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**OAK RIDGE  
NATIONAL  
LABORATORY**



**Availability of Uranium Feed for the Fissile Materials  
Disposition Program**

**Volume 2 : Depleted Uranium Trioxide**

**V. S. White  
R. L. Reid**

MANAGED AND OPERATED BY  
LOCKHEED MARTIN ENERGY RESEARCH CORPORATION  
FOR THE UNITED STATES  
DEPARTMENT OF ENERGY

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**AVAILABILITY OF URANIUM FEED FOR THE FISSILE MATERIALS  
DISPOSITION PROGRAM**

**VOLUME 2: DEPLETED URANIUM TRIOXIDE**

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## ACRONYMS

ADU	ammonium diuranate
AEA	Atomic Energy Act of 1954
ASME	American Society for Mechanical Engineers
CFR	<i>Code of Federal Regulations</i>
DOE	U.S. Department of Energy
DOT	U.S. Department of Transportation
EPA	U.S. Environmental Protection Agency
EPACT	Energy Policy Act of 1992
FEMP	Fernald Environmental Management Project
FMDP	Fissile Materials Disposition Program
HF	hydrogen fluoride (hydrofluoric acid)
HM	heavy metal
INEEL	Idaho National Engineering and Environmental Laboratory
LEU	low-enriched uranium
LWR	light-water reactor
MOX	mixed oxide
MT	metric tons
MTHM	metric tons heavy metal
RCRA	Resource Conservation and Recovery Act of 1976
SRS	Savannah River Site
UNH	uranyl nitrate hexahydrate
USPS	U.S. Postal Service

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# Availability of Uranium Feed for the Fissile Materials Disposition Program

## Volume 2: Depleted Uranium Trioxide

V. S. White  
R. L. Reid

### ABSTRACT

Uranium dioxide (UO<sub>2</sub>) powder makes up more than 95 wt % of the feedstock needed for the domestic mixed-oxide (MOX) fuel fabrication facility to be constructed by the U.S. Department of Energy's (DOE's) Fissile Materials Disposition Program (FMDP) as part of its "dual-path" plutonium disposition strategy. The needed uranium dioxide feed can be derived from natural or depleted uranium compounds. The first volume in this series on uranium feed considered domestic sources of depleted uranium in the form of uranium hexafluoride (UF<sub>6</sub>). This second volume considers depleted uranium trioxide (UO<sub>3</sub>).

Depleted UO<sub>3</sub>, recovered either through chemical separation procedures used in <sup>239</sup>Pu production programs or as a by-product from production reactor target and weapons component fabrication, is now stored primarily in solid form in 55-gal drums located at the Fernald Environmental Management Project and the Savannah River Site. The FMDP's depleted UO<sub>3</sub> requirements are a small portion (~5%) of the total depleted UO<sub>3</sub> available. More problematical is the need for a facility to convert the depleted UO<sub>3</sub> to a stable, high-quality depleted UO<sub>2</sub> powder.

The first volume in this series of reports on FMDP uranium feed materials discussed the depleted UF<sub>6</sub> storage site inventories, cylinder conditions, transportation/regulatory issues, and the chemical conversion of depleted UF<sub>6</sub> to depleted UO<sub>2</sub> powder. This second volume discusses the same issues relative to depleted UO<sub>3</sub>.

The purpose of this document is to support the DOE's FMDP procurement activity for the reactor-based MOX option. This report is one of several topical reports generated to provide background information on various subjects related to manufacturing and burning MOX fuel. This document assumes the use of depleted uranium as the diluent for the weapons-grade plutonium and provides information on the uranium source inventory and subsequent handling and processing of uranium feed.

## 1. OVERVIEW

The U.S. Department of Energy (DOE) currently owns ~19,500 metric tons heavy metal (MTHM) equivalent of depleted uranium trioxide (UO<sub>3</sub>) resulting from historical weapons production programs within the U.S. defense complexes. UO<sub>3</sub> is also referred to as uranyl oxide. The primary sites with UO<sub>3</sub> inventories are the Fernald Environmental Management Project (FEMP) in Ohio and the Savannah River Site (SRS) in South Carolina. These two government installations store depleted UO<sub>3</sub> equivalent to 22,000 MT of depleted uranium dioxide (UO<sub>2</sub>).

The conversion ratios used in this report for determining equivalencies are as follows:

$$\text{U to UO}_2: (270.03 \text{ mol. wt})/(238.03 \text{ mol. wt}) = 1.1344 \text{ .}$$

$$\text{U to UO}_3: (286.03 \text{ mol. wt})/(238.03 \text{ mol. wt}) = 1.2017 \text{ .}$$

$$\text{UO}_2 \text{ to UO}_3: (286.03 \text{ mol. wt})/(270.03 \text{ mol. wt}) = 1.0593 \text{ .}$$

Natural uranium consists of 0.006 wt %  $^{234}\text{U}$ , 0.711 wt %  $^{235}\text{U}$ , and the rest as  $^{238}\text{U}$ . The fissile isotope is  $^{235}\text{U}$ ; uranium is considered depleted if the total  $^{235}\text{U}$  content is less than 0.711 wt %, as found in nature. The percentage composition, referred to as assay, of low-enriched uranium (LEU) necessary for controlled fission in nuclear power reactors is 1.8 to 5.0 wt %  $^{235}\text{U}$ ; the average composition of  $^{235}\text{U}$  in depleted uranium is 0.20 wt %.

Resulting from a combination of changes in DOE's nuclear materials and weapons programs, decreasing federal budgets, and the enactment of the Energy Policy Act (EPACT) of 1992, DOE is reviewing options for disposition of materials in inventory, which includes depleted  $\text{UO}_3$ . Among other requirements, the EPACT instructed DOE to (1) perform a comprehensive inventory of all DOE-owned uranium, including depleted material, and (2) determine the availability of conversion services and possible commercial uses with recommendations for disposition of such inventories. DOE began assessing government-owned depleted uranium inventories through the Materials in Inventory Initiative in February 1995.

Depleted  $\text{UO}_3$  is a recycled product or by-product and is not considered waste. Section 11(z) of the Atomic Energy Act (AEA) of 1954, in concurrence with the U.S. Nuclear Regulatory Commission in the *Code of Federal Regulations* (10 CFR 40.4), defines uranium in any physical or chemical form as source material. The AEA further defines depleted uranium as source material uranium with less than 0.711 wt %  $^{235}\text{U}$ , thus excluding it from jurisdiction under U.S. Environmental Protection Agency (EPA) and Resource Conservation and Recovery Act (RCRA) of 1976 regulations. Management and oversight of depleted uranium by DOE's Office of Nuclear Energy, Science, and Technology and DOE's Office of Environmental Management is not under the control of the EPA or any other government agency.

