**DISPOSITION OF URANIUM-233 (^{233}U) IN
PLUTONIUM METAL AND OXIDE AT THE ROCKY
FLATS ENVIRONMENTAL TECHNOLOGY SITE**

RECEIVED
JUL 14 2000
OSTI

Prepared for:

**Safe Sites of Colorado
Plutonium Stabilization and Packaging System (PuSPS)
Rocky Flats Environmental Technology Site
Golden, Colorado 80403-8200**

Prepared by:

IT CORPORATION

A Member of The IT Group

**Cameron J. Freiboth
Frank E. Gibbs, PhD.
235 Central Park Square
Los Alamos, New Mexico 87544**

**March 1, 2000
Revision 1**

"REVIEWED FOR CLASSIFICATION/UCNI"
BY: Cameron J. Freiboth - 4/21/00
DATE: 03/01/00

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, make any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

DISPOSITION OF URANIUM-233 (^{233}U) IN PLUTONIUM METAL AND OXIDE AT THE ROCKY FLATS ENVIRONMENTAL TECHNOLOGY SITE

Prepared for:

**Safe Sites of Colorado
Plutonium Stabilization and Packaging System (PuSPS)
Rocky Flats Environmental Technology Site
Golden, Colorado 80403-8200**

Prepared by:

IT CORPORATION

A Member of The IT Group

**Cameron J. Freiboth
Frank E. Gibbs, PhD.
235 Central Park Square
Los Alamos, New Mexico 87544**

**March 1, 2000
Revision 1**

TABLE OF CONTENTS

EXECUTIVE SUMMARY	1
1.0 INTRODUCTION.....	2
1.1 Background.....	2
1.2 Preliminary ^{233}U Processing History	2
2.0 ^{233}U HANDLING AND PROCESSING.....	3
2.1 Radiation Concern	3
FIGURE 1 – Decay Scheme for ^{232}U to ^{208}Pb	3
2.2 Initial ^{233}U Processing.....	4
FIGURE 2 - ^{233}U Processing Steps.....	4
2.3 Subsequent ^{233}U Operations.....	5
2.4 Fissile Material Additives	7
2.5 ^{233}U Handling, Processing, and Waste Packaging Summary	7
3.0 ^{233}U INVENTORY / MATERIAL BALANCE AREAS.....	8
3.1 The Inventory Process.....	8
3.2 ^{233}U Inventory / Material Balance Area Information.....	9
TABLE 1 – MBA's / Building Inventories of Uranium-233 for 1973 – 1999	9
TABLE 2 – MBA, Title Identifier and Corresponding Building.....	9
3.3 Potential Processing Areas / Waste Origination Points	10
TABLE 3 – Likely Locations of Uranium-233 Processing at RFP from 1965 to 1982.....	10
3.4 Summary Calculations.....	11
3.5 ^{233}U Assay Limitations	12
3.6 ^{233}U Inventory / Material Balance Area Summary	12
4.0 CONCLUSIONS.....	13
5.0 REFERENCES.....	14

EXECUTIVE SUMMARY

This report documents the position that the concentration of Uranium-233 (^{233}U) in plutonium metal and oxide currently stored at the Department of Energy (DOE) Rocky Flats Environmental Technology Site (RFETS) is well below the maximum permissible stabilization, packaging, shipping and storage limits. The ^{233}U stabilization, packaging and storage limit is 0.5 weight percent (wt%), which is also the shipping limit maximum. These two plutonium products (metal and oxide) are scheduled for processing through the Building 371 Plutonium Stabilization and Packaging System (PuSPS). This justification is supported by written technical reports, personnel interviews, and nuclear material inventories, as compiled in the "History of Uranium-233 (^{233}U) Processing at the Rocky Flats Plant In Support of the RFETS Acceptable Knowledge Program," RS-090-056, April 1, 1999 [1]. Relevant data from this report is summarized for application to the PuSPS metal and oxide processing campaigns, and is based on the following:

^{233}U Handling, Processing, and Waste Packaging

- Uranium-232 (^{232}U), a residual isotope present in ^{233}U metal, produces decay products that result in high-energy gamma emissions that are severe enough to significantly increase radiation fields in Radiological Controlled Areas (RCAs). This radiological concern and potential for contamination of the Plutonium-239 (^{239}Pu) stream, led to a conscious effort to segregate waste associated with ^{233}U processing, followed by the expedient offsite shipment of final product, surplus material, and waste byproducts.
- Small quantities of Americium-241 (^{241}Am) and Neptunium-237 (^{237}Np) were routinely added to ^{239}Pu components as radiochemical tracers (measurement tool) during testing at the Nevada Test Site (NTS). ^{233}U was never purposely added to ^{239}Pu as a tracer material.

