HNF-10919
Revision 0

Plutonium Finishing Plant Plutonium- Uranium Oxide

Characterization of Items with <30 Weight Percent Plutonium

Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management

Project Hanford Management Contractor for the
U.S. Department of Energy under Contract DE-AC06-96RL13200

Fluor Hanford
P.O. Box 1000
Richland. Washington

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D.C. Lini and L. H. **Rodgers**
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Glossary

ANSI	American National Standards Institute
ANL	Argonne National Laboratory
AEC	Atomic Energy Commission
BNW	Battelle Northwest Laboratory
B&W	Babcock & Wilcox
BRWs	boiling water reactors
Cat	Category
CSMO	Central Scrap Management Offices
CTL	Chemical Technology Laboratory
COEI	Code of Ending Inventory
DOE	Department of Energy
DU	depleted uranium
EEI	Edison Electric Institute
EBWR	Experimental Boiling Water Reactor
EBR	Experimental Breeder Reactor
ETR	Engineering Test Reactor
EU	enriched uranium
FFTF	Fast Flux Test Facility
FG	fuels grade
HEDL	Hanford Engineering Development Laboratory
HEU	Highly Enriched Uranium
INEL	Idaho National Engineering Laboratory
INEEL	Idaho National Engineering and Environmental Laboratory
LANL	Los Alamos National Laboratory
LMFBR	Liquid Metal Fast Breeder Reactor
LOI	loss-on-ignition
LANMAS	Local Area Network Material Accountability System
MD	Material Disposition
MTR	Materials Testing Reactor
MO	metal oxide
MT	metric tons
MOX	mixed oxide
Nu	natural/normal uranium

NFS	Nuclear Fuel Services
NUMEC	Nuclear Materials and Equipment Corporation
PFPP	Plutonium Fabrication Pilot Plant
PFP	Plutonium Finishing Plant
PNL	Pacific Northwest Laboratory
PNNL	Pacific Northwest National Laboratory
PPSL	Plutonium Process Support Laboratory
PRCF	Plutonium Recycle – Critical Facility
PRF	Plutonium Reclamation Facility
PRTR	Plutonium Recycle Test Reactor
RFP	Rocky Flats Plant
RFETS	Rocky Flats Environmental Technology Site
SEFOR	Southwest Experimental Fast Oxide Reactor
SRS	Savannah River Site
S&C	slag and crucible
SNM	special nuclear materials
US	United States
WIPP	Waste Isolation Pilot Plant
WG	weapons grade
ZPPR	Zero Power Plutonium Reactor

Plutonium Finishing Plant Impure Plutonium Uranium **Oxide** Items **with <30** Weight Percent Plutonium

1.0 Summary

The Plutonium Finishing Plant (PFP) inventory contains primarily plutonium and plutonium-uranium compounds, metals, and alloys that have been stored for a number of years. The inventory in 1994 contained about four metric tons of plutonium and about the same quantity of uranium. In 1994 this inventory was declared to be excess material for national defense programs and is currently slated for disposal. The PFP is stabilizing and packaging these items for disposition. Plutonium metal and high assay plutonium oxide items (plutonium oxide materials containing greater than 85 weight percent plutonium) are being packaged into long-term storage containers for disposition. Low plutonium assay materials (those containing less than 30 weight percent plutonium), such as incinerator ash and sand, slag and crucible (SS&C) residues are being packaged into Waste Isolation Pilot Plant (WIPP) certified containers and transferred to the Hanford Central Waste Complex pending shipment to WIPP.

The disposition strategy for the plutonium-uranium oxide inventory is evolving. This study focuses on the characterization of the plutonium-uranium oxide (also identified as mixed oxide and mixed plutonium-uranium oxide “MOX”) portion that is less than 30 weight percent plutonium. The MOX powder/pellets encapsulated in fuel pins, pellet stacks and assemblies are included in the fuel pins/assemblies category and are not part of this study. The uranium is depleted uranium (DU), natural uranium **(NU)**, or enriched uranium (EU). The EU is enriched up to 97 percent ^{235}U. **A** total of 2,266 PFP items are classified as MOX material. The MOX materials contain about 316 kg of plutonium and about 2 MT of uranium. There are 393 items for which the net weight was not recorded. For the items with recorded net weights there are 1,780 items that contain less then **30** weight percent plutonium which contain about 222 kg of plutonium and about 917 kg of uranium with a total net weight of about 1.4 MT. The average weight percent plutonium **is** about 18.1 percent. The average weight percent plutonium plus uranium is about ***85*** weight percent. This indicates that the material, on the average, contains only trace levels of other metallic and nonmetallic impurities. Of the MOX items without reported net weights, **344** items contain less then 30 weight percent plutonium containing about **40** kg of plutonium and about 1 MT of uranium. The average plutonium content as a function of plutonium plus uranium is about 11.3 weight percent.

The MOX containing items were generated as a result of prior PFP and other nuclear defense program operations. Most of the inventory was received at the PFP for plutonium recovery from other operations conducted at Hanford, i.e. 300 Area, other Department of Energy (DOE) Sites [or its predecessor organizations], and DOE licensed vendors. The current inventory is material that was not reprocessed/recycled when the decision was made to deactivate the PFP.

The materials stored in the PFP vaults meet stringent vault acceptance criteria in terms of stability, reactivity, corrosivity, moisture content, and pressurization. **A** significant fraction of the MOX material is high fired sintered powder and pellets that were

generated during the manufacture of fuel for the recycled plutonium in power reactor projects. Non-nuclear material, other than oxygen, will include small but measurable quantities of flowsheet process chemicals, process equipment and glovebox corrosion products, and materials introduced into the gloveboxes and process lines.

The inventory can be characterized by material composition, original use, process history/origin, originating contractor, and ^{240}Pu and ^{235}U isotopic content. This characterization provides an understanding of the chemical and physical properties that are pertinent in stabilization, packaging and disposition activities. The majority of the MOX-containing materials are legacy materials, i.e. high quality material originally destined for fabrication in fuel components, from reactor fuel development and fabrication activities conducted at the Hanford 300 Area and in particular in the 308 Building in support of the Fast Flux Test Facility (FFTF) Program. **As** a result of the PFP's involvement with the Central Scrap Management Offices (CSMO), smaller quantities of plutonium and uranium containing scrap and legacy materials were received for nuclear material recovery from other **DOE** sites and DOE licensed vendors.

The packaging dates for items in the PFP inventory range from about the late 1970's to the present. Prior to 1975, plutonium was recovered on a routine bases to satisfy programmatic needs. In 1976 the Plutonium Reclamation Facility (PRF) was shut down as a result of an incident in the americium recovery unit. The PRF came back online for a cleanup mission in 1979 and completed that campaign and then shut down. The PRF only ran a few campaigns after 1980. This absence of plutonium scrap recovery capability has resulted in the current vault residue inventory. From the 1980's, any material going into vault storage was required to meet volatility loss-on-ignition limits of less than 1 wt%. Any item currently in the vault, has been restabilized or verified to meet this criterion.

2.0 Introduction

Process residues were routinely processed at the Plutonium Finishing Plant (PFP) to recover plutonium, nuclear materials, and other special nuclear materials (SNM). **A** significant fraction of the MOX material that was received at PFP consisted of high fired sintered powder and pellet residues that were generated during the manufacture of fuel for the recycled plutonium in power reactor projects. **A** second major source of MOX containing residue was recovered from the thermal stabilization of MOX containing polystyrene compacts (polycubes). With the cessation of process operations, a significant quantity of MOX containing materials is still in vault storage.

As an oversimplification, SNM-containing process residues were generated from weapons programs and defense related power reactor programs. The power reactor program materials tended to generate both plutonium and plutonium-uranium process residues. The Hanford 300 Area had a very significant role in the use of recycled plutonium for power reactor fuel development operations and fabrication of fuel assemblies for Hanford's Plutonium Recycle Test Reactor (PRTR) and the Fast Flux Test

Facility (FFTF) as well as a number of offsite reactors. A major portion of the SNM materials received from the Hanford 300 Area is material remaining from these fuel fabrication development and production operations. Plutonium and uranium oxide process residues were also received from United States (US) commercial organizations licensed to fabricate plutonium fuel such as General Electric (GE) Company-Vallecitos, Nuclear Materials and Equipment Corporation (NUMEC), and Nuclear Fuel Services (NFS) and from DOE Non Weapons Complex sites such as Argonne National Laboratory (ANL).

3.0 Plutonium - Uranium Oxide Inventory Description

For ease of understanding, the PFP inventory has been divided into metal/alloy, plutonium oxide, mixed plutonium-uranium oxide (MOX), fuel pins/assemblies, residue, and solutions categories. This study addresses that portion of MOX inventory that is less than 30 weight percent plutonium. The uranium is depleted uranium (DU), natural uranium (NU), or enriched uranium (EU). The EU is enriched up to 97 percent ^{235}U. The MOX powder/pellets encapsulated in fuel pins, pellet stacks and assemblies are included in the fuel pins/assemblies category and are not part of this study. This study is part of a larger effort to characterize the overall PFP inventory in support of the stabilization and disposition efforts at the PFP. ***An*** earlier study, ***Plutonium Finishing Plant Impure Plutonium Oxide Items with >30 and <85 Weight Percent Plutonium, HNF-10638*** addressed that portion of the plutonium oxide inventory containing greater than 30 weight percent plutonium and less than ***85*** weight percent plutonium and the MOX inventory that is greater than 30 weight percent plutonium + uranium. The less than or equal to 30 weight percent special nuclear material **(SNM)** portion of the inventory was addressed in a study titled ***Characterization of Plutonium Oxide Material as Pipe and Go Candidates- Plutonium Finishing Plant Plutonium Oxide Less Than or Equal to 30 weight percent Plutonium, HNF-10775.***

Other than the MOX material recovered from polycubes, the majority of the MOX containing material is MOX sintered powder and pellets that remained when the plutonium recycle program was terminated. Some of the material was recovered from downloading unirradiated excess fuel pins when a project was completed. The material would have been recycled to fabricate fuel pins if the program had continued.

Most of the process operations associated with the fabrication of mixed plutonium oxide-metal oxide fuel involved few chemicals other than the metal oxide(s) that was uranium oxide in the case of MOX fuel. Nonaqueous processes were the norm in the fabrication of MOX fuel and fuel assemblies. Oxide blending was the preferred method for preparing a mixed oxide fuel. The notable exception was the Kerr-McGee operation where coprecipitation of plutonium nitrate and uranium nitrate solutions was employed to produce the MOX they used for fabricating FFTF fuel pins.

The major PFP role in the plutonium recycle program was as a supplier of plutonium nitrate and oxide. When the program was terminated, PFP was designated to provide interim vault storage for the excess MOX materials as the facilities at Pacific Northwest

National Laboratory (PNNL), Hanford Engineering Development Laboratory (HEDL), GE-Vallecitos, Babcock-Wilcox, etc. were deactivated and the plutonium was returned to the DOE until the materials could be processed to recover nuclear materials.

The PFP also provided the plutonium oxide that was used to fabricate the plutonium-uranium oxide containing polycubes. When that program was completed at Battelle Northwest (BNW) Laboratory (now the Pacific Northwest National Laboratory - PNNL) the materials were returned to PFP for the recovery of the nuclear materials for recycle into other DOE nuclear programs. The MOX material in inventory was intended as feed for the PRF at the time the plant was put in a standby mode.

3.1 Plutonium Less Then 30 Weight Percent In Mixed Oxide Material

This study only addresses mixed oxide material that contains less than 30 weight percent plutonium. Plutonium-uranium material has been categorized as SNM, even though most of the uranium in the PFP inventory is either depleted uranium or natural/normal uranium. Natural uranium was used for the fabrication of FFTF fuel and MOX fuel generated at the 308 Building.

This study focuses specifically on the PFP MOX inventory to provide a better understanding of the inventory including details of its manufacture. Previous studies of the <30 weight percent plutonium inventory, HNF-10775, provided few items and few details on the MOX inventory. This report should provide sufficient processing and manufacture details allowing development of the final disposition path for the MOX inventory.

Based on the September ending FY 2001 Local Area Network Material Accountability System (LANMAS) inventory database, 2,226 PFP vault inventory items are identified as plutonium and uranium oxide (mixed oxide, MOX). Material category codes, i.e. Code of Ending Inventory (COEI), American National Standards Institute (ANSI) and PFP Category Codes (Cat Codes), and engineering judgment were the bases for identifying an item as MOX. The details of the method used to generate the MOX items list from LANMAS is provided in Appendix 1. Classification of an item as MOX is not based on a rigid technical or chemical definition but is consistent with the way the Department of Energy (DOE) has categorized nuclear containing materials.

There are about two metric tons of uranium and 316 kg of plutonium in the 2,226 mixed oxide items (see Table 1). The ^{235}U isotopic enrichment in the EU ranges up to 97 weight percent. For items with reported net weights, 1,780 mixed oxide items contain less than 30 weight percent plutonium and contain about 221.6 kg of plutonium and about 916.7 kg of uranium. For the no reported net weight MOX items, 344 mixed oxide items contain less than 30 weight percent plutonium as a fraction of plutonium + uranium containing about 40 kg of plutonium and about 1,040 kg of uranium. Mixed plutonium-uranium oxide powder or pellets encapsulated in fuel pins, pellet stacks and assemblies are not included in this number.

In a number of cases the reported uranium element weight (61 items) or the plutonium element weight (19 items) is zero or no plutonium weight is reported (23 items). The items with no reported plutonium element weight appear to be uranium only items, since historically the PFP has not identified uranium items as a unique residue category. Those items with zero grams of reported plutonium element weight are either items in which the plutonium element weight is below the reportable quantity for plutonium (one gram +/- 0.5g) or, more likely, uranium items that are potentially cross-contaminated with plutonium. In most cases the MOX items with 0 reported uranium element weight are DU items (51 items).

Table 1 Quantities of Plutonium and Uranium in MOX Materials

Description	Items	Pu –kg	DU-kg	NU-kg	EU-kg	Total U-kg
Reported Net Weight						
<30 Wt % Pu	1,780	221.6	456.8	107.5	352.4	916.7
>30 Wt % Pu	93	30.0	36.5	0.5	2.0	39
No Reported Net Weight						
<30 Wt % Pu	344	40	226.8	767	46.2	1,040
>30 Wt % Pu*	49	24.7	0	0	5.6	5.6
Total	2,226	316.3	720.1	875	406.2	2,001.3

'Based upon the Pu+U reported weights. If actual net weight data were available some of these items could shift into the <30 wt. Percent category.