The smaller component of mixed-oxide (MOX) fuel, plutonium, is composed primarily of the  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  isotopes and is classified in three basic grades for use in (1) weapons (7.0 wt % or less  $^{240}\text{Pu}$ ), (2) breeder reactors (7.0 to 19.0 wt %  $^{240}\text{Pu}$ ), and (3) light-water reactors (LWRs) (greater than 19.0 wt %  $^{240}\text{Pu}$ ). Approximately 50 MTHM of weapons-quality plutonium (plutonium of 93.0 wt %  $^{239}\text{Pu}$  or greater) have been declared surplus to defense program requirements. Additional quantities of weapons plutonium may become surplus in the future.<sup>1</sup>

DOE's Office of Fissile Materials Disposition is currently beginning implementation of options for disposing of weapons-program plutonium that is surplus to national defense requirements based on the *U.S. Nonproliferation and Export Control Policy*<sup>2</sup> and the *Joint Statement Between the United States and Russia on Nonproliferation of Weapons of Mass Destruction and Means of Their Delivery*.<sup>3</sup> These policies focus on five primary efforts:<sup>4</sup>

- security of nuclear materials located in the former Soviet Union,
- long-term, safeguarded storage and disposition of surplus fissile materials,
- nuclear arms reductions that are transparent and irreversible,
- stronger nuclear nonproliferation actions, and
- control of nuclear material exports.

Diluting surplus plutonium with DOE-owned depleted uranium for use as LWR fuel is one of two disposition alternatives being pursued as part of the Fissile Materials Disposition Program's (FMDP's) "dual-path" strategy. The plutonium would be the primary fissile material (~4% of the contents) combined with depleted uranium to produce MOX fuel for commercial reactors. Approximately 35 MTHM of weapons-grade plutonium may be dispositioned as MOX in LWRs. This approach provides a feasible solution that accomplishes the plutonium disposition mission and also affords a disposal method for some environmental legacy depleted uranium resulting from weapons programs.

The plutonium isotopic composition subsequent to usage as fuel would be elevated in  $^{238}\text{Pu}$  (1.5 wt %),  $^{240}\text{Pu}$  (2.2 wt %),  $^{241}\text{Pu}$  (13.5 wt %), and  $^{242}\text{Pu}$  (5.0 wt %) and would be rendered less reactive and more difficult to handle than weapons plutonium. Although disposition of plutonium as MOX fuel in commercial LWRs does not destroy a high percentage of plutonium, fuel irradiation results in increased radioactivity of fission products, which coupled with the fabricated assembly form containing the material, minimizes the potential for unauthorized removal of plutonium from the reactor or repository site. Also, burning of plutonium in a reactor reduces the amount of the highly fissionable isotope  $^{239}\text{Pu}$ , which is the primary isotope used in weapons productions.

The primary steps involved in using depleted  $\text{UO}_3$  as feedstock in the chemical form of depleted  $\text{UO}_2$  for weapons-grade plutonium are as follows:

1. Retrieving the drums from their storage location.
2. Inspecting and certifying the drums for shipping.
3. Preparing the drums for transportation and loading onto the transportation source (truck or train).
4. Transporting the drums, including unloading functions, by road or rail service from their current location (FEMP and SRS) to a commercial facility or DOE site housing the conversion or MOX facility. [The Hanford Reservation, Idaho National Engineering and Environmental Laboratory (INEEL), Pantex, and the SRS are under consideration for the MOX facility].
5. Transferring the depleted  $\text{UO}_3$  to the receiving and storage facility at the conversion facility.
6. Converting depleted  $\text{UO}_3$  to  $\text{UO}_2$  using a commercial conversion facility with approved environmental permits and licenses.
7. Preparing depleted  $\text{UO}_2$  powder for the blending process. This involves (a) filling clean drums for transporting to the MOX facility if not collocated with conversion facility; (b) transporting, including loading and unloading the drums, to the DOE site housing the MOX facility if not collocated with the conversion facility; and (c) transferring materials accountability to the MOX facility.
8. Adhering to EPA- and RCRA-approved methods for disposing of wastes generated from the conversion process.

The depleted  $\text{UO}_3$  being considered from SRS and FEMP contains impurities. The sources of these impurities are discussed in Chap. 2. These impurities may present an obstacle to the commercial fuel fabricators in processing the  $\text{UO}_3$  to  $\text{UO}_2$ . In some instances, the fabricators would require a license modification to allow for trace amounts of plutonium contained in the  $\text{UO}_3$  in their plants. Also, the obstacle of public acceptance of having trace amounts of plutonium on their sites could discourage some of the fabricators from accepting this form of depleted  $\text{UO}_3$ . Once these obstacles are overcome, the depleted  $\text{UO}_3$  could be converted to  $\text{UO}_2$  through either

- a wet process employing a uranium hexafluoride ( $\text{UF}_6$ ) conversion step to remove the impurities,
- a wet process using solvent extraction to remove the impurities, or
- a dry process employing a  $\text{UF}_6$  step to remove the trace impurities.

According to industry representatives, a desired dry conversion process converting the impure  $\text{UO}_3$  directly to reactor-grade  $\text{UO}_2$ , without the  $\text{UF}_6$  step to remove the impurities, is not currently available.

In summary, DOE owns surplus depleted  $\text{UO}_3$  that could be used as feed with plutonium for a MOX facility. It appears possible to convert sufficient quantities of depleted  $\text{UO}_3$  to the chemical form of  $\text{UO}_2$  by either an aqueous process or a dry process employing a  $\text{UF}_6$  conversion step.

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## 2. DEPLETED $UO_3$ INVENTORY AND STORAGE CONTAINERS

Depleted uranium, in the form of 96% pure  $UO_3$ , is stored at FEMP and SRS. The average percentage composition of  $^{235}U$  contained in the depleted  $UO_3$  is ~0.20 wt %  $^{235}U$  at FEMP and ~0.18 wt %  $^{235}U$  at SRS. Depleted  $UO_3$  is a reddish-yellow or orange powder in crystalline form and is primarily stored in plastic-lined, 55-gal steel drums. Packaging and transportation is regulated to ensure that nuclear materials safeguards, control, and accountability measures are followed. Depleted  $UO_3$  must be transported in U.S. Department of Transportation (DOT)-approved containers (7A Type A, LSA-1).

Because of degradation from corrosion or mishandling, the condition of some drums may be such that they are no longer transportable. Drums used for transportation of depleted  $UO_3$  must meet DOT specifications for 7A Type A containers. In addition, depleted  $UO_3$  is packaged, transported, and stored in drums adhering to regulations and policies provided in the various publications listed in Chap. 5 of this report.