^{233}U Material Balance Area / Inventory

- The Material Balance Areas (MBA's) which reported a ^{233}U inventory were those that did not generally have a large or significant amount of plutonium metal or oxide in-process.
- Inventory records support the fact that ^{233}U products, surplus, and waste materials were segregated and expeditiously shipped offsite.
- ^{233}U handling and processing ended in 1982, while the generation of significant amounts of plutonium metal processing and oxide generation continued until 1989.

Reference material utilized to develop the historical summaries and plutonium metal and oxide position are included at the end of this report.

1.0 INTRODUCTION

1.1 Background

Recommendation 94-1 from the Defense Nuclear Facility Safety Board (DNFSB) requires that all Rocky Flats Environmental Technology Site (RFETS) plutonium (Pu) metal and oxide be packaged for long-term storage by May 2002. There are approximately 4,600 items at RFETS that must be processed to meet this requirement [2]. These metal and oxide items must be stabilized, packaged, and stored in accordance with the Department of Energy (DOE) Standard for "Stabilization, Packaging, and Storage of Plutonium-Bearing Materials," DOE-STD-3013-99 [3]. This standard:

"provides criteria for stabilization of plutonium-bearing materials at DOE facilities to safe and stable forms that can be packaged and placed in storage with minimal surveillance for up to 50 years."

Plutonium-bearing metals and oxides must contain at least 30-wt% plutonium, with no lower limit on the amount of uranium (U). The standard does not apply however, to material containing greater than 0.5-wt% Uranium-233 (^{233}U). This 0.5-wt% limit is based on the DOE Office of Material Disposition (MD) "Acceptance Criteria for Plutonium Bearing Materials to be Dispositioned by Immobilization," DOE/MD-0013 [4]. Shipment of this material is planned in Type 9975 shipping containers. The "Safety Analysis Report - Packaging (SARP)" which analyzes shipping activities, also allows for a maximum ^{233}U content of 0.5-wt% [5].

The purpose of this report is to establish the position that plutonium metal and oxide planned to be processed through the PuSPS contains ^{233}U at a level well below the 0.5-wt% maximum permissible stabilization, packaging, and shipping limit.

1.2 Preliminary ^{233}U Processing History

^{233}U was processed at RFETS (also referred to as the Rocky Flats Plant or RFP) in various campaigns between the years of 1965 to 1982. An historical perspective on the processing of ^{233}U at the Rocky Flats Plant was developed in support of the RFETS Waste Isolation Pilot Plant (WIPP) Waste Acceptance Criteria (WAC) Acceptable Knowledge Program [1]. It contains detailed descriptions on ^{233}U metal processing/ component manufacturing, material recovery, and waste handling. It also contains ^{233}U inventory data showing the Material Balance Areas (MBAs), timeframes, and specific locations where it was processed or where inventory was maintained.

While this historical document was developed in support of WIPP/WAC, the information is also applicable to plutonium metal and oxide that will be processed through the PuSPS. Information from this historical report is summarized and presented in the following sections.

2.0 ^{233}U HANDLING AND PROCESSING

2.1 Radiation Concern

The first processing operations at the Rocky Flats Plant (RFP) involving Uranium-233 (^{233}U) occurred in 1965. 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). The material also contained approximately 50 parts per million (ppm) contaminant ^{232}U whose decay scheme daughter products Thorium-228 (^{228}Th), Radium-224 (^{224}Ra), and Thallium-208 (^{208}Tl), emit high-energy gamma radiation [6]. Because of this high-energy radiation emitted from ^{232}U decay, ^{233}U processed from 1974 to 1977 contained approximately 7 to 8 ppm ^{232}U [7,8].