This study focuses on the mixed oxide material that contains less than 30 weight percent plutonium and, for those items with no reported net weight, less then 30 weight plutonium as a function ofplutonium + uranium. The 2,124 items (Table 2) contain about 261 kg of plutonium and about two metric tons of uranium. For the items with net weight, the overall average weight percent plutonium is about 18 percent and the overall average of the plutonium plus uranium is about 85 percent (see Table 2). This indicates that the material is essentially high-grade plutonium-uranium oxide with very little measurable contaminants. The material is dominated by items generated in the 308 Building for the FFTF and Experimental Breeder Reactor (EBR) II driver and experimental fuel manufacture. The plutonium content in driver fuel was nominally about 25 weight percent plutonium as Pu + U with a range from about 22 to 29 weight percent plutonium. About half of the inventory items are Pu-DU that is consistent for fuel used to make FFTF driver fuel. But a large number (661) are Pu-EU. For items without reported net weights, the plutonium as a function of plutonium plus uranium is about 11% but varies widely as a function of uranium enrichment (see Table 2). The 2.7 weight percent Pu in the Pu-NU items with no reported net weight is typical for fuel that was being fabricated by commercial vendors for study in nuclear power reactors. The majority of the enriched uranium in the Pu-EU items with no reported net weight is highly enriched uranium (HEU) with

the ^{235}U enrichment being greater then **20** weight percent. The MOX with no reported net weight is dominated by material from BNW and GE-Vallecitos.

Table 2 Plutonium and Uranium in MOX Materials Containing Less Then 30 Wt% Plutonium

Description	Pu-kg	U-kg	Net Weight-kg	Ave Wt% Pu	Ave Wt % Pu+U	Item Count
Reported Net Weight						
Pu-EU	85.6	352.4	512.2	18.82	85.52	661
Pu-NU	23.4	107.5	150.4	16.71	85.05	162
Pu-DU	112.6	456.8	700.7	18.71	84.02	957
Subtotal	221	916.7	1,363.3	18.08	84.86	1780
Not Reported Net Weight						
Pu-EU	11.4	46.2		19.52*		109
Pu-NU	19.6	767		2.70*		173
Pu-DU	9.0	226.6		11.71*		62
Subtotal	40	1,039.8		11.31*		344
Total	261	1,956.5				2,124

*As a percent of the Pu+U content

3.2 Categorization **of** Plutonium Uranium Oxide Inventory

Items were categorized as mixed oxide based on assignment of the COEI, ANSI and PFP material category codes. Material category codes were assigned to the plutonium-uranium bearing materials based on the chemical and physical properties, original use, process history/origin, originating contractor, and ^{240}Pu and ^{235}U isotopic content. Engineering judgment and institutional knowledge were also invoked on an individual item basis when necessary to complete the category assignment.

Plutonium uranium oxide items having the same material category codes tend to have similar chemical and physical characteristics. The materials within a material category code can vary in the quantity of nuclear material and impurities. This is especially true when the material codes are based on process operations, i.e. PFP Category Code **208,** Plutonium-Uranium oxide weapons grade (WG) that represents material recovered from the processing of polycubes. The recovered plutonium from the polycubes is not weapons grade (WG) but fuels grade (FG) plutonium. It is less true when the material codes are based upon the originating contractor, i.e. PFP Category Code 950, Plutonium/Europium from **324** Building. There is no europium in these items. This is an apparent case of a clerical error in identifying the abbreviation EU as the chemical symbol for europium, Eu. Incidentally, not all the uranium is EU. The items are actually materials resulting from Fast Flux Test Facility (FFTF) fuel production. Most of the mixed oxide inventory was assigned material category codes that signify vendor origin

rather than a process unit operation. Consequently the chemical composition of the non-nuclear constituents in a scrap item could include any flowsheet chemical used in the entire operation rather than those identified with a unit operation. As an example, the non-nuclear constituents in the plutonium-uranium oxide material recovered from the processing of polycubes (PFP category 208) are restricted to a few process chemicals and all items would contain the same impurities, just differing in concentration. Whereas scrap items shipped from BNW would not necessarily contain the same composition, and individual items could contain materials from different process operations. As noted above, most of the MOX-containing material is high quality mixed plutonium-uranium oxide that would have been used to prepare fuel pellets/powder or fuel recovered from unirradiated excess fuel pins if the program had not been terminated.

For items with reported net weight, the largest number of items (about 675) and the largest quantity of material (about 50%) are from process residues generated in the 308 Building for FFTF and Experimental Breeder Reactor (EBR) II fuel experiments (Table 3). The PFP category code for these materials is 950. As indicated by the different COEI and ANSI codes, the MOX items contain different uranium enrichments and were generated at different stages of the fuel fabrication process. Some of the items are in the form of powder and pellets and some of the items (COEI code 771) represent archived samples and standards. Material generated from the processing of MOX containing polycubes (Material Category 208) represents the second largest block of material (about 19% of the items and about 25% of the total material).

For items without reported net weights, PFP Category code 451,303-C Scrap from BNW, comprises the major fraction of the SNM material (about 70%) (Table 4). There are relatively few items because the material is stored in lard cans. Each lard can is considered an item, even though the material is usually packaged in several cans within the lard can. The process residues from GE-Vallecitos (PFP Category Code 475) comprise the next largest fraction of material and about 34% of the number of items

3.3 Source of Plutonium Uranium Oxide Inventory

The material codes provide a linkage to the source that generated the process residue. Using the definition of the PFP category code and an examination of transfer records, the inventory was sorted by PFP category code and assembled by organization/facility that generated the material (Table 5). Materials directly related to FFTF and EBR II fuel pin fabrication and quality testing that were conducted by HEDL in the 308 Building comprise about 68 percent of the items (PFP Category Codes 950 and 955). The next largest component (about 19%) consist of items generated at PFP from the recovery of MOX from polycubes used in criticality studies. A relatively small number of items were generated from other PFP operations. The PFP Category Code 62 could be items that were received and restabilized to meet PFP vault criteria or from stabilization of residues generated at PFP from processing plutonium-uranium materials in PRF.

Another five percent of the MOX-containing material was shipped from the Battelle Northwest Laboratory (BNW, currently Pacific Northwest National Laboratory) and are

PFP Category Codes 451 (303-C Scrap from BNW), 76 (BNW Uranium Scrap WG), 77 (BNW Uranium Scrap FG 04), and **78** (BNW Uranium Scrap FG Other). These items are scrap or legacy material from the BNW operations involving reactor fuel development operations and fabrication of the fuel assemblies for Hanford's Plutonium Recycle Test Reactor (PRTR) and the FFTF as well as a number of offsite test reactors.

Table 3 PFP Plutonium Less Then 30 Wt% in MOX

OEI Code	ANSI Code	Category Code	PFP Category Code Discription	Pu Wt grams	EU Wt grams	DU Wt grams	NU Wt grams	Wt% SNM average	Wt% Pu average	Item Count	Net Weight sum	SNM Wt sum
290	In Fuel Element and Target Fabrication Process			82,101.0	47,425.0	143,034.0	38,584.0	85.71	21.83	421	353,999.4	311,144.0
		950	Plutonium/Europium Scrap from 324 Building*	1,973.0		9,360.0	6.0	87.32	18.62	12	12,822.7	11,339.0
		955	Fuel Pins									
453	In Oxide Conversion Process			228.0		513.0		73.18	22.52	1	1,012.6	741.0
			No PFP Category Code									
725	Compounds (Unirradiated)											
	C01	Oxides (Low-Fired)- Oxides (Relatively Pure)		213.0			828.0	85.54	17.50	1	1,217.0	1,041.0
		43	Analytical Laboratories Scrap with Uranium FG	246.0			784.0	90.37	21.58	1	1,139.8	1,030.0
		62	Stabilized Scrap					87.47	9.90	3	1,204.3	1,052.0
	C04	Oxide with Zirconium/Oxides (Impure)		136.0	916.0							
		950	Plutonium/Europium Scrap from 324 Building*					84.34	11.41	340	347,118.5	295,705.0
	C40	Plutonium-DU Compounds		37,788.0		257,917.0		79.92	18.34	2	3,400.0	2,657.0
		208	Plutonium-Uranium Oxide WG	617.0		2,040.0		58.79	6.88	5	2,456.0	1,502.0
		449	Argonne Scrap CZA	165.0		1,337.0		87.28	7.94	2	3,378.0	2,948.0
		451	303-C Scrap from BNW	268.0		2,680.0		67.86	11.42	4	2,889.2	2,444.0
		478	Plutonium-NU-DEPL Uranium Oxide FG	389.0		2,055.0						
		62	Stabilized Scrap					106.58	26.07	3	997.0	986.0
	C41	Plutonium-DU Oxides (Low-Fired)		269.0		717.0		85.27	16.54	2	3,415.0	2,886.0
		208	Plutonium-Uranium Oxide WG	566.0		2,320.0		59.25	7.33	9	3,762.0	2,359.0
		449	Argonne Scrap CZA	259.0		2,100.0						
		451	303-C Scrap from BNW					4.00	4.00	26	40,145.0	1,098.0
	C43	Plutonium-DU Oxides (High-Fired)		1,098.0		0.0		88.37	17.65	2	4,963.0	4,386.0
		404	Plutonium-Uranium in Mixed Oxide Gneo	876.0		3,510.0		76.72	2.08	6	7,480.4	5,197.0
		449	Argonne Scrap CZA	201.0		4,996.0		87.97	24.92	37	18,764.5	16,518.0
		62	Stabilized Scrap	4,746.0		11,772.0						
		950	Plutonium/Europium Scrap from 324 Building					88.05	3.44	1	10,842.0	9,546.0
	C50	Plutonium-NU Compounds		373.0			9,173.0	31.70	6.34	1	489.0	155.0
		111	BNW Items for Storage	31.0			124.0	88.04	3.44	1	9,744.0	8,579.0
		224	Plutonium-Enriched Uranium Oxide FG	335.0			8,244.0	82.46	18.77	17	15,179.0	12,797.0
		451	303-C Scrap from BNW	2,977.0			9,820.0					
		77	BNW Uranium Scrap FG-04					70.71	12.31	1	2,404.0	1,700.0
	C52	Plutonium-NU Oxides (High-Fired)		296.0			1,404.0	97.36	21.69	1	1,051.0	708.0
		426	Plutonium-NU or DEPL Uraniummoly Alloy FG	228.0			480.0	96.63	2.16	5	7,579.2	7,271.0
		43	Analytical Laboratories Scrap with Uranium FG	162.0			7,109.0	70.98	8.80	1	193.0	137.0
		475	Plutonium Powder Pellets-NU YEB	17.0			120.0	86.34	18.33	38	32,046.7	27,876.0
		478	Plutonium-NU-DEPL Uranium Oxide FG	6,450.0			21,426.0					
		950	Plutonium/Europium Scrap from 324 Building*									

Table 3 (Continued) PFP Plutonium Less Then 30 Wt% in MOX

COE	ANSI	Category	PFP Category Code Discription	Pu Wt grams	EU Wt grams	DU Wt grams	NU Wt grams	Wt% SNM average	Wt% Pu average	Item Count	Net Weight sum	SNM Wt sum
	C70	Plutonium-Enriched Uranium Compounds						88.00	18.58	2	290.0	255.0
		224	Plutonium-Enriched Uranium Oxide FG	53.0	202.0			85.01	2.23	47	86,825.0	73,664.0
		404	Plutonium-Uranium in Mixed Oxide Gneo	1,929.0	71,735.0			78.57	11.84	1	574.0	451.0
		42	Analytical Laboratories Scrap with Uranium wG	68.0	383.0			84.62	26.89	1	1,339.0	1,133.0
		425	Plutonium-ENR Uranium Alloy FG	360.0	773.0			86.53	17.80	19	21,257.0	18,279.0
		436	Plutonium-ENR Uranium Oxide Powder Pellets FG	3,896.0	11,068.0	3,315.0		50.90	5.41	4	2,360.0	1,201.0
		439	Plutonium-Uranium-Zirconium Oxide FG	122.0	1,079.0			83.79	20.55	1	253.0	212.0
		459	Plutonium-ENR Uranium Oxide FG	52.0	160.0			8.65	8.65	3	1,384.0	103.0
		466	Plutonium-ENR Uranium FG 04	103.0	0.0			31.62	5.14	1	778.0	246.0
		476	Mixed Samples	40.0	206.0			47.60	19.53	5	3,660.0	1,892.0
		492	Plutonium-ENR Uranium Oxide FG	703.0	1,189.0			86.02	20.30	39	11,940.8	10,189.0
		77	BNW Uranium Scrap FG-04	2,550.0	7,639.0			151.11	15.78	13	8,577.0	5,707.0
		78	BNW Uranium Scrap FG Other	888.0	4,819.0							
	C72	Plutonium-Enriched Uranium (High-Fired)						87.59	20.76	342	292,159.3	255,920.0
		950	Plutonium/Europium Scrap from 324 Building*	59,753.0	196,167.0							
	C82	Plutonium-Thorium Oxide-Impure						69.64	0.00	5	3,549.3	2,606.0
			PFP Category Code		2,606.0							
727	Noncombustibiles (unirradiated)											
	E70	Plutonium -Uranium Solutions						11.44	10.59	1	236.0	27.0
		425	Plutonium-ENR Uranium Alloy FG	25.0	2.0			9.71	3.94	1	381.1	37.0
		522	PuO2 EU Contaminated WG	15.0	22.0							
746	In Other R&D Usage							81.31	25.57	87	2,769.8	1,404.0
		950	Plutonium/Europium Scrap from 324 Building*	400.0	41.0	963.0						
771	Samples and Standards							86.67	20.00	1	15.0	13.0
		50	Radiation Measurement Laboraory Scrap FG	3.0	10.0			89.14	23.14	264	35,288.9	31,173.0
		950	Plutonium/Europium Scrap from 324 Building*	7,636.0	5,943.0	8,193.0	9,401.0	74.48	13.88	1,780	1,363,329.5	1,138,314.0
		Total		221,604.0	352,385.0	456,822.0	107,503.0					