The depleted  $UO_3$  started out as natural  $UF_6$  in a long series of steps related to the SRS plutonium production mission. Depleted  $UF_6$ , a by-product of DOE's gaseous diffusion uranium enrichment process, is discussed in Vol. 1 of this report. Impurities were introduced through the use of depleted uranium and its compounds in the U.S. weapons program and through the chemical and physical processes that took place at various defense production facilities within the complex. Figure 1 depicts the steps in the transformation of virtually pure depleted  $UF_6$  to a  $UO_3$  by-product containing multiple impurities.<sup>5</sup>

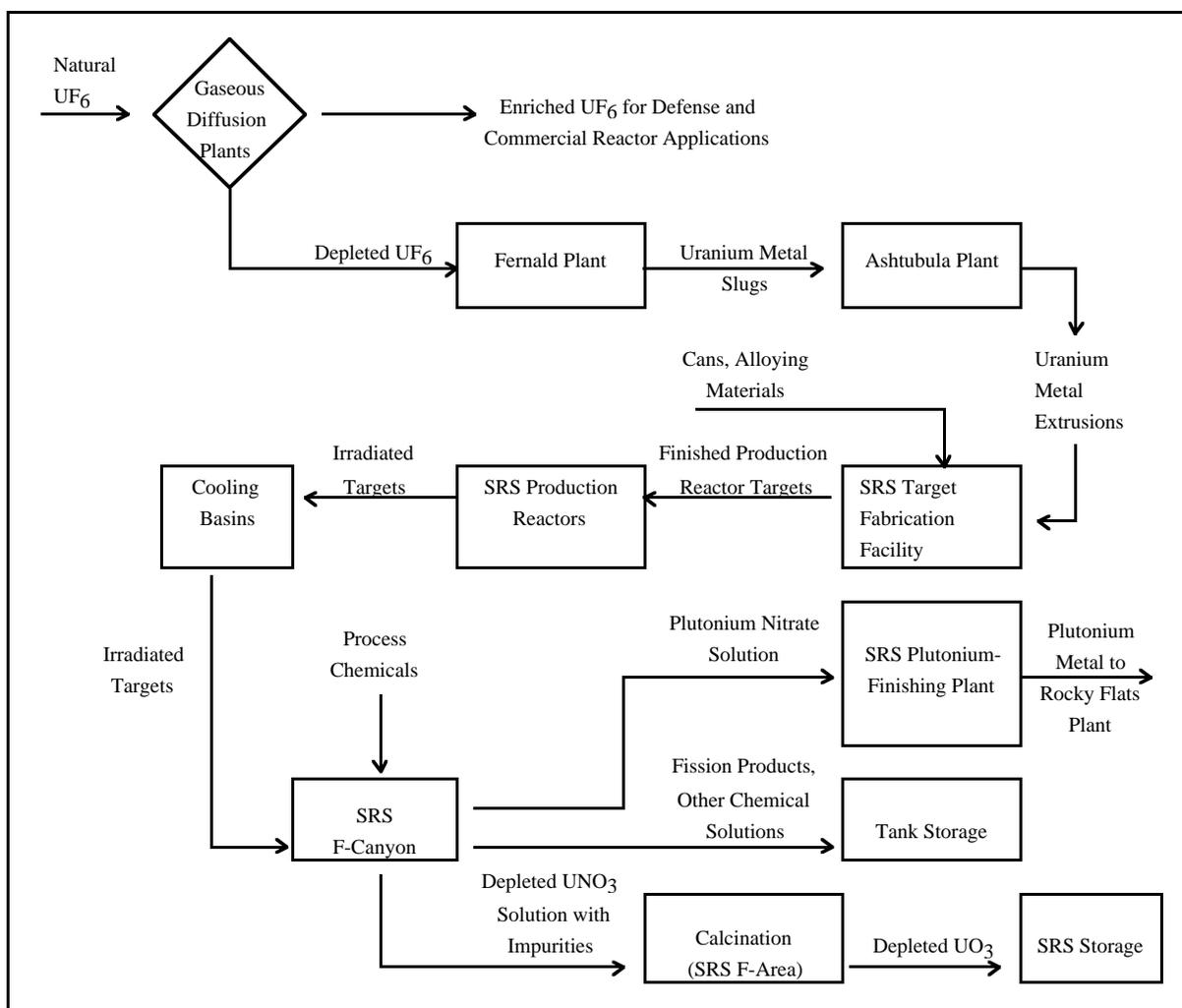


Fig. 1. Steps in generation of  $UO_3$  by-product from plutonium production at SRS.

Historically, depleted uranium had two primary uses in the weapons complex:

1. metal for fabrication of various weapons components and
2. alloyed metal, as the source material, for depleted uranium targets that were subsequently irradiated in the SRS production reactors to produce weapons-grade plutonium.

For weapons use, pure depleted  $UF_6$  was reduced to uranium metal in a process that involved defluorination to the intermediate product uranium tetrafluoride ( $UF_4$ ) and batch reduction of  $UF_4$  to relatively pure depleted uranium metal. These operations took place at two DOE uranium refinery facilities: the Mallinckrodt Facility at Weldon Springs, Missouri, and the Feed Materials Processing Center (Fernald Plant), presently known as FEMP, northwest of Cincinnati, Ohio.

For component fabrication, the depleted uranium metal was processed into weapons shapes at either the Oak Ridge Y-12 Plant or the Rocky Flats Plant. A small amount of  $UO_3$  from SRS was sent to FEMP to enhance the  $UF_4$  reduction step for metal preparation. Much of this  $UO_3$  is still in drums at FEMP. No high-purity  $UO_3$  was produced as part of FEMP operations.

For the SRS plutonium production reactor use, pure  $UF_6$  tails from the diffusion process were reduced to uranium metal through defluorination to the intermediate product  $UF_4$  followed by batch reduction of  $UF_4$  to relatively pure depleted uranium metal. The majority of this activity took place at the FEMP facility. The depleted uranium metal billets were sent to Reactive Metals Incorporated in Ashtabula, Ohio, for extrusion into tubular slugs, which were then sent to SRS for finishing and final bonding into tubular metal cans. These depleted uranium targets were irradiated in the SRS production reactors for a few months for conversion of  $^{238}U$  to  $^{239}Pu$ . This plutonium isotope at concentrations of 93%+ in total plutonium was desired for weapons applications.

After a cooling period, the irradiated targets were transferred to the SRS F-Canyon for dissolution and subsequent solvent extraction removal of the plutonium, fission products, and other dissolved chemical and metal contaminant streams. The SRS plutonium-finishing facility then converted the plutonium solution to pure plutonium metal for shipment to Rocky Flats for use in weapons. The remaining depleted uranium nitrate solution was then calcined to a dry  $UO_3$  powder, which is now stored on site at SRS.

Because solvent extraction could not economically remove or separate 100% of the fission products, plutonium, alloying constituents, or other chemicals added during F-Canyon operations, there are small residual amounts of these impurities left in the dry  $UO_3$ . The radioactivity of the depleted  $UO_3$  results mainly from the small amount of plutonium and fission product impurities.

The  $UO_3$  inventory at FEMP can be found in Ref. 6, with additional background information in Ref. 7.

The  $UO_3$  at FEMP and SRS are similar in composition and level of impurities. The 96% purity level is a result of both the deliberate insertion of contaminants to break down the powder particle size for use in plutonium production processes and the subsequent recycle of production reactor materials. According to information from the Westinghouse Savannah River Company, the impurities consist of the materials and metals shown in Table 1.<sup>8</sup>

During November and December 1994, Carolina Metals, Inc., analyzed SRS  $UO_3$  in a study conducted for Westinghouse Savannah River Company.<sup>9</sup> This analysis consisted of an assessment of current  $UO_3$  management practices, characterization, and condition of packaging containers; determination of the  $UO_3$  chemical and physical properties; and a description of use, storage, and disposal alternatives. Twenty sample drums were selected for the analysis. These drums were placed in overpacks and were shipped off-site to Carolina Metals, Inc., in Barnwell, South Carolina, for evaluation. The drums, stored since 1981 and 1982, exhibited external corrosion. A number of the drums were severely corroded on the bottom because of previously being stored in standing water. None of the 20 drums exhibited internal corrosion, but all had a residual layer of  $UO_3$  powder resulting from damage or embrittlement of the internal plastic liner or tape sealing the liner.