The decay scheme for ^{232}U to stable ^{208}Pb showing these points where high-energy gamma is released is summarized below in FIGURE 1. ^{208}Tl for example, emits gamma in excess of 100 kilo-electron volts (keV), with 36% emitted as 2.614 million-electron volts (MeV) [8,9,10]. A typical package containing 3 kilograms (kg) of ^{233}U with 100 ppm ^{232}U would generate a radiation field of more than 25 rad per hour (r/hr) of gamma radiation 1 foot (30 cm) from the package [9]. A 50 ppm ^{232}U content equates to approximately 13 r/hr at 1 foot, and with extrapolation, a 5 to 10 ppm content would emit approximately 5 r/hr at 1 foot. These high-energy radiation levels present a significant external health hazard to processing workers and because of this, great care was taken to isolate it from the personnel using gloveboxes and from other RFP plutonium and uranium process streams to preclude cross contamination. ^{232}U also has a high alpha activity associated with it, which also dictates isolation and segregation [10].

FIGURE 1 – Decay Scheme for ^{232}U to ^{208}Pb .

2.2 Initial ^{233}U Processing

The first ^{233}U processing operations at Rocky Flats occurred in 1965. This project was initiated as a special order request to fabricate a number of items out of ^{233}U metal. As previously mentioned, great care was taken to isolate ^{233}U from other plutonium and uranium processing streams to reduce the possibility of cross-contamination. Processing steps were expedited to minimize the build-up of ^{232}U decay products. FIGURE 2 below summarizes the flow path for the ^{233}U processing. Descriptions for the individual steps shown follow FIGURE 2 [6].

FIGURE 2 - ^{233}U Processing Steps.

- (1) ^{233}U feed material arrived from the Oak Ridge Plant (ORP) in special transport casks as uranyl nitrate solution ($\text{UO}_2(\text{NO}_3)_2$).
- (2) The nitrate solution was transferred to receiving tanks. Fluoride precipitation was used to remove the highly radioactive daughter products (primarily ^{228}Th).
- (3) Once the precipitation was completed, the uranium was converted to peroxide, which was then shipped to Building 881, (4) where it was calcined to an oxide.
- (5) The calcined oxide was then converted to uranium tetrafluoride (UF_4 or green salt), (6) that was then reduced to ^{233}U metal using a Thermit reaction.
- (7) The ^{233}U 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.
- (8) 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 ORP in the original shipping casks. 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. Aqueous wastes went to Building 774 for disposal, while low level wastes were placed in drums and shipped to the Atlantic Richfield Company (ARCO) in Idaho for burial.

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

2.3 Subsequent ^{233}U Operations

Inventory records indicate that kilogram (kg) quantities of ^{233}U were also received, processed, and shipped at the RFP over the next three years, 1966 to 1968. Smaller quantities, typically less than 1 kg, are reported during the period 1969 to 1973, where projects included casting small metal ^{233}U disks [11]. During 1974 to 1982, kg quantities of ^{233}U were again processed at the RFP [12,13]. By 1974, ^{233}U processing in Building 881 was stopped. During this time frame, two Special Order projects were worked which again involved manufacturing a number of components from ^{233}U metal. Feed material arrived at RFP from ORP as uranium oxide that was converted to UF_4 , and reduced to ^{233}U metal by a Thermit reaction [14]. 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 the material processed from 1974 to 1977 was substantially lower than the 1965 level of 50 ppm, running around 7-8 ppm [7,8]. Gamma radiation levels were still fairly high, however, and a fluoride precipitation step was used on feed material to remove the decay products and reduce the radiation hazard to workers. In addition, a study was conducted where fluoride precipitation was used on in-process material. It was observed that gamma radiation surface readings taken from in-process material dropped with each successive processing step: fluorination, casting, and recasting. Concurrently, radiation levels from the casting skulls increased, suggesting segregation of ^{233}U high-radiation decay products was taking place in the skulls [8].

As with the previously discussed 1965 processing activities, the radiation concern within the ^{233}U stream as well as possible contamination of other production process streams still existed. The 1974 to 1977 feed, finished part, surplus and waste materials generated during the ^{233}U Special Order processing activities were again carefully isolated from the normal RFP plutonium and uranium process streams to preclude cross-contamination. These isolated waste materials including metal reduction residues, casting skulls, machine turnings, oily machine filters and towels, and contaminated glovebox materials, were treated, packaged and disposed of in the same manner as in 1965.