* There is no europium in these items

Table 4 PFP Plutonium MOX With 0 Grams Reported Net Weight

COEI Code	ANSI Code	PFP CAT Code	PFP Category Code Discription	Pu Net Weight grams	Net Weight DU grams	Net Weight EU grams	Net Weight NU grams	Sum Pu+U grams	Wt% Pu Ave.	Item Count
637	Sintered Products									
		451	303-C Scrap from BNW			2,777.00		2,777.00	0	1
725	Compounds (unirradiated)									
	C40	Plutonium-DU Compounds						31,909.00	14.78	30
		208	Plutonium-Uranium Oxide-WG	4,692.00	27,217.00			1,381.00	21.41	2
		35	Oxide from Polystyrene	272.00	1,109.00			5,996.00	20.41	6
		56	CTL Scrap with U FG	521.00	5,475.00			149.00	15.44	1
		77	BNW Uranium Scrap FG 04	23.00		126.00				
	C41	Plutonium-DU Oxides- Low Fired						161,706.00	1.09	9
		451	303-C Scrap from BNW	1,748.00	159,958.00			21,903.00	2.67	9
		85	Plutonium/DU/NU Fuel Rods	584.00	21,319.00					
	C43	Plutonium-DU Halides						4,587.00	18.88	3
		416	Plutonium-ENR Uranium Oxide FG	842.00	3,745.00			197.00	13.20	1
		453	Plutonium-ENR Uranium-Sn FG	26.00	171.00			6,157.00	1.67	1
		77	BNW Uranium Scrap FG 04	103.00	6,054.00					
	C50	Plutonium-NU Compounds					8,603.00	8,734.00	1.50	1
		527	Pu02-UO2 Fuel Pins	131.00			8,814.00	9,624.00	11.57	6
		77	BNW Uranium Scrap FG 04	810.00			3,628.00	3,652.00	0.66	1
		98	Repacakged Plutonium	24.00						
	C51	Plutonium-NU Oxides (low-fired)					590,664.00	605,687.00	2.78	44
		451	303-C Scrap from BNW	15,023.00			4,531.00	4,625.00	2.03	2
		85	Plutonium/DU/NU Fuel Rods	94.00						
	C52	Plutonium-NU Oxides (high-fired)					1,637.00	1,686.00	2.91	1
		456	Plutonium/NU Standards YEB	49.00			149,130.00	152,572.00	2.25	118
		475	Plutonium Powder Pellets YEB	3,442.00						
	C70	Plutonium-Enriched Uranium Compounds						1,149.00	21.38	9
		224	Plutonium-Enriched Uranium Oxzide FG	242.00		907.00		4,959.00	16.00	12
		416	Plutonium-ENR Uranium Oxide FG	763.00	1,560.00	2,636.00		1,713.00	20.39	7
		417	Plutonium-ENR Uranium Oxide CR FG	350.00		1,363.00		1,805.00	10.40	7
		436	Plutonium-ENR Uranium Oxide Powder-Pellets FG	189.00		1,616.00		40.00	20.00	1
		441	Plutonium-ENR Uranium Oxide -Grinder Filt FG	8.00		32.00		869.00	13.65	5
		452	Plutonium-ENR Uranium Oxide -Sic FG	77.00		792.00		1,115.00	18.16	4
		453	Plutonium-ENR Uranium-Sn FG	190.00		925.00		439.00	24.66	3
		459	Plutonium-ENR Uranium Oxide FG	109.00		330.00		13,172.00	20.92	18
		492	Plutonium-ENR Uranium Oxide FG	2,776.00		10,396.00		134.00	0.00	1
		56	CTL Scrap with Uranium FG	0.00		134.00		29,326.00	23.06	32
		77	BNW Uranium Scrap FG 04	6,590.00		22,736.00		1,606.00	17.39	5
		78	BNW Uranium Scrap FG Other	281.00		1,325.00				

Table 4 (Continued) PFP Plutonium MOX With 0 Grams Reported Net Weight

COEI Code	ANSI Code	PFP CAT Code	PFP Category Code Discription	Pu Net Weight grams	Net Weight DU grams	Net Weight EU grams	Net Weight NU grams	Sum Pu+U grams	Wt% Pu Ave.	Item Count
727	Noncombustibles (unirradiated)									
	E70	Plutonium and Enriched Uranium Combustibles						53.00	7.55	1
		523	PuO2 EU Contaminated FG	4.00		49.00				
729	Process Residues (unirradiated)									
	G70	Pu-EU Residues						9.00	22.22	1
		477	Plutonium-ENR Uranium Grinding Wheel Res FG	2.00		7.00				
746	In Other R&D Usage							44.00		1
		111	BNW for Storage			44.00				
771	Samples and Standards							10.00		1
		111	BNW for Storage			10.00				
			Total	39,965.00	226,608.00	46,205.00	767,007.00	1,079,785.00	12.30	344

* Items do not contain europium

Table **5** Origin of Plutonium-Uranium Oxide Residues

COEI code	ANSI code	Category code	Item count
HEDL Process Operations in 308 Building			
290		950	421
290		955	12
725	C04	950	3
725	C43	950	37
725	C52	950	38
725	C72	950	342
746		950	87
771		950	264
		subtotal	1,204
BNW Process Operations in 308 Building			
725	C50	77	17
725	C70	77	39
725	C50	111	1
725	C70	78	13
725	C40	451	5
725	C41	451	9
725	C50	451	1
637		451	1
725	C40	77	1
725		451	9
725		85	9
725	C43	77	1
771		111	1
746		111	1
725	C50	77	6
725	C51	451	44
725	C70	77	32
725	C70	78	5
		subtotal	195
PFP Polycube Stabilization			
725	C40	208	340
725	C41	208	3
725	C40	208	30
725	C40	35	2
		subtotal	375
PFP Analytical Laboratory Operations			
725	C52	42	1
725	C01	43	1
725	C52	43	1
725	C40	56	6
725	C70	56	1
		subtotal	10

Table **5** Continued Origin of Plutonium-Uranium Oxide Residues

	COEI code	ANSI code	Category code	Item count	
PFP Stabilization Operations					
	725	C01	62	1	
	725	C40	62	4	
	725	C43	62	6	
			subtotal	11	
General Electric Process Operations					
	725	C50	224	1	
	725	C70	459	1	
	725	C70	224	2	
	725	C52	475	5	
	725	C52	456	1	
	725	C52	475	118	
	725	C70	224	9	
	725	C70	459	3	
			subtotal	140	
Nuclear Fuel Services Process Operations					
	725	C43	404	26	
	725	C70	404	47	
			subtotal	73	
Nuclear Materials Equipment Corporation Process Operations					
	725	C70	436	19	
	725	C70	439	4	
	725	C70	436	7	
			subtotal	30	
Babock-Wilcox Process Operaitons					
	725	C70	492	5	
	725	C70	492	18	
			subtotal	23	
Argonne Laboratories Process Operations Chicago and INEL					
	725	C40	449	2	
	725	C41	449	2	
	725	C43	449	2	
	725	C40	478	2	
	725	C52	426	1	
	725	C52	478	1	
	725	C70	425	1	
	727	E70	425	1	
			subtotal	12	

Table 5 Origin of Plutonium-Uranium Oxide Residues
3 of 3 pages

	COEI code	ANSI code	Category code	Item count	
Battelle Memorial Laboratory Process Operations - Columbus					
	725	C70	476	1	
			subtotal	1	
Miscellaneous Process Operations					
	453			1	
	725		466	3	
	725	C82		5	
	727	E70	522	1	
	771		50	1	
	725	C43	416	3	
	725	C43	453	1	
	725	C50	527	1	
	725	C50	98	1	
	725	C51	85	2	
	725	C70	416	12	
	725	C70	417	7	
	725	C70	441	1	
	725	C70	452	5	
	725	C70	453	4	
	727	E70	523	1	
	729	G70	477	1	
			subtotal	50	
			Total	2,124	

The remaining MOX containing materials were generated at a number of facilities that supported the use of plutonium as MOX fuel for test power reactors. Notable is the material received from commercial vendors that had licenses to fabricate/ process plutonium-uranium fuel including; General Electric-Vallecitos, Nuclear Fuel Services (NFS), Nuclear Material Equipment Corporation (NUMEC), Babcock-Wilcox, and Battelle Columbus Laboratory. The largest single lot is the Pu-NU material from GE-Vallecitos.

The items listed under Miscellaneous Process Operations represent material that has not yet been linked to a process operation or the vendor that generated the material.

3.4 Chemical Characterization of MOX Residues

The MOX residues in this study are plutonium-uranium oxide, which have met the PFP vault acceptance criteria. The material was generated from two primary sources; MOX prepared in support of the recycled plutonium program, and MOX recovered from the processing of polycubes. The process operations used in the fuel fabrication process did not add appreciable chemical constituents above the trace level. The different potential impurities that can be in the MOX material are summarized in Table 6. The scrap materials tend to be high quality plutonium-uranium oxides. The feed specifications for the ceramic grade uranium dioxide and ceramic grade plutonium dioxide used in the FFTF fuel manufacture is shown in Tables 7 and 8, respectively. The total maximum impurity limit is about 0.4 weight percent with no single element being greater than 400 to 500 parts per million. The impurity specification for plutonium oxide for MOX is about 0.6 weight percent. This is slightly higher than the PFP plutonium oxide specification. This specification places limits on more nonmetal elements and identifies specifications on more transition and rare earth metal elements. The materials with lower concentration of plutonium and uranium oxides would be expected to also contain metal oxides used in preparing ternary oxides such as zirconium or molybdenum and material that was swept up from the floors and other surfaces of the gloveboxes. After pyrolysis and combustion, the MOX recovered from polycubes would contain measurable quantities of aluminum from the paint that had been used to coat the cubes and residual carbon. The metal oxides from the process equipment, gloveboxes, and hood sweeps would also be expected in the lower nuclear material content items. A summary discussion of the processes used at the PFP and the other facilities that are the sources of the plutonium oxide and MOX material is provided in Section 3.5

Table *6* Potential Chemical Impurities in MOX

Material Source	Process Operations **and/or** Chemicals	Potential Impurities
PFP	Polycubes & Thermal Stabilization	C, Al, Ta, Mo
MOX Powder	Die lubricants, Zirconium oxide, Thorium oxide, Aluminum, Stainless Steel, Molybdenum oxide	C, Zr, Th, Mo, Fe, Al, Cr, Ni
MOX Pellets	Die lubricants, Grinding Media,	Al, Mo, C

3.4.1 Chemical Composition of MOX Generated in Support of FFTF

PFP Category Codes 950 and 955 are comprised of MOX containing material shipped to PFP from the 324 Building. The material was packaged at the 308 Building for storage in the 324 Building. This represents materials that were generated by HEDL in support of the recycled plutonium program. The HEDL was responsible for several operations in the 308 Building. They performed quality assurance on the FFTF fuel pins that were used to fabricate the driver fuel assemblies used in Cores 1, 2, 3, and 4 for irradiation at FFTF. Kerr McGee at Crescent, Oklahoma, and NUMEC and Babcock and Wilcox at Parks Township, Pennsylvania fabricated the driver fuel pins. More than 60,000 driver fuel pins were fabricated. About one out of every 100 driver fuel pins was cut open in the 308 Building and the pellets were tested to ensure compliance with the specifications. Some of the fuel pellets were retained as samples and the remaining pellets were packaged for recycle. Standards were also fabricated for calibration of the various testing equipment. The HEDL also fabricated test fuel pins and assemblies for testing at the FFTF at Hanford the EBR II reactor at the Idaho National Engineering Laboratory (INEL, now the Idaho National Engineering and Environmental Laboratory). Over 8,000 FFTF test pins and EBR II MOX test fuel pins were fabricated at the 308 Building.

The PFP received items categorized as PFP category code 950 or 955 (see Table 5). The ANSI codes indicate that the uranium component is NU, DU, or EU. The COEI codes categorize the materials as 290, In Fuel Elements and Target Fabrication Process, 725 Compounds (Unirradiated), 746 In Other R&D Usage and 771, Samples and Standards. Correspondence related to the transfer of the material to PFP described the materials as "oxide/pellets Pu- NU/DU/EU high fired", "oxides/pellets Pu-NU/DU/EU", "recycle powder Pu-DU", and standard powder Pu-DU"[Brooksher 1985] These materials tend to be high quality fuel grade plutonium uranium oxide. Test pins were fabricated that included zirconium, molybdenum, and thorium in combination with MOX or with uranium oxide which would account for some of the low nuclear material content. Some of the material could be materials gathered during the cleanout and shutdown of the plants and material returned for recycling from the vendors fabricating the fuel pins. However there were very few additional process chemicals introduced into the MOX fuel fabrication process.