Radiation readings from the sampled drums ranged from 3.0 to 3.5 mrem/h. Following sampling taken from the top of each drum, repackaging in new drums, and insertion into overpacks, all readings were well below the 200 mrem/h allowable limit. Table 2 provides a chemical analysis from the sampled drums.

The current DOE depleted  $UO_3$  inventory is summarized in Table 3. All inventories are small, with the exception of  $UO_3$  stored at FEMP and SRS. SRS inventory is the only single storage source large enough to meet  $UO_2$  program requirements.

**Table 1. Average impurities found in DOE-owned depleted UO<sub>3</sub>**

Impurity	Average level
Plutonium	<10 ppb
Trace materials	
Sodium	300 ppm
Iron	200 ppm
Aluminum	100 ppm
Phosphorus	100 ppm
Chromium	75 ppm
Silicon	50 ppm
Nickel	30 ppm
Copper	15 ppm
Calcium	10 ppm
Molybdenum	10 ppm
Lead	5 ppm
Environmental protection toxicity metals	
Barium	<2.0 mg/L
Chromium	<1.5 mg/L
Arsenic	<1.0 mg/L
Lead	<0.3 mg/L
Selenium	<0.3 mg/L
Silver	<0.1 mg/L
Cadmium	<0.1 mg/L
Mercury	<0.02 mg/L

**Table 2. Chemical analysis from sampling of SRS depleted UO<sub>3</sub> (Ref. 9)**

Chemical component	Range	Average level
Uranium	78.47–82.25%	80.34%
Plutonium	0.333–9.52 ppb	2.85 ppb
Trace materials		
Sodium	<10–300 ppm	48 ppm
Iron	29–323 ppm	100 ppm
Aluminum	20–>100 ppm	68 ppm
Phosphorus	<25–100 ppm	90 ppm
Chromium	20–100 ppm	41 ppm
Silicon	<10–150 ppm	41 ppm
Nickel	< 10–50 ppm	21 ppm
Copper	<2–15 ppm	4 ppm
Calcium	<5–10 ppm	6 ppm
Molybdenum	<10 ppm	10 ppm
Lead	<5–5 ppm	5 ppm

**Table 3. Total inventory of DOE-owned depleted UO<sub>3</sub>**

Location	Uranium (MTHM)	Drums	Reference/comments
FEMP	34.452	126	B. Ison, Fluor Daniel Fernald Interoffice Memorandum, "Uranium Inventory Position at Fernald as of January 1, 1997," FD Fernald #: M:WMTSP(NMDP):97-0002, January 24, 1997.
Hanford	0.607		Hanford transferred 103 MT of uranium to low-level waste in FY1996; quantity remaining is residual powder in drums. D. C. Lini and G. B. Hulse, <i>Uranium Disposition Strategy for the Hanford Site</i> , U.S. Department of Energy, HNF-MR-0533, February 1997.
Paducah	0.712		J. M. Bird, U.S. Department of Energy, Oak Ridge Operations, facsimile to V. S. White, Oak Ridge National Laboratory, May 1, 1997 (data as of December 31, 1996).
Portsmouth	0.068		J. M. Bird, U.S. Department of Energy, Oak Ridge Operations, facsimile to V. S. White, Oak Ridge National Laboratory, May 1, 1997 (data as of December 31, 1996).
SRS	<u>19,436.276</u>	<u>35,835</u>	S. D. Brady et al., Depleted Uranium Trioxide (UO <sub>3</sub> ) Characterization and Storage, 94AB652387P-00F, Carolina Metals, Inc., 1995 (data as of January 6, 1995).
Total	19,472.115	35,961	

Table 4 provides the depleted UO<sub>2</sub> equivalencies for the primary stocks of government-owned depleted UO<sub>3</sub> inventories. Assuming a 4.0 wt % fissile composition, the drums contain sufficient inventory to accomplish a blend ratio of 1 part plutonium to 25 parts depleted uranium. A 35-MTHM plutonium program would require ~875 MTHM of depleted uranium, equating to 1000 MT of depleted UO<sub>2</sub> or ~1050 MT of depleted UO<sub>3</sub>. Approximately 4.5% of the total government-owned inventory of depleted UO<sub>3</sub> would be required for blending the currently identified weapons program surplus of 35 MTHM of plutonium. Drums are available for shipping from FEMP or SRS. The contents may need to be transferred to other containers or placed in overpacks before transporting to the conversion facility because of drum degradation.

Several government sites contain additional depleted uranium inventories in the forms of UF<sub>6</sub>, alloyed and unalloyed metals, oxides, nitrates, hydrides, and aqueous solutions. Even though inventory data have routinely been classified at most weapons manufacturing facilities, those facilities no longer in production mode are releasing inventory data subsequent to the DOE Openness Initiative.<sup>1</sup> The availability of UF<sub>6</sub> is discussed in Vol. 1 of this topical report series (ORNL/TM-13417 Rev. 1).

**Table 4. Depleted UO<sub>2</sub> equivalent inventory of DOE-owned depleted UO<sub>3</sub> at primary repositories**

Location	Uranium (MTHM)	UO <sub>2</sub> Equivalent (MT)	UO <sub>3</sub> Equivalent (MT)
FEMP	34.452	39.082	41.401
SRS	<u>19,436.276</u>	<u>22,048.512</u>	<u>23,356.573</u>
Total	19,470.728	22,087.594	23,397.974

### 3. DEPLETED $\text{UO}_3$ TO $\text{UO}_2$ CONVERSION PROCESS AND FACILITIES

For the purposes of the MOX fuel fabrication facility acquisition strategy and conceptual design, it is assumed that MOX requirements are a dry, clean, free-flowing depleted or natural  $\text{UO}_2$  powder capable of being readily blended with a plutonium dioxide ( $\text{PuO}_2$ ) powder. The  $\text{PuO}_2$  will be derived from a hydride process such as the Advanced Recovery and Integrated Extraction System/Thermally Induced Gallium Removal.

No commercial or DOE materials inventories contain the 1000 MT or more of depleted  $\text{UO}_2$  of high-quality powder needed for the MOX plutonium disposition mission. This report does not consider the near-term problem of providing  $\text{UO}_2$  blend stock for the MOX test rods required for irradiation in the INEEL Advanced Test Reactor.

Conversion refers to the process of transforming depleted  $\text{UO}_3$  to  $\text{UO}_2$ . This conversion step is similar to that for converting plutonium "pits" or other materials to  $\text{PuO}_2$  powder for ultimate blending with the depleted  $\text{UO}_2$  powder. If such a service is not available elsewhere, the DOE-funded MOX fuel fabrication facility will be required to provide this conversion step of depleted uranium to  $\text{UO}_2$  as part of the MOX fuel fabrication facility. This will mean additional investment costs for the government. There are also environmental consequences of adding the conversion step, which would have to be addressed in the National Environmental Policy Act of 1969 process.