The process of carefully segregating the materials and waste is mentioned in a series of RFP Research and Development (R&D) Technical Reports. Typical statements contained in various reports includes:

"The equipment in intimate contact with the ^{233}U ... was all new" [11]

"...the integrity of ^{233}U was maintained throughout this project and no cross contamination with other actinide elements or uranium isotopes occurred" [11]

"The casting skulls and machine turning will be converted to an oxide for shipment to LLL" [7], where LLL was the Lawrence Livermore Laboratory

"Machining scrap will be processed for return to LLL, and all contaminated waste will be packaged for disposal" [7]

"All of the metal scrap and residues from this project were to be recovered and returned to LLL or shipped to the Idaho waste storage site" [14]

"Contaminated glove-box materials were segregated into combustible and noncombustible categories. Five drums were generated and will be shipped to the Idaho waste storage site" [14]

"All metal scrap and residues generated during fabrication of the uranium-233 device were to be recovered and returned to LLL or shipped to the Idaho waste storage site" [15]

"The casting skull, machine turnings, and other residues from this project are being collected" [15]

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 glovebox materials were segregated into combustibles and non-combustibles, packed into 55-gallon drums, and shipped to the Idaho waste storage site. Interviews with a number of personnel directly involved with these activities supported the Technical Reports statements regarding segregation of in-process ^{233}U and the expedient shipment of wastes to Idaho [1].

The final disposition of oily machine filters and oily towels is somewhat uncertain. One of the Technical Reports states:

"The oily machine filters and towels were disposed of in the Rocky Flats incinerator" [14].

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 [1]. 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. While this presents the possibility that incinerator ash and other residue clean-out materials may contain ppm quantities of ^{233}U contamination, it would not show up in the plutonium metal and oxide streams. By not being a part of the plutonium metal and oxide streams, it would not be processed through the PuSPS and therefore would not be subsequently shipped to the Savannah River Site (SRS).

Inventory data, which is discussed in more detail in Section 3 of this report, 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 a residual level of 267 grams, and in December 1983, was recorded as 0 grams. From August 1984 through March 1988, the last date in the inventory summary documentation, ^{233}U inventory is listed at a residual level of around 14 grams from month to month. This suggests that all operations with ^{233}U essentially ceased after 1982 [13].

2.4 Fissile Material Additives

Radiochemical tracers were typically added to Special Order ^{239}Pu components for measurement purposes during testing at the Nevada Test Site. Two examples include Neptunium-237 (^{237}Np) and Americium-241 (^{241}Am). ^{237}Np typically was added at a level of 0.1-wt %, while ^{241}Am was added to feed ingots at levels of 3000 to 4000 ppm (0.30 to 0.40-wt %) with a final component concentration of 1500 ppm (0.15-wt %). A review of technical reports and personnel interviews indicated no evidence of the use of ^{233}U as a radiochemical tracer. As discussed previously, ^{233}U items were manufactured separately from ^{233}U metal and specifically segregated from the ^{239}Pu stream to prevent cross-contamination. This contamination not only represented a radiological concern, but could significantly affect the manufacture and performance of ^{239}Pu components. A representative study of the chemistries associated with twenty-one Special Order castings manufactured between 1975 and 1980 for LANL was made which showed that 4 of the components contained ^{237}Np , and confirmed the levels did not exceed 0.1-wt %. All of the castings contained small amounts of ^{241}Am , which were at levels less than 0.15-wt %. The chemistries indicated uranium levels up to 550 ppm. These levels of uranium are not broken out by isotope, however, so it can not be stated that there was absolutely no ^{233}U contained in the plutonium metal and oxide [1].

2.5 ^{233}U Handling, Processing, and Waste Packaging Summary

Various activities and operations involving ^{233}U were carried out at the Rocky Flats Plant starting in 1965 and ending in 1982. This included the chemical processing of various uranium compounds, conversion of oxides to metal, casting, metal fabrication, and waste and residue disposal. Great care was taken to keep the ^{233}U activities separate from all other plutonium and uranium operations. This was due to the gamma radiation concern associated with ppm contaminant levels of ^{232}U and subsequent decay products, as well as the need to avoid material cross-contamination. Technical reports state that ^{233}U materials and wastes were carefully monitored and segregated from other Rocky Flats processing streams. Residues were shipped to either Lawrence Livermore Laboratory or the Oak Ridge Plant, while wastes were sent to Idaho for burial. Some combustible materials may have been incinerated at the Rocky Flats Plant, as they would have not been shippable in their initial condition. Interviews with current and former RFP personnel support these reports with statements regarding the conscious segregation of ^{233}U feed, process, product and waste materials. Based on this information, and the indications that ^{233}U was not purposely added to plutonium castings as a radiochemical tracer, there is no concern over ^{233}U being contained in plutonium metal and oxide at levels which exceed the Materials Disposition and SARP level of 0.5-wt%. While incinerated materials (ash) may contain some ^{233}U , material from this waste stream will not be processed through the PuSPS, and therefore would not end up being transferred to the SRS.