Table 7 Uranium Dioxide Impurities Standards 1994

Impurity Group	Element	Maximum Impurity Limit µg/g
1	Na K	SUM = 700 (No one > 500)
2	Cl F	SUM = 30
3	C N	SUM = 350 (No one > 250)
4	Sm Eu Gd Dy	SUM = 100
5	Fe Cr Ni	SUM = 1000 (No one > 400)
6	P S	SUM = 350 (No one > 300)
7	Zn Pb Sn	SUM = 220 (No one > 100)
	Cd	20
8	Be	20
	Ca Al	SUM = 450 (No one > 250)
	Si	150
9	B	20
10	Li	10
11	Co	10
	Ti V Ta W Cu Ag Mo Mn Zr	SUM = 900 (No one > 400)

Table 8 Impurity Limits for Plutonium Dioxide for the Fabrication of Mixed Oxide Fuel Nuclear Standards 1981

Element	Maximum Impurity Limit. µg/g PuO_2
Aluminum	250
Beryllium	20
Boron	10
Cadmium	20
Calcium	500
Carbon	200
Chlorine	50
Cobalt	20
Fluorine	25
Lithium	10
Magnesium	100
Nitride Nitrogen	200
Phosphorus	200
Potassium	200
Silicon	150
Sodium	300
Sulfur	300
Tantalum	400
Tungsten	100
Vanadium	200
Zirconium	500
copper + Zinc + Titanium	650
silver + Manganese + Molybdenum + Lead + Tin	200
Samarium + Europium + Gadolinium + Dysprosium	100
Chromium + Iron + Nickel (No one > 500)	1200

The 308 Building received material and scrap from the vendors during the cleanout and shutdown of their plants at the close the FFTF fuel fabrications contracts. Material from the cleanout of the B&W plant at the close of the FFTF Core 3 and 4 fuel fabrication contract was shipped to the Savannah River Plant. The "scrap collected by B&W during the course of the FFTF contract for the purpose of recycling to the fabrication program" was included in the material shipped to the PFP. [Brooksher 1985a] The material was described as MOX powder and pellets. The PFP Scrap Evaluation Team evaluated and approved the material with respect to stability, packaging and capability for receipt and storage. [Washburn 1985]

3.4.2 Chemical Composition of MOX Generated by BNW

Before the contract was awarded to HEDL, the BNW conducted research and development activities in the 308 Building on the use of recycled plutonium in test power reactors. In addition to the initial work on FFTF, BNW prepared test fuels for the PRTR, and test reactors located at Idaho. The initial test fuels were metal alloys. But very quickly the choice of fuel became plutonium oxide –uranium oxide. Before the advent of sintered fuel pellets, much of the early experience was obtained with packed-particle fuels prepared by a swaging process or by vibrational compaction. Based on PFP category codes and material item names, BNW packaged and sent MOX materials to PFP (Table *5)*. Reactor quality plutonium oxide and uranium oxide powders were blended and loaded into fuel pins. Very few process chemicals, other then the metal oxides being blended into fuel were required. **Any** lubricants or binders that were used were thermally removed before the powders were loaded into the fuel tubes. Scrap material that would have been recovered from cleanout and maintenance operations could contain metals from the equipment/ tools used in the blending, grinding, loading, cutting operations. The metals would have been converted to the metal oxides during thermal stabilization of the scrap for packaging and shipment to PFP. However, the majority of the material would be excess MOX feed material that did not meet the stringent fuel specifications, and material recovered from downloading unirradiated fuel pins that were packaged and stored for recycling. In almost all fuel pin fabrication operations excess fuel pins were fabricated in case a fuel pin is damaged or fails quality assurance testing or is archived.

In the case of the "no net weight" MOX items, there is about one metric ton of Pu - NU/DU powder that was recovered from the down loading of Vipac-loaded mixed oxide zircalloy-clad fuel pins (Table 4, PFP Category Code 451). The material can be segregated into three homogeneous lots; a DUO_2 –0.9 Wt% PuO_2 Vipac mixed oxide unmoderated powder lot, a NUO_2 –2 Wt% PuO_2 Vipac mixed oxide unmoderated powder lot, and a DUO_2 -4 Wt% PuO_2 Vipac mixed oxide unmoderated powder lot. Notes on the transfer of BNW material from the 303-C building to PFP described three lots of material. One lot consisted of 150 4-¼ inch by 4-7/8 inch cans of NuO_2 (439 kg) 2 Wt % PuO_2 (8.8 kg, 24 Wt % ^{240}Pu) Vipac mixed oxide unmoderated powder. The second lot consisted of 56 4-1/2 inch by 4-7/8 inch cans of NUO_2 (156 kg) 0.9 Wt % PuO_2 (1.6 kg, 7.36 Wt % ^{240}Pu) Vipac mixed oxide unmoderated powder. The third lot consisted of 55 4-¼ inch by 4-7/8 inch cans of DUO_2 (439 kg) 2 Wt % PuO_2 (8.8 kg, 24 Wt % ^{240}Pu)

Vipac mixed oxide unmoderated powder. There should be about 261 inner cans packaged in 53 lard cans based upon this accounting.

The material was recovered from the Plutonium Recycle – Critical Facility (PRCF) rod cutting and dumping operations carried out during 1978. [Laming 1983] The Zircalloy-clad PRCF (36" long) Vipac loaded mixed oxide rods were transferred from the 303-C Building to the 308 Building. The Vipac powder was removed, canned and returned to the 303-C building. The operation was carried out in three separate campaigns, corresponding to the three separate PuO_2 enrichments of the fuel. Table 9 summarizes the downloading operation and quantities and isotopics of the nuclear material. Figures 1 and 2 provide the fuel specifications for the UO_2- 0.9 Wt% PuO_2 and the UO_2 – **2** Wt% PuO_2 fuel rods.

These materials were shipped to PFP during 1983 from the 303-C Building as part of the larger transfer of nuclear material from Pacific Northwest Laboratory **(PNL)** to Rockwell Hanford. [SD-CP-TI-0131The 303- C Shipping and Receiving Plan indicates that the individual inner cans were mechanically sealed and placed into a mechanically sealed overpack and shipped inside a DOT-6M or AL-M6 shipping container. The items were placed into lard can storage initially in 224-T. When 224-T was deactivated, the material was transferred to PFP. [Copies of the shipping and receiving reports providing net weight, plutonium and uranium weights, and percent ^{240}Pu for each can are on file with the PFP Technical Support Department, and **NDA** data on the items are on file at PNNL Safeguards and Security Services].

The materials are high quality plutonium-uranium oxide. Since the recovered material was intended for use in reactor experiments, the MOX would have met the specifications for reactor fuel. Chemical impurities are at the measurable trace level and there are no volatile, reactive, or corrosive constituents. The fuel downloading campaigns in the 308 Building would have not introduced any measurable impurities other than traces of zircalloy from the cutting operation. The materials have subsequently been in vault storage in mechanically sealed cans since 1979.

3.4.3 Chemical Composition of MOX Generated By Other MOX Vendors

The majority of the remaining MOX containing materials were shipped to the PFP from several DOE laboratories and licensed plutonium vendors. All of these shippers were involved with the several programs that were conducted in the U.S. during the 1950's through the 1970's to investigate the use of recycle MOX in power reactors [DOE/DP 1996, Cowell 1997]. The shipper/source identity was determined based on a Combination of the PFP category code description and the item material name. Items from General Electric-Vallecitos, Nuclear Fuel Services (NFS) and Nuclear Material and Equipment Corporation (NUMEC) (Table 10) make up the majority of the materials.

Table 9 Summary of Downloading Vipac Fuel Pins
Laming 1983

Dates of Campaign	Nominal Description of Material Involved	Number of Rods to 308	Number of Cans to 303-C	Approx Total Net Wt., Kg	Approx. Total U Wt., Kg U
3/16/78 –11/20/78	2% PuO_2-UO_2	453	150	510	**440** (N)
11/28/78-1/10/79	0.9% PuO_2-UO_2	163	55	180	157 (D)
8123179- 9/28/79	4% PuO_2-UO_2	168	56	182	155 (N)

Summary of Pu Content and Isotopic Information

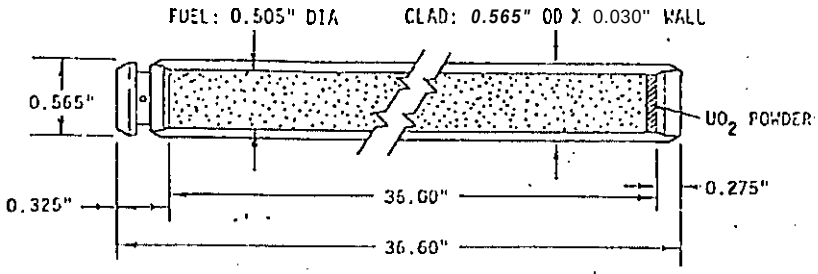
Nominal Material	Pu Factor Used in Transfer	Pu Factor From Analytical Measurement Ave. 3 Samples	Pu Isotopics Used in Transfer		Pu Isotopics from Analytical	
			Pu-240	Pu 239+241	Pu 240	Pu 239+241
2% PuO_2-UO_2	0.007867	0.009033	7.26	92.72	7.36	92.61
0.9% PuO_2-UO_2	0.017265	0.01710	23.84	75.49	24.08	75.19
4% PuO_2-UO_2	0.03444	NA	18.42	80.12	18.61	79.94

Figure 1 Fuel Description Pu - **NU** Vipac Fuel Pin

FUEL SPECIFICATIONS: UO_2 - 2 WT% PuO_2

FUEL RODS

1. ROD DIMENSIONS



2. CLADDING: ZIRCALOY-2 TUBING WITH PLUGS SEAL WELDED AT BOTH ENDS.
3. TOTAL WEIGHT OF LOADED FUEL RODS: 1340 gms (AVERAGE)

FUEL LOADINGS

1. PuO_2 MIXED IN NATURAL UO_2 AND VIBRATIONALLY COMPACTED.
2. 1128 gms OF UO_2-PuO_2 MIX/ROD.
3. CHEMICAL COMPOSITION WT%: Pu/PuO_2 = 88.1 U/UO_2 = 88.0 Pu/MIX = 1.760; 1.617 1.765
4. PuO_2 IS 2.00 WT% OF TOTAL MIXTURE.
5. FUEL DENSITY - 9.54 gm/cc (~87% THEORETICAL DENSITY).
6. UO_2 POWDER AT THE END OF FUEL COLUMN.
7. THE ISOTOPIC DISTRIBUTION OF PLUTONIUM IN THE TWO TYPES OF RODS REFERRED TO AS 8% AND 24% IS GIVEN BELOW:

1. 8% (NOMINAL) ^{240}Pu ATOM PERCENT		2. 24% (NOMINAL) ^{240}Pu ATOM PERCENT	
91.615	^{239}Pu (1.13d)?	71.762	^{239}Pu
7.654	^{240}Pu (7.67)?	23.503	^{240}Pu
0.701	^{241}Pu	4.08	^{241}Pu
0.031	^{242}Pu	0.656	^{242}Pu

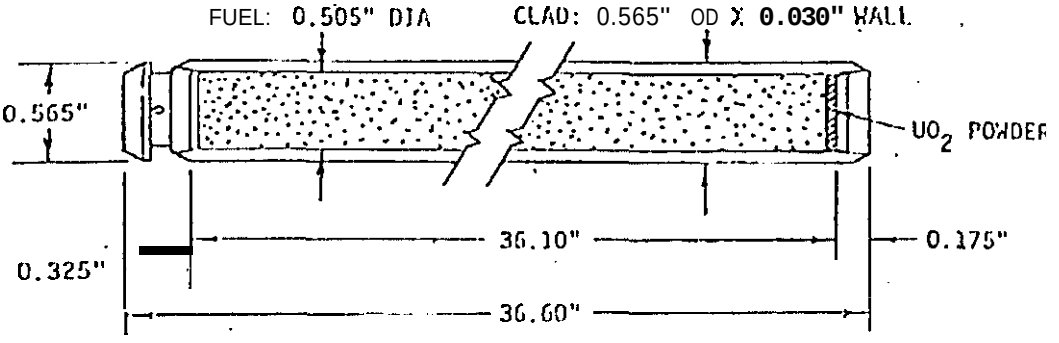
8. ANALYSIS DATE: JANUARY 1965 , MAY 1965
9. SEPARATIONS DATE: APRIL 1962 , OCTOBER 1953
10. ^{241}Am CONTENT: NOT KNOWN , 4.89 X 10^{-6} nuclei/bn-cm
11. DATE MEASURED: NOT MEASURED , MARCH 1970

Figure 2 Fuel Description of Pu - DU Vipac Fuel Pin

FUEL SPECIFICATIONS: UO_2 - 0.3 WT% PuO_2

FUEL RODS

1. ROD DIMENSIONS



2. CLADDING: ZIRCALOY-2 TUBING WITH PLUGS SEAL WELDED AT BOTH ENDS.
3. TOTAL WEIGHT OF LOADED FUEL RODS - 1316 gms (AVERAGE).

FUEL LOADINGS

1. PuO_2 MIXED IN DEPLETED UO_2 AND VIBRATIONALLY COMPACTED.
2. 1107 gm OF UO_2-PuO_2 MIX/ROD.
3. CHEMICAL COMPOSITION WT %: Pu/PuO_2 = 88.2 U/UO_2 = 88.0 Pu/MIX = 0.766.
4. PuO_2 IS 0.068 WT% OF TOTAL MIXTURE.
5. FUEL DENSITY - 9.38 gm/cc (~86% THEORETICAL DENSITY).
6. UO_2 POWDER AT THE END OF FUEL COLUMN.
7. ISOTOPIC COMPOSITION.

PLUTONIUM - ATOM%		URANIUM - XTON %	
92,133 ± 0.050	^{239}Pu	0.005 ± 0.001	^{234}U
7.241 ± 0.055	^{240}Pu	0.234 ± 0.002	^{235}U
0.595 ± 0.005	^{241}Pu	0.006 ± 0.0005	^{236}U
0.025 ± 0.001	^{242}Pu	99.755 ± 0.002	^{238}U

8. ANALYSIS DATE: NOVEMBER 1964
9. SEPARATIONS DATE: UNKNOWN

Table 10 MOX Items From Minor Sources

PFP Cat or ANZI Codes	Item Count	Shipper/Source
404	73	Nuclear Fuel Services
436, 439	30	NUMEC
224, 475, 459	140	General Electric
423, 425, 426, 449, 478	12	Argonne-Chicago & Idaho
492	23	B&W
476	1	Battelle-Columbus
	52	Miscellaneous
Total	331	

Between 1965 and 1972, uranium and plutonium oxides were dry blended and pressed to fabricated MOX fuel pellets at the NFS fuel fabrication facilities at Erwin, TN. Uranium and plutonium were also coprecipitated, dried, and fired to produce MOX. The fuel pellets were loaded into rods and end caps were welded in place. The material appears to be high quality MOX. The Pu-EU items contain an average of about 87 weight percent plutonium + enriched uranium. The DU element weights are reported as 0 for a number of items, but the material contains about the same Pu content as the Pu-EU items. If the depleted uranium weight had been recorded it is highly probable that the material could be shown to be high quality MOX.