The overall generic fuel-fabrication flow sheet consists of feed receipt; powder preparation (blending, milling, granulating, and incorporating additives); pellet fabrication (sintering, grinding, and inspection); rod fabrication; packaging; and assembling of fuel bundles required by the specific reactor's core loading requirements. The chemical conversion of the depleted  $\text{UO}_3$  to oxide powder for subsequent blending with  $\text{PuO}_2$  powder is an initial step for MOX fuel processing, as shown in Fig. 2.

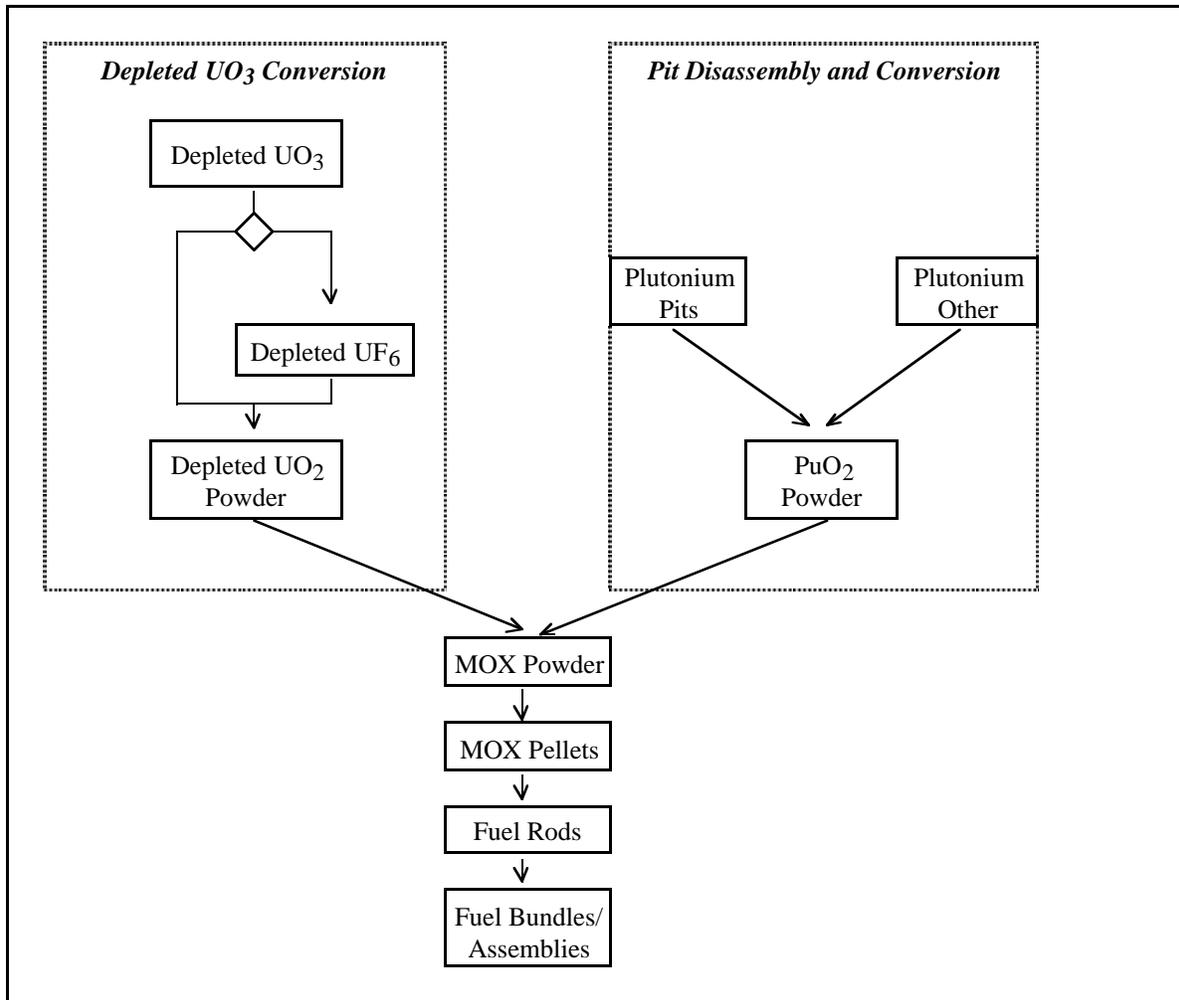
The health, safety, and environmental risks involved in the conversion process and handling of materials are not discussed in this report.

The process of converting  $\text{UO}_3$  powder to  $\text{UO}_2$  can be accomplished directly by a calcination/reduction process (heating the  $\text{UO}_3$  in flowing hydrogen gas), and pellets can subsequently be pressed from the resulting  $\text{UO}_2$  powder.<sup>10</sup> However, the depleted  $\text{UO}_3$  powder that is being considered in this program is contaminated with trace impurities, as indicated in Tables 1 and 2. Removing these impurities requires either a solvent extraction step in the conversion process or a step converting the depleted  $\text{UO}_3$  to  $\text{UF}_6$  to remove the impurities via  $\text{UF}_6$  distillation with subsequent conversion of  $\text{UF}_6$  to  $\text{UO}_2$ .

There are programmatic objections to converting the depleted  $\text{UO}_3$  to depleted  $\text{UF}_6$  because copious quantities of depleted  $\text{UF}_6$  currently exist, as indicated in Vol. 1 of this report. The conversion process of choice is an all-dry process because it is the more environmentally acceptable of the available conversion alternatives. However, based on informal discussions with domestic LEU fuel fabricators, there is no current way to remove impurities from depleted  $\text{UO}_3$  without an aqueous step or conversion to  $\text{UF}_6$ .

Commercial nuclear fuel fabricators indicated that at the appropriate step they could put clean  $\text{UO}_3$  directly into either their wet or dry nuclear fuel process lines. However, there was no assurance that the impure  $\text{UO}_3$  would be accepted for fabrication in their lines. Some of the fuel fabricators' licenses preclude accepting any plutonium on site, even in trace amounts. Other fabricators' licenses allow plutonium on site in quantities less than 20 ppb, which would not preclude the impure  $\text{UO}_3$  considered in this study based on plutonium content alone. It is uncertain and considered highly improbable that the LEU fabricators would risk the adverse public opinion of having any plutonium in any quantity on site.

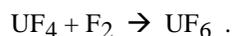
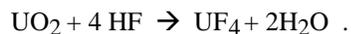
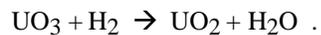
As previously mentioned, before the depleted  $\text{UO}_3$  enters the depleted  $\text{UO}_2$  conversion facility, the  $\text{UO}_3$  may need to be either converted to  $\text{UF}_6$  or go through an aqueous solvent extraction process to remove the impurities. It is highly unlikely that current fuel fabricators would want to contaminate their lines with recycled  $\text{UO}_3$  from government-owned inventory. Process diagrams of the various options available to FMDP for conversion of  $\text{UO}_3$  are shown in Figs. 2 through 6. These are in no particular order of importance.