3.0 ²³³U INVENTORY / MATERIAL BALANCE AREAS

3.1 The Inventory Process

Inventory information for ²³³U is tracked to the nearest gram because it is considered accountable material. Plutonium and uranium facilities at the RFP are sub-divided into Material Balance Areas (MBAs), where inventories of accountable nuclear materials are maintained. The sum of individual nuclear material/isotope numbers for each MBA provides the total plant-site inventory. During the time that ²³³U existed in fairly large quantities (1965 to 1982), inventories were conducted monthly, and the starting values for each MBA was the ending inventory number from the month before. All materials received into an MBA were added to this starting value, and materials shipped out of the MBA were 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), which was also 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). These 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. An example is a weighed piece of ²³⁹Pu where the inventory value recorded included all plutonium isotopes present. Standard Nuclear Material Safeguards procedures required that actual constituent isotope amounts were calculated using analytical chemistry information. Any difference between expected/calculated inventory on hand and the measured inventory became material unaccounted for, or MUF. This calculation is summarized below.

Material Unaccounted For (MUF)	=	Expected/ 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. For example, plutonium oxide sent for isotopic assay would have an estimated plutonium value that 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.

²³³U inventory data reviewed indicated that NOL and MUF were reported in various years that inventory was present at the RFP.

3.2 ²³³U Inventory / Material Balance Area Information

A review of various historical RFP non-plutonium inventory records shows various quantities of ²³³U present from 1973 to 1999. TABLE 1 below summarizes the MBA's reporting inventories of ²³³U during these years and includes the buildings associated with these MBA's [16-37]. Inventory records for 1965 to 1972 were not available and therefore excluded from this table.

MBA	Building	73	74	75	76	77	78	79	80	81	82	83	84	85	86	87	88	89	90	91	92	93	94	95	96	97	98	99
1375-47	371																											
0361-31	559	X	X		X	X	X	X	X	X	O	X	X	X													O	O
1576-07	707						X																					
0374-32	750						X	(NOL Write-off)																				
0217-80	771														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																			X									
1371-31																				O	O	O	O	O	O	O		
1371-43																								O				
1374-31						X	X		X	X																		
1347-34						O	O																					
0233-76	776				X																							
0233-77	777	O			O	O	X																					
1177-53		O					O	O		O	O																	
0233-80	779				X	X	X																					
0233-79	779A	X	X		X	X	X																					
0216-71	881	O	O																									
0362-31		X																										
1144-91	991										O																	
0218-71	Unknown	X																										
0233-81		O																										

X = MBA's reporting inventory and MUF losses/gains. O = MBA's reporting inventory and no MUF. Unknown = Unknown Building

TABLE 1 – MBA's / Building Inventories of Uranium-233 for 1973 – 1999.

Titles of MBA's reporting ²³³U inventory in TABLE 1 above are summarized in TABLE 2. Listed are the MBA titles for those years when inventories were reported and the associated Building where the MBA was located [38].

MBA	Material Balance Area Title	Building	MBA	Material Balance Area Title	Building
0216-71	R&D Chemistry Instrumentation	881	0367-71	Analytical Labs. Building 771	771
0217-80	R&D Chemistry Tech Development	771	0374-32	Solid Waste NMC	750
0218-71	Unknown	UNK	0383-33	Chem. Standards Lab.	771
0223-71	R&D Plutonium Metallurgy	771	1144-91	Product Warehouse Shipping	991
0233-71	R&D Chem. Tech. – U-233 Account	771	1177-53	Metal Fabrication Disassembly – BBIS	777
0233-74	Special Recovery – U-233 Account	771	1371-30	Drum & Crate Storage	771
0233-76	Quality Acceptance – U-233 Account	776	1371-31	Non-Specification In-Process	771
0233-77	Assembly – U-233 Account	777	1371-43	Building 771 Drum Holding	771
0233-79	R&D Machining – U-233 Account	779A	1374-31	Special Recovery Operations – Aqueous	771
0233-80	R&D Lab. Chem. Tech – U-233 Account	779	1374-34	Special Recovery – Special Alloy	771
0233-81	Unknown	UNK	1375-47	Calorimetry/POC Packaging	371
0361-31	Analytical Labs. Production Support	559	1576-07	Metal Fabrication Machining	707
0362-31	Lab. Service Uranium Analysis	881			

TABLE 2 – MBA, Title Identifier and Corresponding Building.