In addition to fabricating fuel pins for FFTF from Hanford-supplied plutonium oxide, NUMEC had an ongoing plutonium-uranium fuel fabrication program and prepared fuel for other test programs. Some of the MOX fuel tested in the Saxton Plutonium Project was prepared by NUMEC. The NUMEC also conducted research on the preparation of MOX from coprecipitation of plutonium and uranium nitrate solutions [Caldwell 19631. Kilogram size batches of sinterable-grade PuO_2-UO_2-ZrO_2 ceramic powders were also prepared as test fuel at NUMEC [Fisher 1966]. The plutonium-uranium content of the 73 items ranges from about 85 weight percent to less then 50 weight percent. The lower concentration items could be from cleanout of gloveboxes and equipment or material containing a third metal oxide.

General Electric operated the Vallecitos Atomic Laboratory at Pleasanton CA. The GE-Vallecitos laboratory was engaged with the Edison Electric Institute (EEI) to evaluate the utilization of plutonium in boiling water reactors (BWRs). [Walke 1971] Fuel fabricated by GE-Vallecitos was tested in the Big Rock Point and the Quad Cities 1 Reactors. The Government supplied the plutonium. [Nixon 19841 The material shipped to the PFP appears to be material from the termination of their fuel fabrication activities. The Pu-EU and Pu-NU (PFP Cat Code 475) items tend to contain greater than 85 weight percent Pu + U. A few Pu-NU item (PFP Cat Code 224) contain less then 50 weight percent weight percent Pu + NU and could be material recovered from the cleanout of gloveboxes and equipment.

In the case of the "no net weight" MOX items, 118 Pu + **NU** items shipped from the GE-Vallecitos facility appear to be similar to the five Pu-NU items with net weight. The same weight is used for both the **NU** value and the net weight value in three of the items. In the two that have correct net weights, the percent Pu + **NU** is 88 Wt% and the plutonium in **SNM** percentage is 2.13 Wt% and 2.23 Wt%. In the corresponding "no net weight" items, the plutonium in **SNM** percentage ranges from 2.01 Wt% to 2.94 wt%. This suggests that the material is primarily mixed plutonium oxide- uranium oxide that was excess pellets and powder from fuel pin fabrication with only trace quantities of metal impurities. ***An*** internal Rockwell Hanford Operations letter [Black 1980] that indicated that of the eight FL-10 shipping containers [in this specific shipment] received from GE Vallecitos, the containers in seven were to be buried and only one was to be opened and the containers put in vault storage. The contents were reported to be plutonium, and plutonium-uranium mixed pellets and powder.
A fuel pin recently shipped to PFP from GE-Vallecitos contained six archived MOX fuel pellets designated Quad Cities 001. The MOX pellets were made from the batch feed material used in the fabrication of fuel rods for the Quad Cities reactor. The plutonium concentration in the pellets was 3.91 Wt%, 4.02 Wt%, 2.9 Wt%, 2.68 Wt%, 4.02 Wt%, and 3.9 Wt% with a ^{240}Pu plutonium isotopic content of either 12.2 Wt% or 18.67 Wt%. The fuel rod was made in the early 1970's. [Murray 19991

The Argonne National Laboratory at Argonne, IL (Argonne-East) and at Idaho (Argonne-West) sent plutonium-containing scrap to the PFP. In addition to MOX, plutonium oxide, metals and alloys have been received. Some of the materials were determined to be unprocessable at the PFP and either sent to Savannah River for processing or disposed of as transuranic waste (TRU). The MOX items have a plutonium plus uranium content of from 69 to 88 weight percent indicating they have high quality nuclear material oxides. Correspondence from Argonne- East describes MOX shipped to the PFP as PuO_2/UO_2 and pellets, pieces, and powders. Some items are contaminated with stainless steel fines and chips. Passivated Zero Power Plutonium Reactor (ZPPR) material has also been received. This is a plutonium, uranium, and molybdenum alloy that was converted to the oxide by burning in a tantalum crucible. The records are insufficient to link the remaining inventory items with specific shipping documents, but the documentation does indicate the types ofmaterials that were being transferred to the PFP. [Skiba 1983, Nelson 1983, Skiba 1983a]

Information is unavailable on the remaining items. The Battelle Columbus Laboratory had been involved with a number of the recycled plutonium utilization programs such as the Saxton Plutonium Project and the Electric Power Research Institute research into the use of MOX. Numerous plutonium-containing materials were received at the PFP for plutonium recovery. Likewise Babcock and Wilcox (B&W) was another licensed vendor involved in the fabrication of MOX fuels. They took over the NUMEC operation in 1971. The nuclear material content of these materials is about 48 weight percent suggesting that this is sweeps recovered from the cleaning out of gloveboxes and process equipment. The expected non-nuclear constituents would likely be the same as those found in any MOX fuel fabrication process.

3.4.4 Chemical Composition of The MOX Recovered from Polycubes

The PFP Category Code 208 represents nuclear material recovered from the pyrolysis and burning of polycubes (Table 5). The polycubes were Polystyrene- Plutonium-Uranium fuel compacts that were fabricated to measure plutonium criticality in various conditions and arrays in the Critical Mass Laboratory. Plutonium and uranium oxides were uniformly dispersed in polystyrene cubes. The components used in the fabrication of the compacts would have met very stringent material quality controls. The polycubes were typically clad with approximately 6-mil thick tape and spray painted with about a 1-mil thick coat of aluminum paint. The pyrolysis/combustion operation should have only introduced trace quantities of corrosion products from the furnace equipment and boats. **All** moisture and volatile components were driven-off during the high temperature oxidation phase. Vault records indicate that these materials were generated in 1984-1985. Shipping and Receiving reports of 1983 indicate that PNNL shipped to PFP some PuO_2- depleted UO_2 polycubes. The material was 8.1 weight percent PuO_2 in PuO_2-UO_2 with a plutonium isotopic of 11.5 Wt% ^{240}Pu. The Pu-DU oxide in PFP Category Code 208 has the same ^{240}Pu isotopic as the material received from PNNL in 1983. If this is similar material, the expected weight percent plutonium oxide would be about 8 %. The plutonium weight percent values for the PFP inventory items indicates that most of the items are essentially pure plutonium-uranium oxide. Some of the items have a plutonium oxide content greater than 8 weight percent which is not readily explained, while those less than 8 weight percent plutonium oxide suggest the presence of aluminum, corrosion products and residual carbon.

Some of the plutonium-uranium oxide recovered from polycubes was sent to LANL as part of the Materials Identification Surveillance Program. One item that contained 16 weight percent plutonium and 69 weight percent uranium was analyzed LANL. The non-nuclear components, other than oxygen, were at the trace level, i.e. 0.036 Wt % carbon, 0.030 Wt % chromium, and 0.043 Wt% nickel. Photographs of the material indicated that the material is a free flowing powder.

3.4.5 Chemical Composition of Other MOX Generated Materials at PFP

Based on the assignment of material category codes, MOX containing residues were generated at PFP either in the Analytical Laboratory and Plutonium Process Support Laboratory (PFP Category Codes 42, 43, and 56) or by PFP stabilization processes (PFP Category Code 62) (See Table 5). Since the PFP did not routinely process plutonium-uranium items, the materials are not of PFP origin, but are combination of samples received by the Laboratory from polycube burning and PRF or from offsite for characterization and materials that were restabilized at the PFP to meet vault acceptance criteria. The PFP Category Code 62 material could also be material recovered from the PRF gloveboxes after a plutonium-uranium scrap recovery campaign. Little can be deduced about the chemical composition based on the material category codes. The non-nuclear components would be a cross section of those materials found in the PFP Plant and Laboratory operations and introduced in MOX fuel fabrication processes. There should be nothing unique about the chemical composition of these items.

3.5 Process Operations Related to Plutonium Uranium Oxide Generated Materials

The MOX-containing material at PFP is from two primary sources. The PFP recovered plutonium-depleted uranium oxide from the pyrolysis and burning of MOX containing polystyrene compacts. The majority of the remaining materials are from programs that supported the use ofrecycled plutonium as a MOX fuel for test power reactors.

The MOX fuel was fabricated either from plutonium-uranium oxide powder or plutonium-uranium oxide sintered pellets. Much of the early development and testing using plutonium-uranium oxide powders was conducted at Hanford in the 308 Building in the 300 Area. The same processes were generally used by the different facilities to prepare plutonium-uranium oxide pellets for fuel and the fabrication of fuel pins. The one notable exception being whether the fuel fabrication facility started with plutonium and uranium oxide or with plutonium nitrate and uranium nitrate. Plutonium-uranium oxide sintered pellets were produced in the 308 Building, as was also the quality assurance testing of the MOX fuel and driver FFTF fuel pins. The MOX containing materials generated at facilities producing MOX pellets are expected to contain essentially the same non-nuclear chemical constituents.

3.5.1 Description of Polycube Recovery Operations

The PFP Category Code 208 and ANSI code C40 items were generated from the burning of plutonium-depleted uranium polystyrene compacts (polycubes) that were manufactured to measure plutonium criticality in various conditions and arrays in the Critical Mass Laboratory. The plutonium and uranium oxides were uniformly dispersed in polystyrene cubes. The polycubes were typically clad with approximately 6-mil thick tape and spray painted with about a I-mil thick coat of aluminum paint. The C40/208 items contain Pu + DU generated in the 1984-1985-time period and except for eight items that appear to have been generated near the conclusion of the burning campaign are high assay plutonium-uranium oxides.

The items were processed at PFP to recover the nuclear material. Glove Box 4 in the PFP housed the equipment used in distilling polystyrene blocks (cubes). The process recovered the nuclear material by pyrolysis and distillation of polystyrene and burning of the residual carbon. The cube coatings were split to allow the escape of distilling gases and the coatings were burned rather than mechanically removed. The still temperature was maintained at 450^0C and alarmed at 490^0C. The off-gas from the still was drawn through a scrubber containing carbon tetrachloride. No more than 8-2" x 2" x 2" cubes were placed in the still portion of the Glove Box 4 at any time. The 2" x 2" x 2" cubes were broken in half and placed in the boats. The boats were charged to the still on a 45 minute cycle.

The boats were then transferred to the furnace portion of Glove Box 4. Each distilled cube was crushed and evenly distributed into two boats. The boats were charged to the burning furnace. The burning furnace was maintained at 810^0C and alarmed at 950^0C.

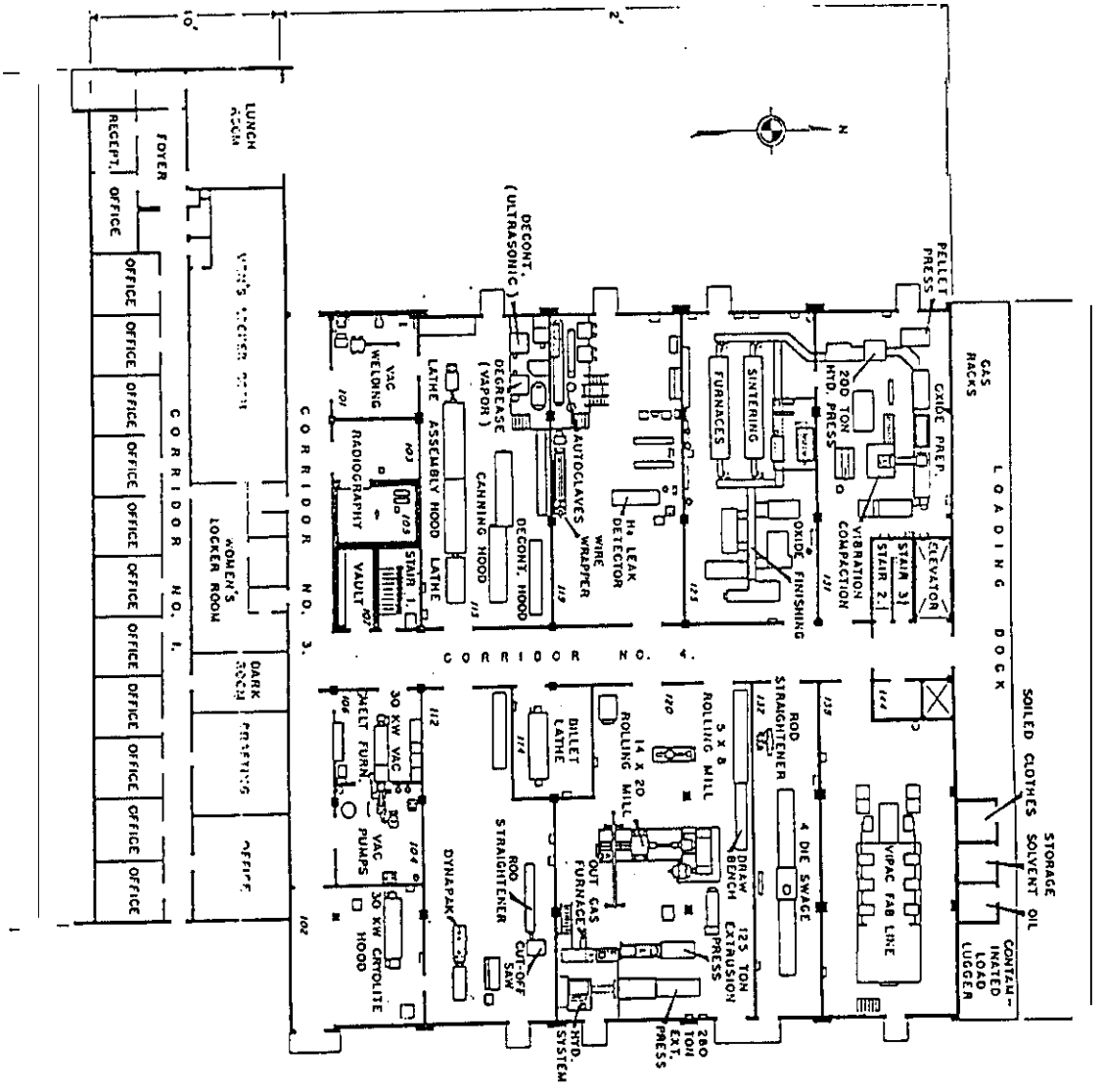
3.5.2 MOX from Fuel Fabrication Processes

Several programs were conducted in the U.S. between the late 1950's and 1970's to investigate the use of recycled plutonium as a MOX fuel for power reactors. Hanford had a very significant role in this work. In late 1956 the Atomic Energy Commission (AEC) initiated a program of research and development at the Hanford Laboratories (in the 300 Area). The prime goal of the program, termed the Plutonium Recycle Program, was the development of safe and economical methods for the recycle of plutonium bearing fuels, with major emphasis on thermal-heterogeneous reactors. The General Electric Company initially conducted the work. The work transitioned first to BNW in 1965 and in 1970 to the HEDL. Work conducted in the 300 Area played a very significant role in reactor fuel development operations and fabrication of fuel assemblies for Hanford's PRTR and the FFTF, as well as a number of offsite reactors. Related work on plutonium fuels was also initiated at the Argonne National Laboratory primarily devoted to fast reactor technology, and at the Los Alamos Scientific Laboratory (now called LANL) focusing on other plutonium fueled reactor systems. Nine commercial organizations had licenses to fabricate/ process fuel rods - Atomics International, Babcock and Wilcox, Exxon Nuclear, GE, Gulf United Nuclear, Kerr-McGee, NSF, NLJMEC and Westinghouse. Exxon Nuclear, GE and Westinghouse were major players. Kerr-McGee, NLJMEC, and Babcock and Wilcox (B&W) manufactured driver fuel pins for FFTF. The MOX fuel fabricated by the commercial vendors tended to be MOX ceramic pellets.