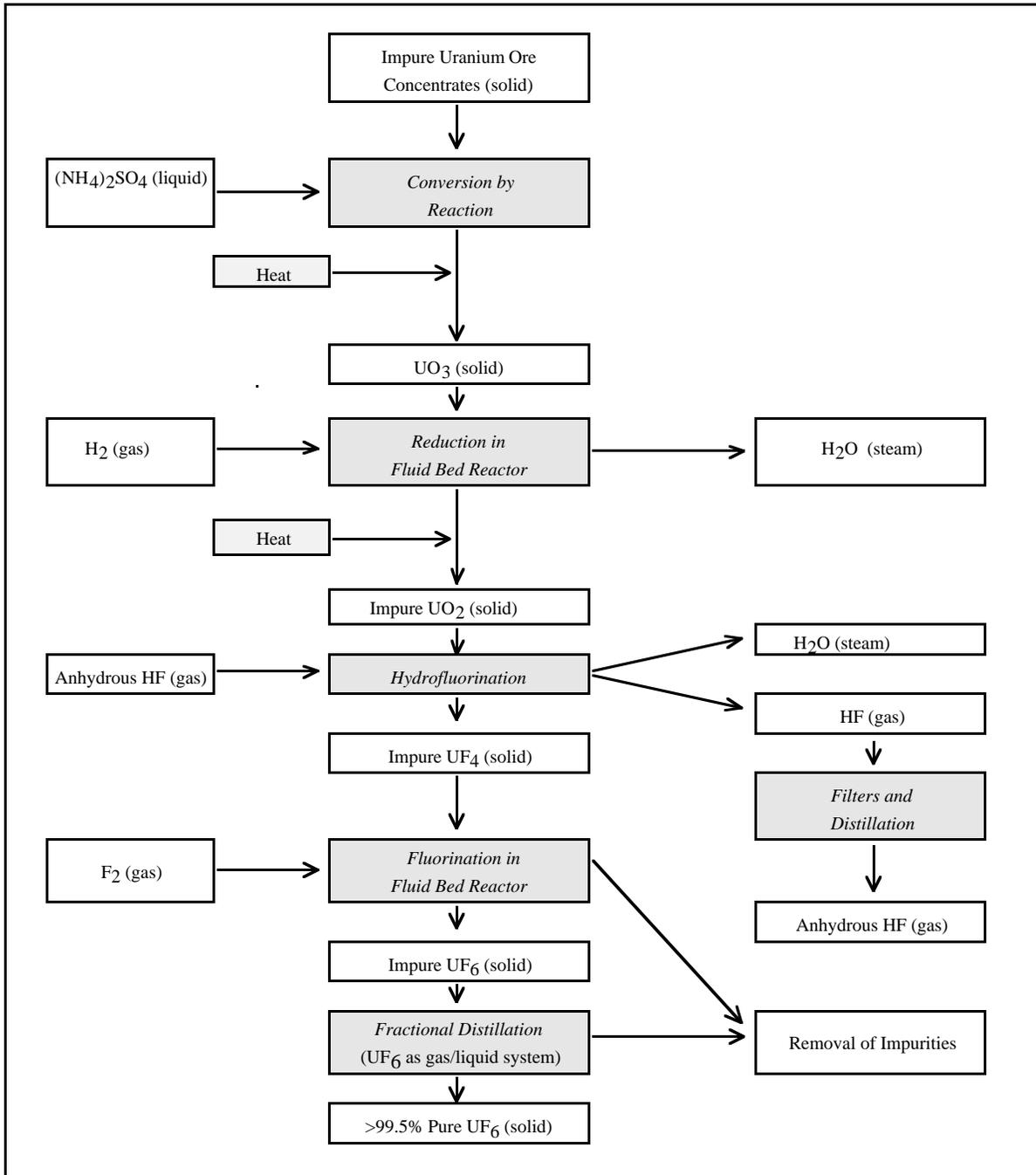


**Fig. 2. Material process flow for MOX fuel.**

Figure 3 shows a process flow diagram of the natural uranium concentrates to UF<sub>6</sub> dry conversion process. The UO<sub>3</sub> is initially reduced to impure UO<sub>2</sub> before the fluorination steps. The UF<sub>6</sub>, purified through fractional distillation, would be available for transportation to a UF<sub>6</sub>-to-UO<sub>2</sub> conversion facility. The primary obstacle to conversion of UO<sub>3</sub> to UF<sub>6</sub> is that the current domestic facility is not used for conversion of depleted uranium nor is it likely to be used for depleted uranium because of potential contamination of the natural uranium process lines. Additional information on activities regarding conversion alternatives for depleted UF<sub>6</sub> can be found in Vol. 1 of this report.

The dry process is given by the following set of equations





**Fig. 3. Dry uranium to UF<sub>6</sub> conversion process.**

As previously mentioned, there are programmatic objections to converting the depleted UO<sub>3</sub> powder to UF<sub>6</sub>, to remove the impurities as an intermediary step in the conversion to pure UO<sub>2</sub> because of large amounts of depleted UF<sub>6</sub> that already exist and that could be available to the FMDP. However, this is the only known procedure that uses an all-dry process, such as that shown in Fig. 3 in combination with the dry conversion process diagram shown in Fig. 5. An alternative wet process, using a UF<sub>6</sub> intermediary step, is shown in Fig. 4 in combination with the wet conversion portion of Fig. 5.