3.3 Potential Processing Areas / Waste Origination Points

While inventory records by MBA are an indicator on potential ²³³U processing in an area, some of the areas may have just been used for material storage. Waste containing ²³³U could have been generated wherever the material was processed. This includes wastes from specific operations at the time, as well as potential residual contamination in waste generated from the gloveboxes during subsequent clean-ups, long after processing was over. TABLE 3 below identifies those buildings where ²³³U handling and processing most likely occurred [1]. The assignment of buildings in any given year is based on the interpretation of Technical Report information and interviews conducted with personnel involved with or who had knowledge of ²³³U operations at the RFP. It is possible that operations were conducted in additional or fewer years or in other buildings.

YEAR	Building 559	Building 771	Building 774	Building 777	Building 777A	Building 779A	Building 881	Building 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		
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								

TABLE 3 – Likely Locations of Uranium-233 Processing at RFP for 1965 - 1982.

It should be noted that TABLES 1 and 2 list some RFP buildings that are not in TABLE 3. Buildings 371, 707, 750, 776, 779, and 991 reported inventories at various times between 1973 and 1999. While inventories were reported, it does not necessarily mean that material was processed in an MBA and corresponding building. Material (including waste) may have been in storage or in transit when an inventory was being conducted. In addition, Buildings 774, 777A, and 883 are processing buildings identified in TABLE 3 that are not in TABLES 1 or 2. This is most likely due to (1) the difference in the table reporting years, (2) the two unknown MBA's associated with these buildings, or (3) during these various years, MBA titles, areas of coverage, and locations changed.

Excluding the unknown MBA's listed in TABLE 1, all can be separated into distinct four categories: (1) Aqueous (liquid) or Analytical, (2) Storage, and (3) Research and Development (R&D), and (4) Production. The aqueous/analytical MBA's were those where primary activities were conducted using liquids. These areas did not have significant amounts of plutonium metal and oxide, and thus the potential for cross-contamination in PuSPS metal and oxide is very small. Storage MBA's were used to store sealed drums of material or waste, and therefore there is no potential for cross-contamination to occur. R&D areas did not contain large quantities of plutonium metal at any given time when compared to the production areas. There is a small possibility that plutonium metal and oxide contained in these areas could potentially have been contaminated with ^{233}U . The production areas identified were those areas where plutonium processing was stopped when the majority of production operations were transferred to Building 707 during the 1970's. There is a small possibility that any residual plutonium metal from these operations could potentially have been contaminated with ^{233}U . These possibilities are extremely unlikely, however, again based on the segregation summary previously discussed.

When looking at the small possibility that some small ^{233}U cross-contamination of plutonium metal or oxide could have occurred, it is important to note that all activities associated with ^{233}U were stopped in 1982. Plutonium part fabrication operations continued for an additional 7 years, where significant amounts of plutonium were handled, and large quantities of metal were cast, oxide generated, and reprocessed. Even if small quantities of plutonium metal or oxide were cross-contaminated with ^{233}U , the processes associated with plutonium use and recycle would have diluted the amount to one that would be undetectable.

3.4 Summary Calculations

To further support the notion that any ^{233}U in PuSPS oxide and metal is significantly below the 0.5-wt% limit, the following three (3) summary calculations are presented. These calculations are global in nature and do not address individual containers. They are provided to illustrate the gross magnitude of the differences between worst case and known MUF and NOL values.