3.5.2.1 Process Operations to Fabricate MOX Powder Containing Fuel Pins

Hanford became involved in the Plutonium Recycle Program in the early 1950's. Most of the fuel development and fabrication work at Hanford was conducted in the **308** Building which was constructed in 1960. The building was initially named the Plutonium Fabrication Pilot Plant (PFPP) and later was called the Fuels Development Laboratory. Figure *3* shows the layout of the PFPP as it was initially configured. The initial work at Hanford focused on metallic core fuel. The use of aluminum-plutonium core alloy offered the surest way of fabricating plutonium-bearing fuel elements. But very quickly the choice of fuel became plutonium oxide – uranium oxide. Before the advent of sintered fuel pellets, much of the early experience was obtained with packed-particle fuels prepared by a swaging process or by a vibrational compaction process in which mixtures of high-density fuel particles were packed into metal cladding tubes by mechanical reducing of the tube diameter or by electronic or pneumatic vibration of the tubes. The cold swaging process was used in fabricating fuel for PRTR. Hanford initiated experiments in high-energy vibrational compaction in 1959. Figure **4** is a depiction of the PRTR fuel element history. It shows the evolution from plutonium-aluminum alloy fuels to MOX fuel and the transition from swagged loaded fuel pins to Vipac loaded fuel pins.

Figure 3 Plan of Plutonium Pilot Plant
(Merker 1963)



The practice used to the greatest extent to obtain the MOX powders was mechanical blending of the mixed oxides. In the mechanical blending process, individually prepared oxides of uranium and plutonium were intimately mixed together in some predetermined proportion. Mechanical techniques that were used for the mixing include ball milling (wet and dry), paste blending (wet), conical shell blending (dry), mulling (dry to damp), and V-blending with an internal intensifier bar. The operations involved in the preparation of plutonium oxide and uranium oxide feeds are shown in Figure 5.

Hanford prepared powders by a high energy-rate pneumatic impaction process (Nupac). In the high-energy pneumatic impaction process the mechanically blended mixed-oxide powders were heated under vacuum to 1,200^{0}C and compacted in a die at 200,000-500,000-psi pressure by the kinetic energy of a massive pneumatically accelerated **ram.** [Sharp 19641The fuel was recovered from the container by crushing, and additional crushing and screening was performed to prepare the optimum particle mixtures for vibrational compaction or swaging.[Brite 19641These operations are depicted in Figure ***6*** which shows an overall PRTR flow chart.

Hanford initially developed cold and hot swaging-compaction processes. The process consisted of swaging a physical mixture of UO_2-PuO_2 powder into a 0.5 inch diameter, eight-foot long, Zircalloy-clad rod. The cladding tubes for swage compaction have larger diameters and thicker wall and are shorter in length than the final rod. The welded fuel pins were loaded into a rotating feed device and passed through the swage. Three reducing passes plus several finishing passes were used to obtain a final length with a length elongation from 63 to 90 inches. Reduction in area was 42 percent. [Bardsley 19621 Figure **7** depicts the swage compaction fabrication process. Other than the mixed oxides very few chemicals are introduced in the process. Hot swaging process provided densities more than 10 percent greater than those obtainable by cold swaging by heating the fuel rods to approximately 800^{0}C as they entered the swaging machine. The process was phased out in the favor of vibrational compaction. Experiments with high-energy vibrators were initiated at Hanford in 1959.

The feed material for vibrational compaction consisted of crushed and sized arc-fused UO_2 and high temperature calcined PuO_2. The process is essentially the same as that used in the swage compaction process except for the compact method (see Figure 8). The difference between the two processes begins at the tube loading step. Again, there are very few process chemicals other than feed associated with this process. The cladding tube, with the first end cap welded in place, was firmly attached in **a** vertical position to either an electromagnetic or pneumatic vibrator. The open end of the tube was attached by a bag to the bottom of a glove box. The sized particle fractions were dried and then sequentially loaded into the tube as small proportional increments to give a uniform longitudinal distribution of thin layers. A follower rod with a weight of a few pounds

Figure **4** PRTR Fuel Element History

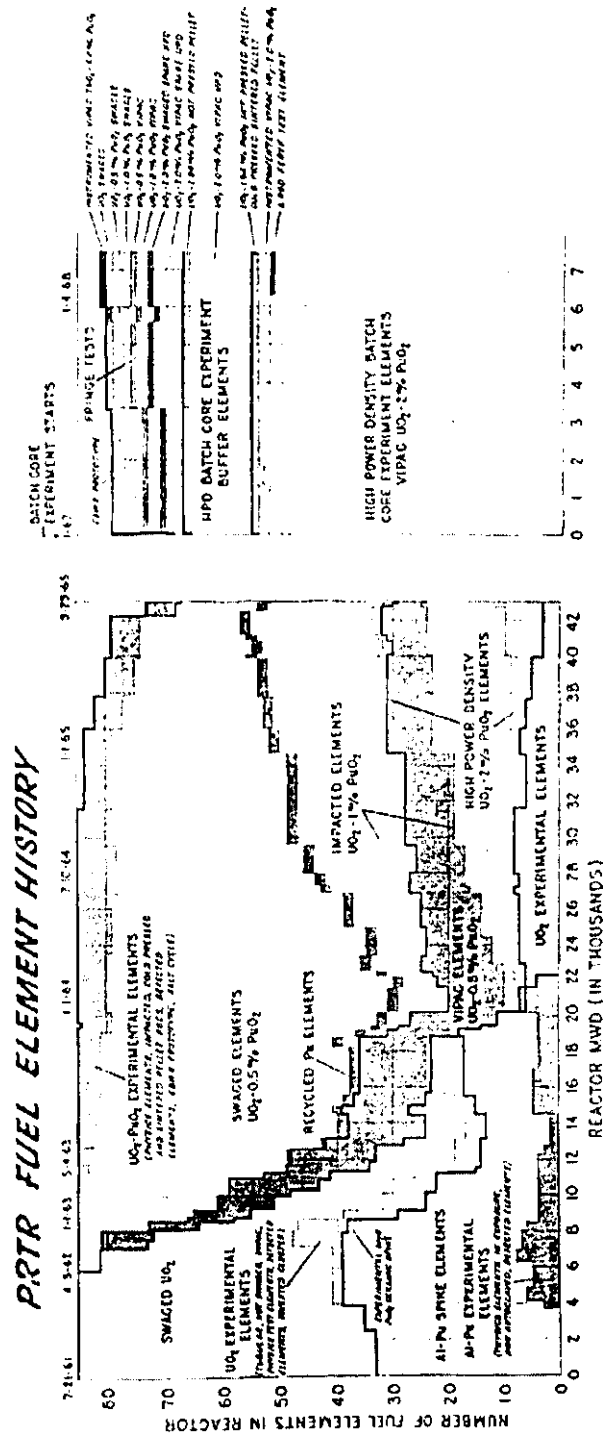


Figure 5 Characterization of Fuel Materials
(Laming 1969)

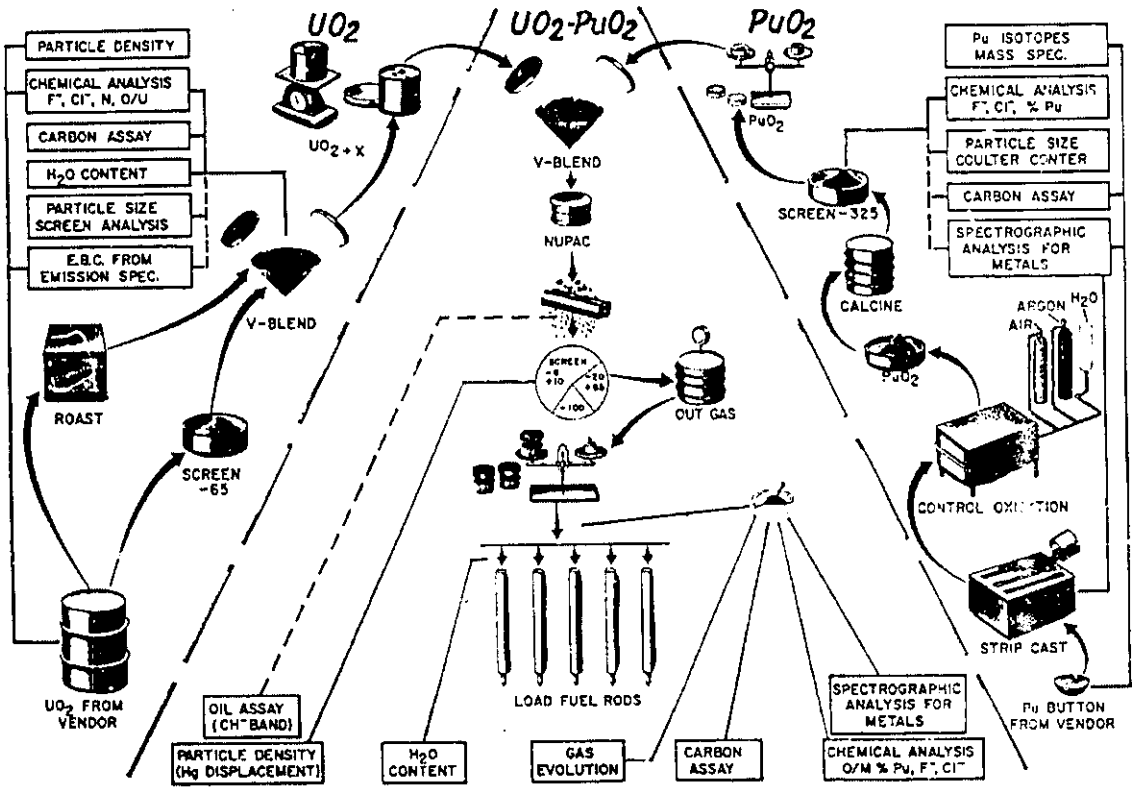


Figure *6* PRTR Flow Chart
(Brite 1973)

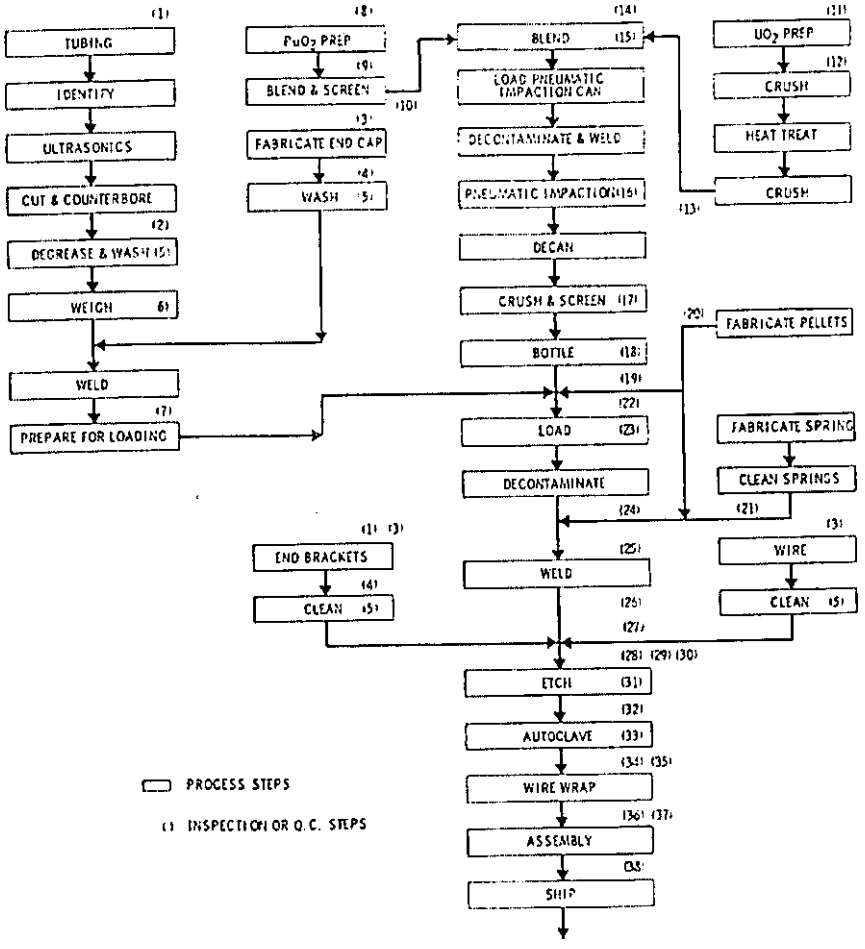


Figure 7 Mark I-M $UO_2 - PuO_2$ **Swage Compaction Fabrication Process (Thomas** 1967)