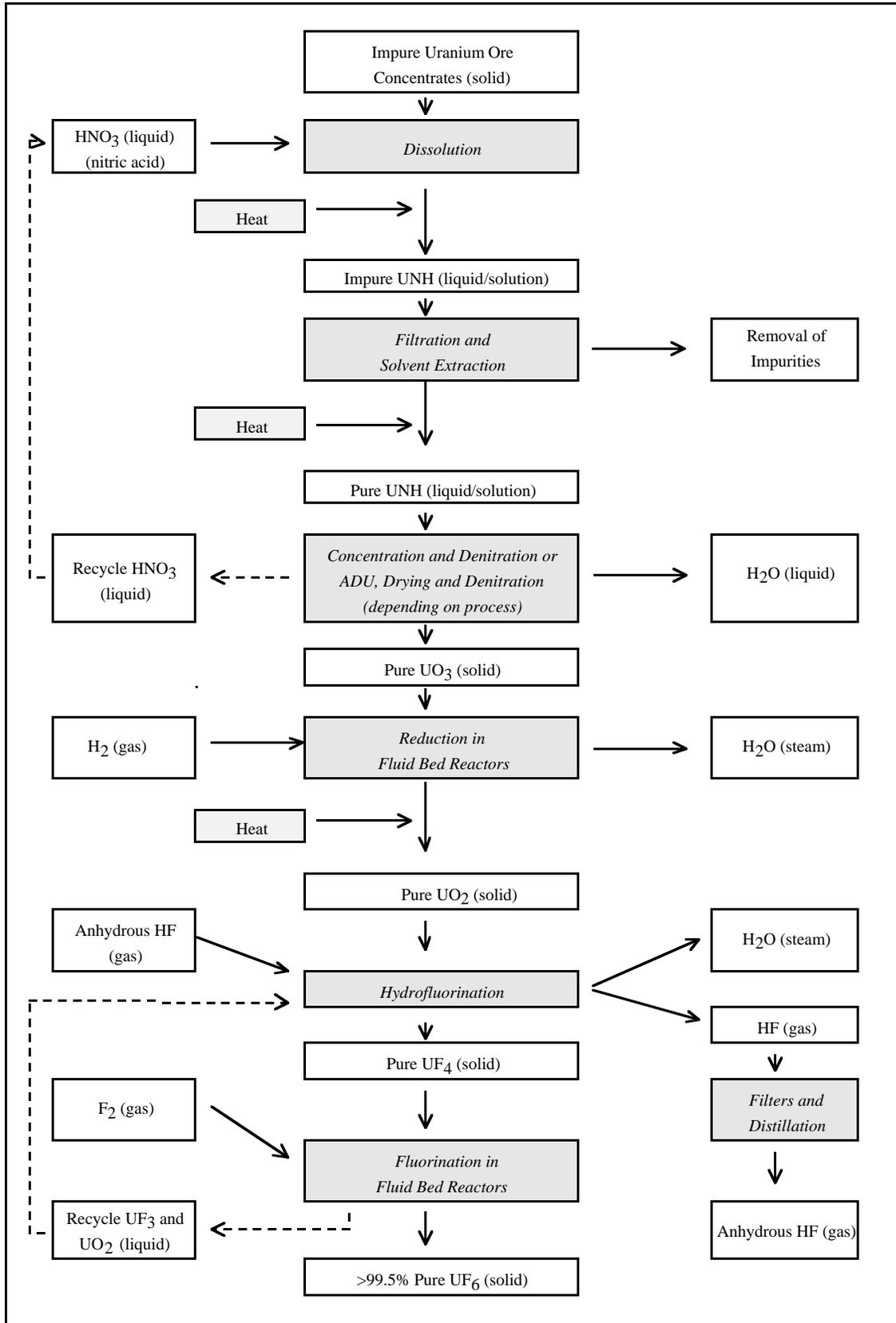


Fig. 4. Wet uranium to  $\text{UF}_6$  conversion process.

Figure 5 provides the process flows for both traditional wet and dry enriched UF<sub>6</sub>-to-UO<sub>2</sub> conversion processes. The dry process, which generates virtually no waste, for conversion of UF<sub>6</sub> to UO<sub>2</sub> has less waste streams than the aqueous process. The only by-product, hydrogen fluoride (HF), is captured and sold as a viable commercial product.

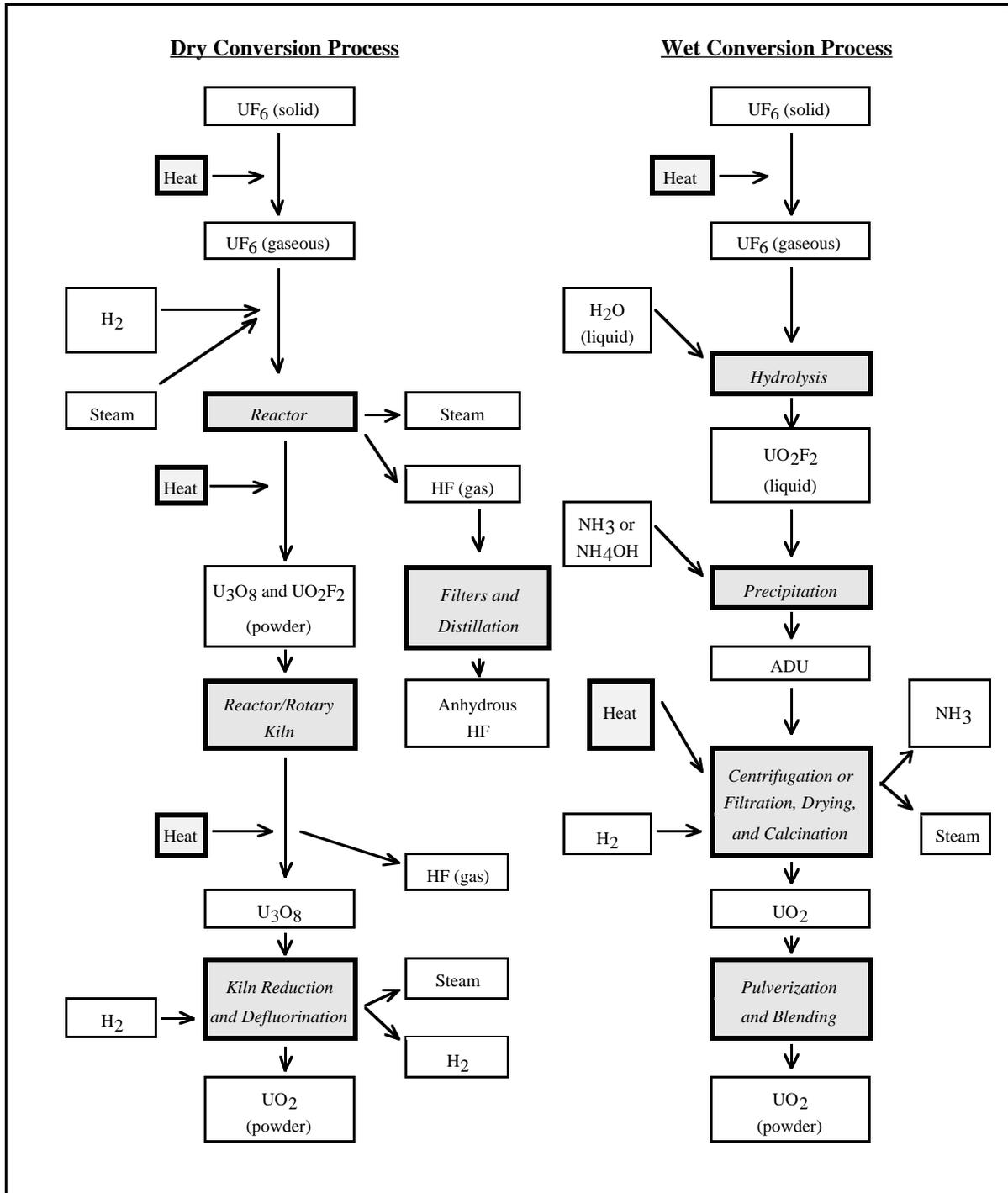
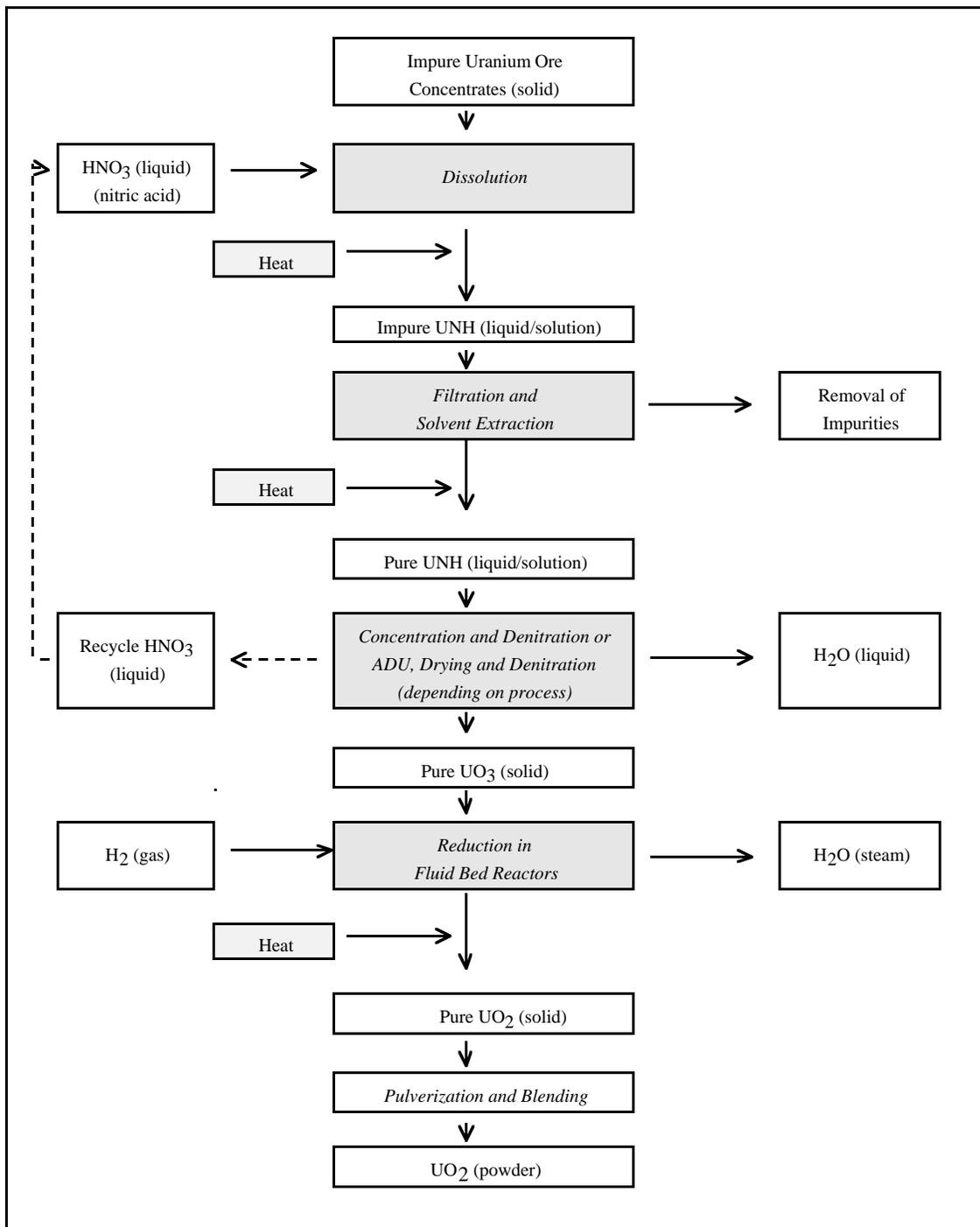