- (1) The maximum amounts of metal and oxide that can be packaged into a PuSPS container are 4.4 and 5.0 kgs respectively [2]. A 0.5-wt% ^{233}U maximum content would equate to 22 grams of ^{233}U allowed in metal, and 25 grams allowed in oxide. Assuming 700 containers of metal and 1300 containers of oxide are estimated to be produced by the PuSPS, a worst case scenario assumption would indicate that $[(700 \times 22 \text{ grams}) + (1300 \times 25 \text{ grams})] = 47,900$ grams or 47.9 kgs of ^{233}U could be contained in the PuSPS metal and oxide. ^{233}U MUF and NOL inventory values are in the gram range, which indicates that the worst case assumption bounds the 0.5-wt% individual container limit.
- (2) March 3, 1997 inventory data estimates that there is approximately 9,600 kgs of plutonium compounds at the RFP [39]. Assuming that all of this were to be processed through the PuSPS, this would equate to $(9,600 \text{ kgs} \times 0.005) = 48.0 \text{ kgs}$ of ^{233}U (assuming a 0.5-wt% maximum) allowed in the processed oxide and metal. Again, ^{233}U MUF and NOL inventory values were in the gram range, which indicates that the worst case assumption bounds the 0.5-wt% individual container limit.

- (3) Historical plutonium chemistries indicate that non-isotope specific uranium was at a stream level maximum of 550 ppm [1]. 550 ppm = 0.055-wt%, and if it was assumed that all of this was ^{233}U , it is well below the limit of 0.5-wt%.

3.5 ^{233}U Assay Limitations

RFP drum counters, calorimeters, gamma spectroscopy, and other Non-Destructive Assay equipment are set up to look primarily for plutonium isotopes, ^{235}U / ^{238}U , ^{237}Np , and ^{241}Am . ^{233}U is not currently measured. Previous attempts to measure for ^{233}U have been unsuccessful, as several difficulties associated with qualifying and quantifying NDA results were identified when attempts were made to measure ^{233}U [40]. If there were an initiative to investigate determining the actual amount of ^{233}U in stream plutonium, a significant amount of NDA research and development would need to occur.

3.6 ^{233}U Inventory / Material Balance Area Summary

A review of ^{233}U inventory information, Technical Reports, and personnel interviews indicates that ^{233}U was processed in various buildings at the RFP. Changes in processing locations throughout the years can be attributed to the changing mission at the RFP, as well as consolidation of operations into newer and upgraded facilities on plant-site. MBA's reporting ^{233}U inventories were primarily storage areas, aqueous (liquid) areas, or R&D areas, where the amount of oxide generated or metal handled would be none to a limited amount. Inventory records support the fact that ^{233}U products, surplus, and waste materials were segregated and expeditiously shipped offsite. The ^{233}U processing operations were completed in 1982, and metal processing and oxide generation continued for an additional 7 years. Any ^{233}U contained in the plutonium stream would have been significantly diluted over time. Summary calculations show that a significant amount of ^{233}U would need to be present to exceed the two limits.

4.0 CONCLUSIONS

Based on process knowledge, obtained through interviewing personnel familiar with RFP ^{233}U activities, and by reviewing Technical Reports discussing various ^{233}U activities, it appears likely that little ^{233}U material is contained in PuSPS plutonium metal and oxide, and is well below the 0.5-wt% maximum allowable limit.

- (1) A conscious effort to segregate ^{233}U from other processing streams to preclude cross-contamination and avoid the associated gamma radiation concerns is evident.
- (2) The use of ^{233}U as a tracer material in plutonium metal was not conducted.
- (3) Material Balance Areas reporting ^{233}U inventories did not generally have a significant amount of plutonium metal and oxide in process.
- (4) The ^{233}U inventory records indicate that products, surplus, and waste materials were segregated and promptly shipped offsite.
- (5) The handling and processing of ^{233}U ended in 1982, and a significant amount of plutonium metal and oxide was processed through 1989. The ongoing processing of plutonium would dilute any ^{233}U residual contamination to levels that are not detectable.

While it is likely that ^{233}U is present in some incinerator ash, this material type will not be processed through the PuSPS, and therefore will subsequently not end up at the SRS.