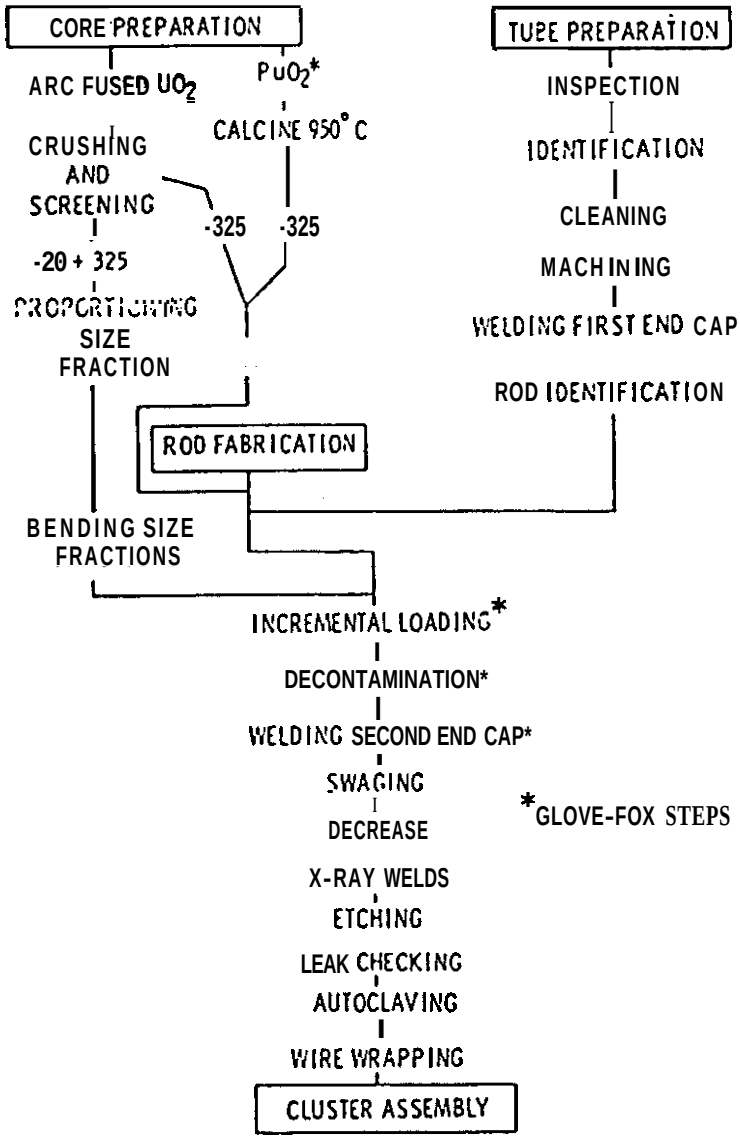


Fig. 19.14—Mark I-M UO_2–PuO_2 swage compaction fabrication process. From R. E. Bardsley and R. E. Sharp, *Trans. Am. Nucl. Soc.***, 5 (2): 453 (1962).**

Figure 8 **Mark** I-L UO_2 - PuO_2 Vibratory Compaction Fabrication Process (Thomas **1967)**

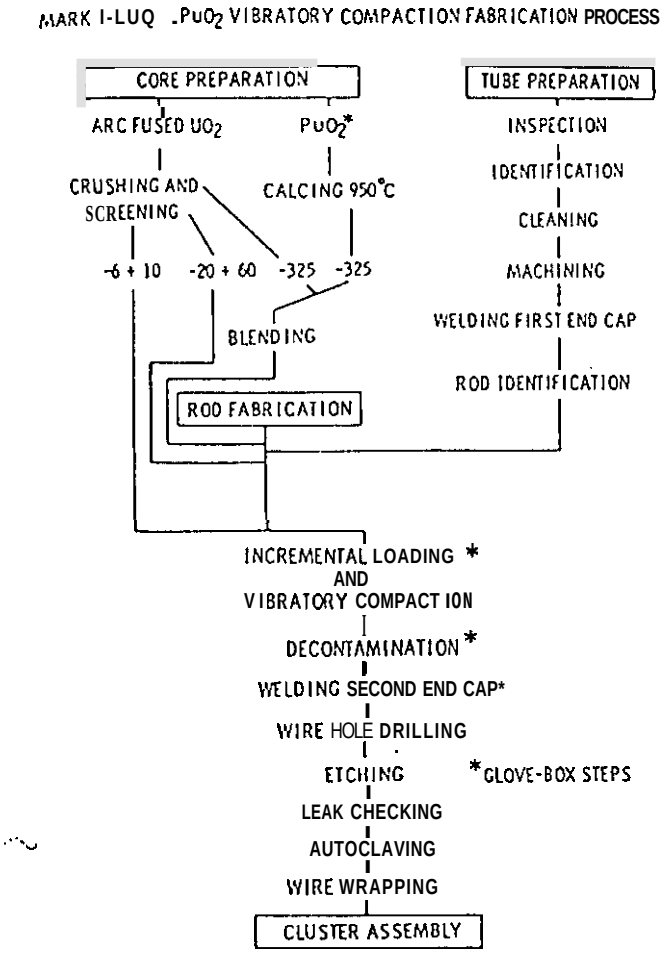


Fig. 19.12—Mark I-L UO_2–PuO_2 vibratory compaction fabrication process. From R. **K.** Koler and R. E. Sharp, *Trans. Am. Nucl. Soc.*, **5** *(2)*: **454 (1962).**

was placed on the particle column and the tube was vibrated. This procedure was repeated until the tube was full and the particles compacted. Vibrational compaction of the fuel rods was achieved using an electrodynamic system with a rating of 1750 lb force. **A** novel resonant plate coupling between the rod and the vibrator produced high compaction rates of seven to eight minutes per rod. [Hauth 19611

Irradiation tests on Vipac fuels were conducted in the PRTR at Hanford and the Materials Testing Reactor (MTR), the Engineering Test Reactor (ETR), and the Experimental Boiling Water Reactor (EBWR) at the ANL in Idaho. The primary objective of the EBWR was to demonstrate successfully the use of plutonium in a boiling-water reactor and to obtain fuels and physics data on PuO_2 - UO_2- fuel. [Dawson 19671

3.5.2.2 Process Operations to Prepare MOX Pellets for the Fabrication of Fuel Pins

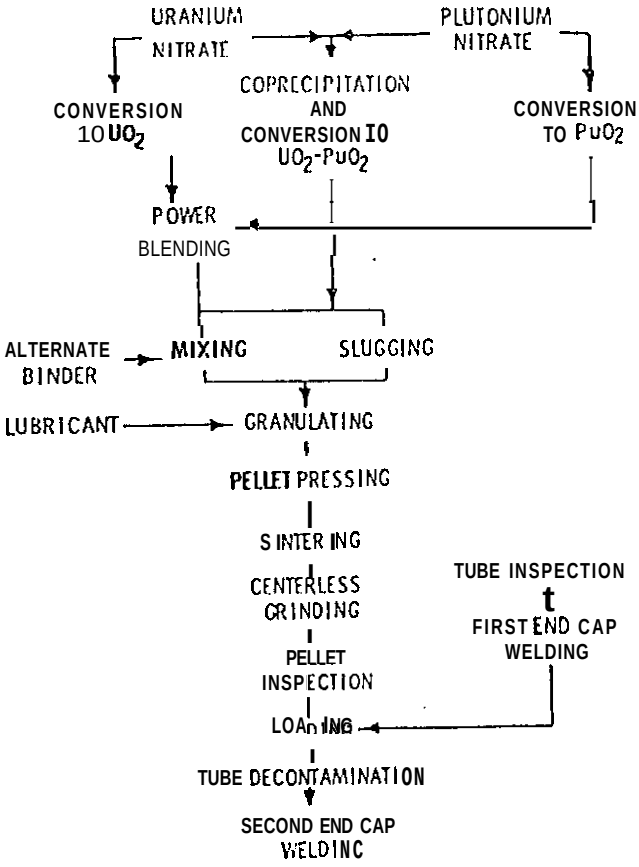
The greatest volume of mixed-oxide fuel used was in the form of pellets prepared by pressing and sintering of well-blended plutonium oxide and uranium oxide [Thomas 1967]. The only significant difference among the various manufacturers of MOX fuel was the starting material in their facility. In the majority of operations plutonium oxide and uranium oxide was used. A minor number of vendors started with plutonium nitrate - uranium nitrate and coprecipitated, with additions of ammonia solution, plutonium hydroxide and an ammonium diuranate. The coprecipitated material was converted to a sinterable grade oxide powder by thermal treatment under a suitable atmosphere. A generic flow diagram of the pelleted fuel process is depicted in Figure 9.

In either case the powders for pelleting had to be free flowing mixtures for pellet pressing in automatic presses. This was achieved by either a wet process, wherein an organic binder was added by blending a solution containing the binder with the powder mixture, evaporating the solvent, and granulating the dried mixture through a screen, or a dry process, in which the powder mixture was prepressed into slugs, at a pressure somewhat less than the ultimate pressure to be used in the final pressing and granulated. A die lubricant was usually blended with the granulated powder prior to final pellet pressing, to reduce die wall friction and thus facilitate ejection of the pressed pellet. Alternatively, the die wall could be lubricated prior to die filling by spraying or wiping with a lubricant solution.

The ultimate goal in the powder compaction process was to continuously and automatically fabricate a fuel shape by mechanical compression of a powder that possessed suitable characteristics. Coprecipitated and mechanically blended powders of MOX were generally compacted at 10 to 30 $ton/in.^2$ to give a resultant green shape with a density of 40 to 55% theoretical.

High temperature sintering at 1,500 to 1,700' C was used to convert mixed-oxide pressed powder pellets into dense, solid ceramic bodies suitable for reactor fuel. During the sintering process, volatile impurities, adsorbed gases, organic binders, or die lubricants were driven off and interdiffusion of PuO_2 and U02 occurred, resulting in a more

Figure 9 Process Outline - Pelleted Fuel
(Thomas 1967)



homogeneous plutonium distribution than existed in the powder mixtures. In general, the system can be described as one of complete solid solution between the component oxides Different types of sintering furnaces were used for sintering mixed-oxide pellets. The batch-type furnaces include molybdenum-wound alumina furnaces, refractory metal foil insulated furnaces using refractory metal wire mesh or rod-type heating elements, and graphite resistance-heated furnaces using a nonporous ceramic tube to contain the mixed-oxide pellets in a carbon-free reducing atmosphere. Continuous furnaces used for the sintering of mixed-oxide pellets have been almost exclusively the pusher type utilizing a flat alumina hearth, molybdenum elements, and molybdenum pusher plates and trays.

Two types of centerless grinders that have been used for obtaining close control of pellet diameter were belt centerless grinders and abrasive wheel grinders. Machines of both types ordinarily required a recirculating water spray for cooling and lubrication during grinding.

The hot pressing of mixed-oxide fuel pellets provided an alternate fabrication process affording the advantage of forming the pellets to the exact diameter required, obviating the pellet grinding operation.

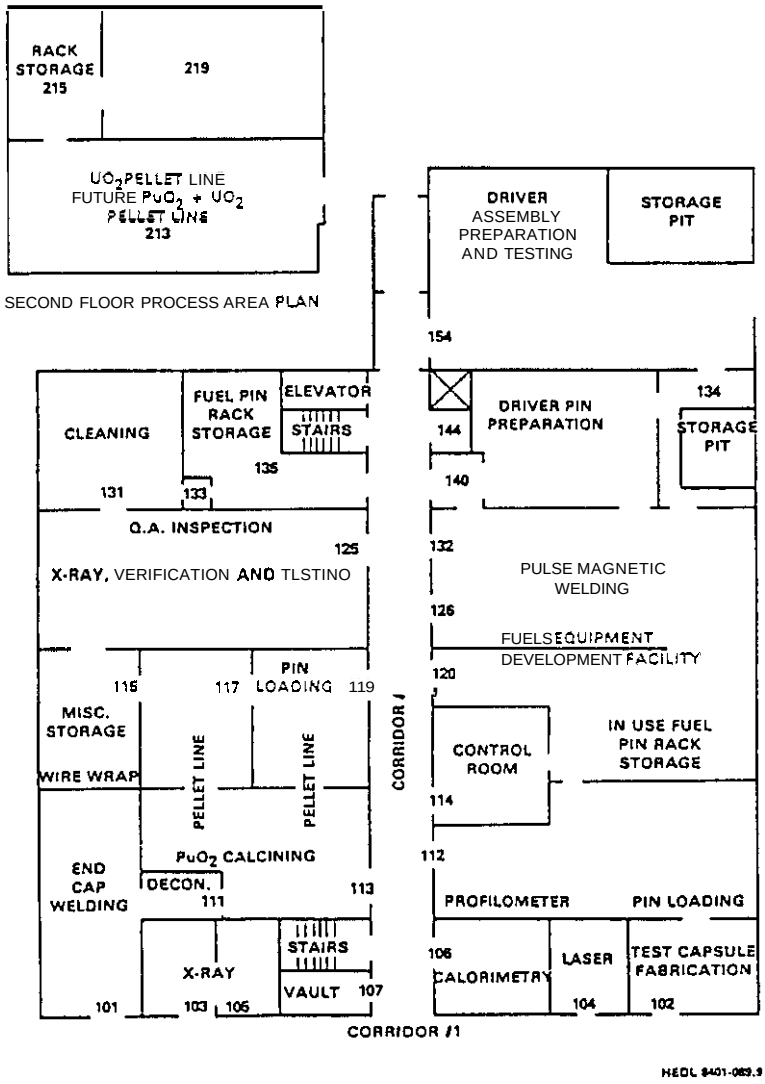
3.5.2.3 FFTF Support Activities Conducted in the 308 Building

The major portion of the MOX-containing materials that were generated in support of the recycled plutonium programs was in support of the FFTF program. The items were generated by HEDL from 308 Building operations. They were stored in the 324 Building and transferred to the PFP and stored in the vault as PFP Category Codes 950 and 955. The materials resulted from; the fabrication of FFTF test pins and EBR-II fuel pins, quality assurance inspections of FFTF driver fuel pins fabricated by Kerr-McGee and NUMEC, which later was taken over by B&W, and materials recovered from the closeout of fabrication operations by the vendors when their contracts were completed and subsequently shipped to HEDL.

Under HEDL the 308 Building served as a multipurpose nuclear facility containing varied activities of reactor fuel development, fuel fabrication, assembly inspection, engineering, quality assurance, fuels research, fuels design, ceramic research and nuclear material management. From its initial construction in 1960, the 308 Building was expanded several times to handle new missions. A large fuel assembly laboratory, Room 154, was added in 1970. Between 1975 and 1979 the laboratory areas was expanded several times, creating the 309-A Annex. [Tomlinson 1985] The floor plan of the facility as it was configured for the FFTF program is shown in Figure 10.