Fig. 5. Dry and wet UF<sub>6</sub>-to-UO<sub>2</sub> conversion processes.

In the  $UF_6$ -to- $UO_2$  dry process, the gaseous  $UF_6$  reacts with superheated steam to form gaseous HF and solid uranyl fluoride ( $UO_2F_2$ ) powder, which is conveyed to the kiln for further defluorination and reduction to triuranium octaoxide ( $U_3O_8$ ). This oxide is further reduced to stoichiometric  $UO_2$  through introduction of hydrogen ( $H_2$ ). Out-of-specification uranium can be sent back through the hydrolysis zone for further processing.

Pure  $UO_2$  could be achieved directly by an aqueous means, as shown schematically in Fig. 6. This would involve introduction into the LEU fuel fabricator's process lines at the appropriate step without going through the  $UF_6$  conversion.



**Fig. 6. Wet uranium to  $UO_2$  conversion process.**

Once converted, the depleted  $\text{UO}_2$  would be available for transportation to the MOX fabrication facility for further processing if the conversion plant is not collocated. The MOX facility does not necessarily require collocation with the depleted  $\text{UF}_6$  conversion plant, which will have a small depleted  $\text{UO}_2$  product storage vault for inventory awaiting transport to the MOX fuel fabrication plant.

Depleted  $\text{UO}_2$  is packaged in a heavy plastic-lined, 55-gal drum that holds up to 880 lb of  $\text{UO}_2$  powder. Depleted  $\text{UO}_2$  is not a fissile material and does not have a packaging limit to prevent criticality. The lining is secured with a wrapping tie and is used to prevent contamination from contact with the drum. Limited storage space for  $\text{UO}_2$  awaiting shipment to the MOX fuel fabrication facility would be available based on commitments to other customers.

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#### 4. RECOMMENDATIONS FOR FMDP

A new technical study would involve an interest search among DOE and commercial uranium facilities to obtain a better understanding of chemical, isotopic, and powder morphology requirements. Planning activities would require close interaction with the commercial sectors interested in providing conversion services and LEU fuel fabrication services for DOE. (Some of these corporate entities may become part of the MOX fabrication/irradiation consortium.) Issues relating to transportation to the conversion and MOX facilities, drum and cylinder handling, feed storage, and waste disposal would need to be resolved in connection with program requirements.

In conclusion, a portion of DOE's surplus depleted  $UO_3$  could be converted to the oxide form of depleted  $UO_2$ . The depleted  $UO_2$  could be used as feed in combination with plutonium for a MOX fuel fabrication facility. However, since the  $UO_3$  would first require conversion to  $UF_6$  to remove impurities, use of DOE's surplus of virtually pure  $UF_6$  would be more cost effective than use of its surplus impure depleted  $UO_3$ . Alternatively, the impure  $UO_3$  could be purified using aqueous means (solvent extraction).

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## 5. REGULATORY REQUIREMENTS

Handling, disposition, packaging, and transportation of drums containing depleted UO<sub>2</sub> must adhere to the following laws, policies, and regulations.

<b>Document</b>	<b>Description</b>
ASME NQA-1	<i>Quality Assurance Program Requirements for Nuclear Facilities</i> , American Society for Mechanical Engineers.
DOE Order 1270.2	<i>Safeguards Agreement with the International Atomic Energy Agency</i> , U.S. Department of Energy, June 1992.
DOE Order 1540.1	<i>Materials Transportation/Traffic Management</i> , U.S. Department of Energy, July 1992.
DOE Order 1540.2	<i>Hazardous Material Packaging for Transport—Administrative Procedures</i> , U.S. Department of Energy, September 1986.
DOE Order 5480.3	<i>Safety Requirements for the Packaging and Transportation of Hazardous Materials, Hazardous Substances, and Hazardous Wastes</i> , U.S. Department of Energy, July 1985.
DOE Order 5630.11	<i>Safeguards and Security Program</i> , U.S. Department of Energy, August 1994.
USPS Pub. 52	<i>Acceptance of Hazardous or Perishable Articles</i> , U.S. Postal Service, Publication 52.
10 CFR	Parts 40.4, 50.34, 70, 71(H), and Appendix B to Part 50, <i>Code of Federal Regulations</i> , U.S. Nuclear Regulatory Commission.
42 USC 2296b-5	<i>Energy Policy Act (EPACT) of 1992</i> , Public Law 102-486, Sect. 1016—Uranium Inventory Study, October 24, 1992.
49 CFR	Parts 173.420, 173.425, 173.474, and 173.475, <i>Code of Federal Regulations</i> , U.S. Department of Transportation, Research and Special Program Administration.

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