5.0 REFERENCES

- 1 RS-090-056, "History of Uranium-233 (²³³U) Processing at the Rocky Flats Plant In Support of the RFETS Acceptable Knowledge Program," April 1, 1999.
- 2 Plutonium Stabilization and Packaging System Campaign Report, Revision 0a, May 7, 1999.
- 3 DOE-STD-3013-99, [DRAFT] "Stabilization, Packaging, and Storage of Plutonium-Bearing Materials," DOE Standard, November 1999.
- 4 DOE/MD-0013, "Acceptance Criteria for Plutonium Bearing Materials to be Dispositioned by Immobilization," Department of Energy, Office of Materials Disposition, October 1, 1998.
- 5 [DRAFT], "Safety Analysis Report – Packages (SARP) 9965, 9968, 9972-9975 Packages (U)," Revision 9, February 2000.
- 6 CD65-3184, "Investigation of Uranium 233-235 Crossover Incident," June 11, 1965, S/RD.
- 7 RFP-2546, Chemistry Research & Development Semiannual Progress Report for the period January – June 1976, Article "Uranium-233 Processing," February 14, 1977, S/RD, (²³³U portion is Unclassified).
- 8 RFP-2817, Chemistry Research & Development Semiannual Progress Report for the period January – July 1978, Article "Uranium-232 Daughter Segregation Study," February 10, 1979, S/RD, (²³³U portion is Unclassified).
- 9 DNFSB/TECH-13, "Uranium-233 Storage Safety at Department of Energy Facilities," Defense Nuclear Facility Safety Board Technical Report, February 1997.
- 10 DOE-STD-XXXX-YR, [DRAFT], "Criteria for Packaging and Storing Uranium-233-Bearing Materials," DOE Standard, Proposed.
- 11 RFP-1848, "Casting of Multiple ²³³U Metal Target Disks," W. V. Conner and D. L. Baaso, May 1972 (U).
- 12 CD73-5096, Non-Pu Physical Inventories, Inventory Differences, and NOL – Operations to Date (information through FY-1988), S/RD.
- 13 Inventory Summary Data on Pu-238 and U-233 for 1957 through April 1977, C/RD.
- 14 RFP-2680, Chemistry Research & Development Semiannual Progress Report for the period January – June 1977, Article "Uranium-233 Processing," October 12, 1977, S/RD, (²³³U portion is Unclassified).

- 15 RFP-2745, Chemistry Research & Development Semiannual Progress Report for the period July – December 1977, Article “Uranium-233 Fabrication Project,” May 29, 1978, S/RD, (²³³U portion is Unclassified).
- 16 CD74-4225, Non-Pu Workbook, FY-74 & FY-75, S/RD.
- 17 CD76-2492, Non-Pu Workbook, FY-76 & FY-76A, S/RD.
- 18 CD77-648, Non-Pu Workbook, FY-77 & FY-78, S/RD.
- 19 CD78-4074, Non-Pu Workbook, FY-79, S/RD.
- 20 CD80-1627, Non-Pu Workbook, FY-80, S/RD.
- 21 CD81-533, Non-Pu Workbook, FY-81, S/RD.
- 22 CD84-1550, Non-Pu Workbook, FY-82, S/RD.
- 23 CD84-1543, Non-Pu Workbook, FY-83, S/RD.
- 24 CD85-118, Non-Pu Workbook, FY-84, S/RD.
- 25 CD85-4954, Non-Pu Workbook, FY-85, S/RD.
- 26 CD86-317, Non-Pu Workbook, FY-86, S/RD.
- 27 CD86-5665, Non-Pu Workbook, FY-87, S/RD.
- 28 CD87-5410, Non-Pu Workbook, FY-88, S/RD.
- 29 CD88-5008, Non-Pu Workbook, FY-89, S/RD.
- 30 CD89-01361, Non-Pu Workbook, FY-90, S/RD.
- 31 RFCD90-02563, Non-Pu Workbook, FY-91, S/RD.
- 32 RFCD91-03823, Non-Pu Workbook, FY-92, S/RD.
- 33 RFCD92-02257, Non-Pu Workbook, FY-93, S/RD.
- 34 RFCD93-01653, Non-Pu Workbook, FY-94, S/RD.
- 35 NMA94-0148, Non-Pu Workbook, FY-95, S/RD.
- 36 NMA95-0167, Non-Pu Workbook, FY-96, S/RD.
- 37 Safeguards Accountability Network (SAN) and RockMAS Printouts (DRAFT WORKSHEETS), 1997-1999, S/RD.

- 38 RS-090-065, "Historical Material Balance Area (MBA) Designator Usage, and Isotope Inventory Review for Uranium 235 (²³⁵U), Uranium 238 (²³⁸U), Plutonium 238 (²³⁸Pu), Neptunium 237 (²³⁷Np), Californium 252 (²⁵²Cf), Curium 244 (²⁴⁴Cm), and Thorium (Th)," August 31, 1999.
- 39 RFP-4790, "Rocky Flats Security Classification Guide," May 1995, UCNI.
- 40 RF/RMRS-018, "RFETS TRU Waste Acceptable Knowledge Supplemental Information," Revision 3, 1/4/99.