The FFTF project had its beginning in September 1963 with a small task force study. In January 1966, BNW was assigned project management responsibilities for design and construction of FFTF. Work associated with MOX fuel was conducted in the 308 Building. In July 1970 HEDL, operated by Westinghouse Hanford, assumed responsibility for the FFTF Project. Beginning in 1976, the main mission became the preparation and Quality Assurance testing of all FFTF fuel assemblies. [Gerber 19921

Figure 10 308 Building Process Area Plans
(Tomlinson 1985)



Licensed commercial vendors fabricated the driver FFTF fuel pins. The reference fuel fabrication process is depicted in Figure 11. NUMEC and Kerr-McGee prepared the fuel pins for FFTF core 1 and **2** between 1973 and 1975. B&W (which had purchased NUMEC) prepared the driver fuel pins for FFTF Cores 3 and **4** between December 1975 and August 1979. [Williams 1980] The driver fuel pins were $PuO_2 - DUO_2$ and $PuO_2 - NUO_2$. The **NU** was natural uranium. The assembly of driver fuel pins into completed assemblies was conducted in Rooms 138, Driver Pin Preparation, and 154, Driver Assembly Preparation and Testing. .

The DOE-licensed commercial vendors shipped driver fuel pins to HEDL. The fuel pins were fabricated into fuel assemblies in the 308 Building. All the fuel pins were subjected to stringent quality assurance verification. About one out of every 100 driver fuel pins was downloaded to ensure that the fuel pellets met the fast breeder reactor mixed oxide fuel pellet standard. Samples, standards, excess material and materials for recycle were stored in the 324 Building. At the completion of the vendor contracts, all remaining nuclear materials including those recovered during the cleanout of gloveboxes and equipment were shipped to HEDL. Some of the scrap material was sent to the SRS for plutonium recovery and the remainder was shipped to the PFP

Between 1971 and 1986 HEDL fabricated over 8,000 fuel pins in the 308 Building primarily in the form of EBR-II MOX test fuel pins or FFTF MOX test fuel pins. The uranium used in MOX test fuel prepared by HEDL was DU, **NU** or EU. Test Pins were also fabricated that contained only uranium oxide in varying ^{235}U isotopic enrichments. The operations used a variety of highly specialized fabrication and test equipment including inerted gloveboxes and open-face hoods. The following equipment was normally found in the gloveboxes and hoods used to fabricate test pins: V-blenders, Hex-blenders, ball mills, crushers, pellet presses, debinding and calcining furnaces, vacuum sintering furnaces, centerless grinders, vacuum storage modules, component cleaning equipment, welding equipment, leak testers, x-ray examination units, "gas-tagging'' equipment, NaK and Na filling equipment, and test-capsule fabrication equipment.

The test pin fabrication process is essentially the same as that for the driver fuel pins. The process is shown in Figure 12. Typically the PuO_2 and UO_2 for blending were passed through a screen a number of times until the material was deemed adequate for ball milling. The material was normally ball milled for **20** hours. The ball milled material was blended for 15 minutes in a twin shell or Hex blender. In the dry blend process, dry binder and MOX were mixed and tumbled for 10 minutes. The mixture was preslugged and retumbled to provide granules for pellet pressing. Dry binders used included Carbowax 3350, Carbowax 8000, Acrowax, methyl cellulose, and polyvinyl alcohol (powder). Pellets were pressed at pressures in the range of 30,000 to 80,000 psi. The binder was removed from the green pellets by heating the pellets to 600^0 C for four hours in a debinding furnace. The pellets are next sintered in a furnace at 1690^0 C for six hours. Total furnace running time including warm up and cool down was 20 hours. The pellets were analyzed for % Pu, %U, Pu and U isotopics, F, Cl, N, C, S and other impurities at the test designer's request. Pellets were usually ground to a controlled range

Figure 11 Reference Fuel Fabrication Process
(Williams 1980)

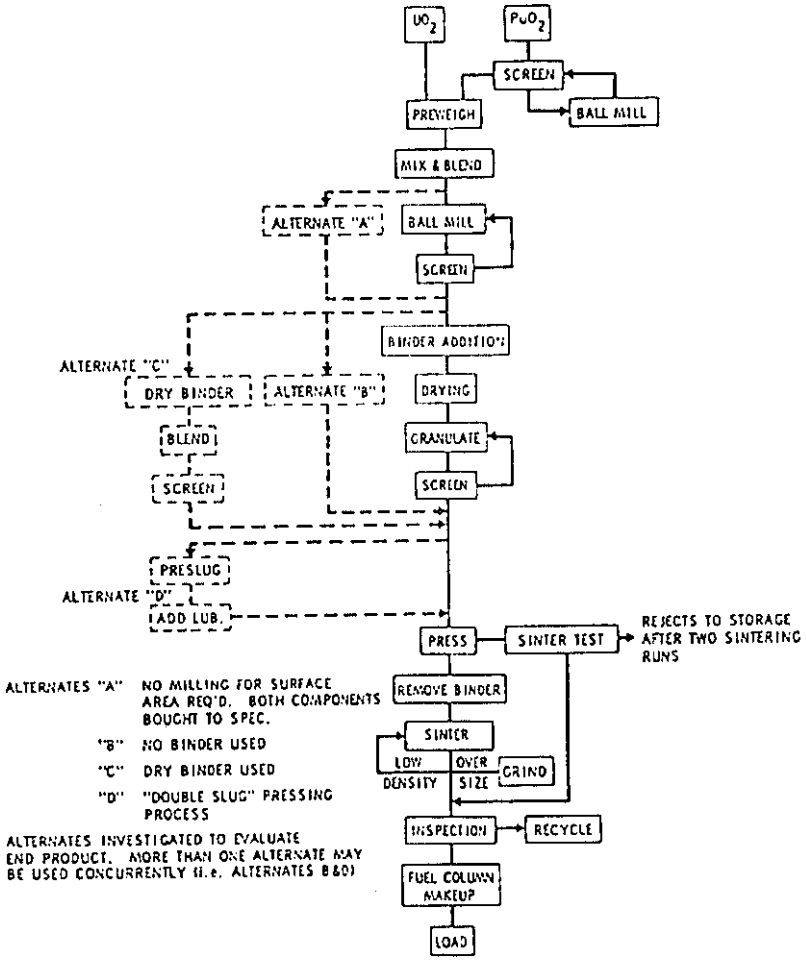
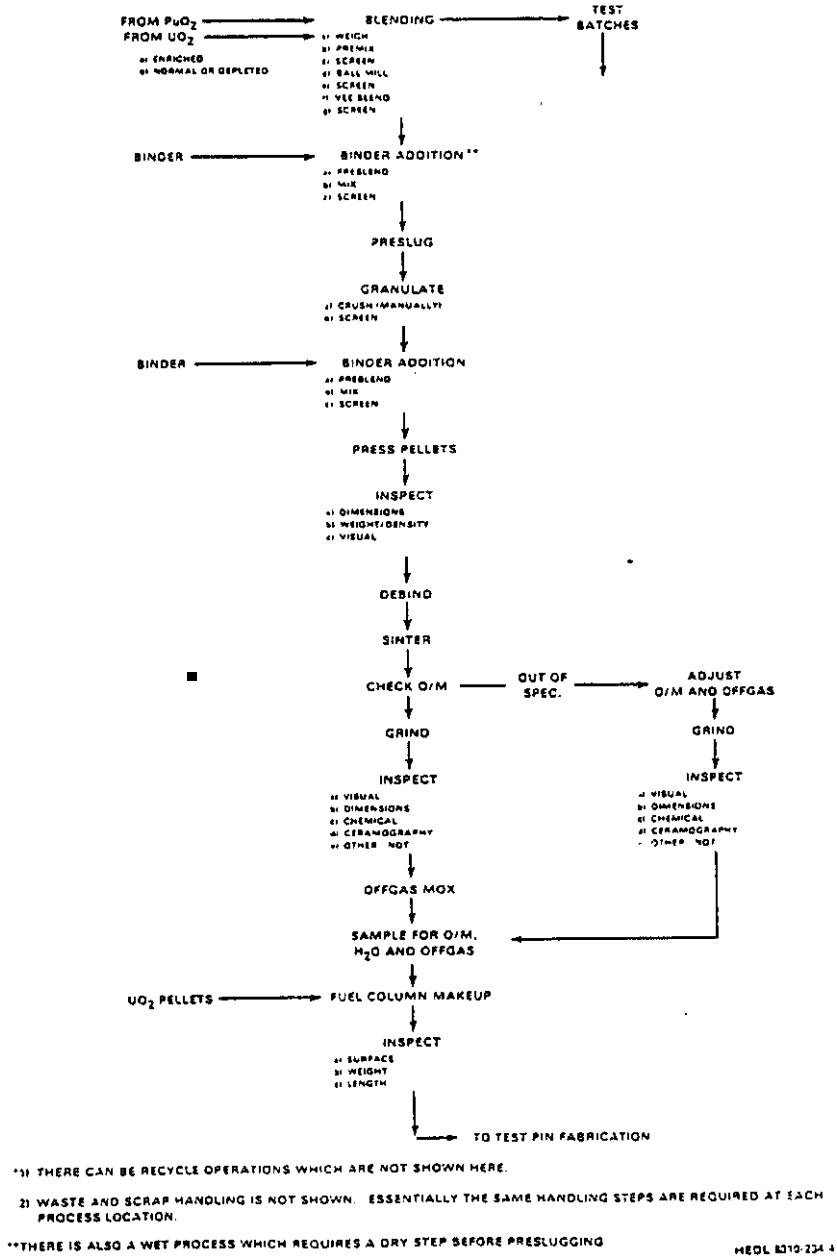


Figure **12** Mixed Oxide to MOX Pellets
(Tomlinson 1985)



of outer diameters. Pellet grinding was done on a centerless grinder with a diamond impregnated grinding wheel. Grinding was preformed without a coolant to eliminate impurity pickup. The pellets were then loaded into pins and the welded closed. The Non-destructive analysis of fuel and test pins took place in "clean" rooms. The last plutonium oxide fuels for FFTF were fabricated in October 1986. During the 1989-1990 time frame, test assemblies were fabricated containing MOX pellets made offsite, enriched uranium metal alloy fuel, and enriched uranium oxide fuel pellets.

Starting in the mid1980's the legacy FFTF plutonium-containing materials in the 324 Building in the 300 Area were transferred to the PFP for vault storage. The last of the 308 **SNM** inventory was transferred to the PFP in May 1992. Deactivation of the 308 Building was completed on March 31, 1994. [Gerber 19941

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Appendix 1 Development of the Plutonium Oxide and MOX Database

The list of PFP plutonium oxide items containing less than or equal to30 weight percent plutonium database was generated from the September 2001 ending Local Area Network Material Accountability System (LANMAS) database. The database was queried for items located in PFP material balance areas (MBAs) [>210, <290, and not 233,235,271 or 272]. The query identified 7,558 unique items by material name. Many items contain more than one type of nuclear material. Based on summary material type, i.e. plutonium, normal uranium, enriched uranium, thorium, etc., there are 10,105 SNM entries in the LANMAS database (Table A.l.) (Note: the accountability database tracks nuclear material by material types and not as discrete items)

Table A 1 Nuclear Material Inventory at PFP

Nuclear Material	Material Type	Number of Items
Plutonium	50	7,439
Depleted uranium	10	1,285
Enriched Uranium	20	914
Normal Uranium	81	427
Thorium	88	22
Plutonium-238	83	8
Americium-241	44	3
Californium-252	48	3
Neptunium-237	82	2
Uranium-233	70	2
	Total	10,105

The plutonium oxide and plutonium-uranium oxide (MOX) database was generated by the elimination of items based on COEI, ANSI, and PFP category codes, material name and SNM type. Most items were removed based on material codes. Items having the material codes listed in Table A.2 are not considered plutonium oxide and MOX and removed from the database on the basis of COEI, ANSI or PFP Category Code. Additional items were removed individually based on process knowledge and engineering judgment.

Table **A** 2 PFP Nuclear Material Not Identified as Plutonium Oxide or MOX

Category Name	Code
Unalloyed Buttons Product	132
In Fuel Element and Target Fabrication Process	291
Assembled Items Product	309
Nitrate Solutions Product	409
In Recovery Process – Unirradiated	700
Nitrate Solutions Product	703
Unalloyed metal – Unirradiated	721
Alloyed metal – Unirradiated	722
Combustibles – Unirradiated	726
Solutions – Unirradiated	728
Incinerator Ash/Residue	G03
Reduction Residues	G05
PuF_3/UF_6	C10
PuF_4/UF_4	C11
Encapsulated Pu Compounds/UR Carbides	C30
Line System Holdup	614
Plutonium Dylene Scrap	65
Waste Drums for Burial	37
Radiation Sources for Recovery	849
Radiation Sources	850
Awaiting Burial	34
FAC SRV Equipment Holdup-Filter	603
Hood Holdup	611
FAC SRV Equipment Holdup-Hood	612
Filterbox Holdup	613
Miscellaneous MBA Holdings	630
Prd H&O equipment Holdup - Hood	632
Incinerator Ash Standards	31

After removal of the majority of the non plutonium oxide and MOX items based on material codes, the list was screened to remove items that were not plutonium oxide or MOX based on item material names, location, container code, and process knowledge. Items in PFP category code **453** with item material **names** starting with H5###, RHZ###,

E3##, E4#3, E5##, E6## and E-4### were removed. Some of these items contain plutonium oxide packaged in the Bagless Transfer Can (BTC) or the 3013 container, but the **LANMAS** database doe not provide distinction between metal items and stabilized plutonium oxide items. Also items whose item material names included HC##, -M##, In Process, and "Case" **were** eliminated. Items in MBA 211 were also deleted. Items stored in PR cans or FL10 containers were also deleted. Items listed stored in the 2736ZB Repackaging GB were also eliminated. Items, listed as containing only plutonium-238, americium-241, californium-252, and uranium-233, were also deleted. What resulted is a proposed list of 5,425 items that contain plutonium oxide or plutonium-uranium oxide and that were slated for disposal under the 94-1 directive.