in the chemistry R&D Semiannual Progress Report, RFP-2680 dated 10/12/77, "must be true" as he would not have made it if he didn't have knowledge of this at the time.
A2.6 **Chuck Tesitor**	**Incinerator operations, including manager, from 1968-1982** • Because of the combustible/fire hazard associated with oily waste, small quantities containing contaminants deemed to be at insignificant levels so as not to pose a stream contamination risk were burned in the Building 771 incinerator. The resulting ash was dissolved in HNO_3, filtered, and the filtrate processed through ion exchange to recover metal. • All inputs to the incinerator went through NDA. • R&D would have burned chips on hot plates or in muffle furnaces. • It is not likely that muffle furnaces would have been used to burn oily waste because the gases generated would have created a potentially explosive mixture.
A2.7 **Larry Crisler**	**Engineer in chemistry R&D** • It is likely that ^{233}U machining took place in Building 779A. • Scrap was sent to Building 771 for further treatment. • Oily filters were likely cut up and calcined in small muffle furnaces. • There was a project in the early 1970s that made some ^{233}U metal disks. • Building 881 was removed from uranium handling and processing around 1967. • There was another ^{233}U project in the early 1980s. Work on this project was done in Building 771 and Building 779.

A2.8 **Jerry Haynes**	**Health Physics Safety** • He had no recollection as to where ^{233}U might have been machined in the late 1970s. This, in spite of his involvement with protecting workers from excessive radiation hazards.
A2.9 **Ed Kinneson**	**Special Orders Engineering** • Ed could not remember where ^{233}U machining might have taken place in the late 1970s.
A2.10 **Dick Cleavenger**	**Special Orders Engineering** • He could not remember where ^{233}U machining might have taken place in the late 1970s. • Remembers some machining of special materials in Building 777A, but doesn't know whether these included uranium or ^{233}U.
A2.11 **Jim Deyo**	**Special Orders Engineering** • He could not remember where ^{233}U machining might have taken place in the late 1970s.
A2.12 **Bill Conner**	**Engineer in Chemistry R&D groups** • Bill's initial reaction was that he didn't remember sending any wastes to the Building 771 incinerator. • After reviewing the Chemistry R&D Semiannual Progress Reports describing ^{233}U work on special order projects, he believes that the statements mentioning Building 771 incineration of oily filters and towels from machining operations were indeed true. • Machining of ^{233}U metal took place in Building 779A. • There were some ^{233}U metal target disks cast on another special project in 1971. • A special order in the early 1980s involved ^{233}U. This is likely associated with the ^{233}U inventory changes recorded at that time. • Total mass was weighed during inventory. It was "implicitly" understood that isotopes other than the primary one were present. Safeguards was expected to calculate isotope amounts based on average stream assay knowledge. • Drum counters were being developed in the 1960s. They used a combination of gamma and neutron detectors and accuracy was ± 100%. • There was no way that RF drum counters could assay for ^{233}U. • Drum counters were not calibrated for ^{233}U. • Special drums were used to collect miscellaneous wastes of non-routine isotopes, typically from special order work.
A2.13 **Duane Dunn**	**Nuclear Records Management** • Inventory numbers were used to maintain a record of material handling. • They could not be relied upon as a 100% accurate statement of inventory. • Weighing, recording, and other administrative errors occurred, leading to corrections being entered from time to time.

A2.14 **Brenda Douglass & Fred Lyons**	**Members of Nuclear Materials Safeguards group; involved with inventory records management** • NOL typically was measured material in discard waste that was below the economic discard limit (EDL). • Process hold-up was accounted for in MUF. • Do not believe there was any gamma counter scanning for ^{233}U. • There could be traces of ^{233}U in drums containing Pu below the EDL that had not been checked for ^{233}U. Pu waste from glovebox lines would be an example of this. • One needs to identify MBAs for areas that handled ^{233}U and look at waste packaged during the time period to find those drums which might contain ^{233}U. MBAs with ^{233}U inventory MUF would be likely candidates, both at the time of processing and over successive years when waste from clean-up could also contain ^{233}U from residual contamination.