STABILIZATION OF DEPLETED URANIUM HEXAFLUORIDE TAILS MATERIAL FOR DISPOSITION

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ABSTRACT

Uranium Disposition Services, LLC (UDS) has been contracted by the U.S. Department of Energy (DOE) to stabilize approximately 700,000 metric tons (MT) of legacy tails material from uranium enrichment activities. To accomplish this mission, UDS will design, construct, and operate two Conversion Facilities: one in Paducah, Kentucky and one in Portsmouth, Ohio. These facilities will convert the DOE inventory of depleted uranium hexafluoride (DUF_6) now located at the Paducah Gaseous Diffusion Plant, the Portsmouth Gaseous Diffusion Plant, and the East Tennessee Technology Park to uranium oxide.

Legacy tails material is considered a potential hazard as it is stored in the volatile DUF_6 form, which is highly reactive when exposed to water including the moisture (or humidity) in air. Additionally, it is stored outside in approximately 60,000 steel cylinders that could potentially corrode to the point where the primary containment is unreliable.

The patented conversion process, based on Framatome ANP proprietary technology, utilizes a fluidized bed conversion unit to convert the DUF_6 to its most benign form, uranium oxides, in a simple one-step operation. This simple process ensures that the Conversion Facility is cost effective and has maximum operability, reliability, and maintainability. The uranium oxide product is nonvolatile and nonreactive. Hydrofluoric acid (HF acid), a byproduct of the conversion process, is recovered thereby eliminating a liquid process effluent stream. The recovered HF acid will be sold to the chemical industry.

Other key elements to minimizing the waste of processing this material are:

- DUF_6 heels (process residue) are typically extracted and treated in alternate equipment. The UDS process will stabilize the heels in-situ, eliminating heel treatment equipment and associated waste streams.
- After processing, the uranium powder (uranium oxides) will be loaded back into modified DUF_6 cylinders (with the heel). This innovative strategy eliminates the need for special cylinder disposal equipment, recurrent operational cost for powder packaging, different types of container handling equipment, etc.
- The offgas treatment chemicals are regenerated for reuse, eliminating this as a liquid waste stream and minimizing the operating cost of treatment chemicals.

INTRODUCTION

This paper will focus on the conversion technology used by UDS to convert DUF_6 to uranium oxides and highlight the key elements that are incorporated in the process design to provide safe, environmentally sound operations and to minimize generation of process wastes.

The technology, design, and operating bases for the DUF_6 Project is based on Framatome ANP's proven commercial process and facilities in operation today that convert uranium hexafluoride (UF_6) to uranium oxide. Figure 1 shows the Framatome ANP Conversion Facilities at Lingen, Germany and Richland, Washington that produce nuclear-grade uranium dioxide (UO_2) for nuclear fuel fabrication. These facilities have demonstrated reliable performance over a combined 19 years of commercial operation.

Fig. 1. Framatome ANP Conversion Facilities

UDS is an integrated team formed by Framatome ANP, Inc., Duratek Federal Services, Inc., and Burns and Roe Enterprises, Inc. for the single purpose of dispositioning DOE's legacy DUF_6 and cylinders. UDS is currently focused on designing and constructing the facilities to convert the DOE's inventory of DUF_6 to uranium oxide in 25 years. See Figure 2 for a computer-generated model of the Paducah Conversion Facility.

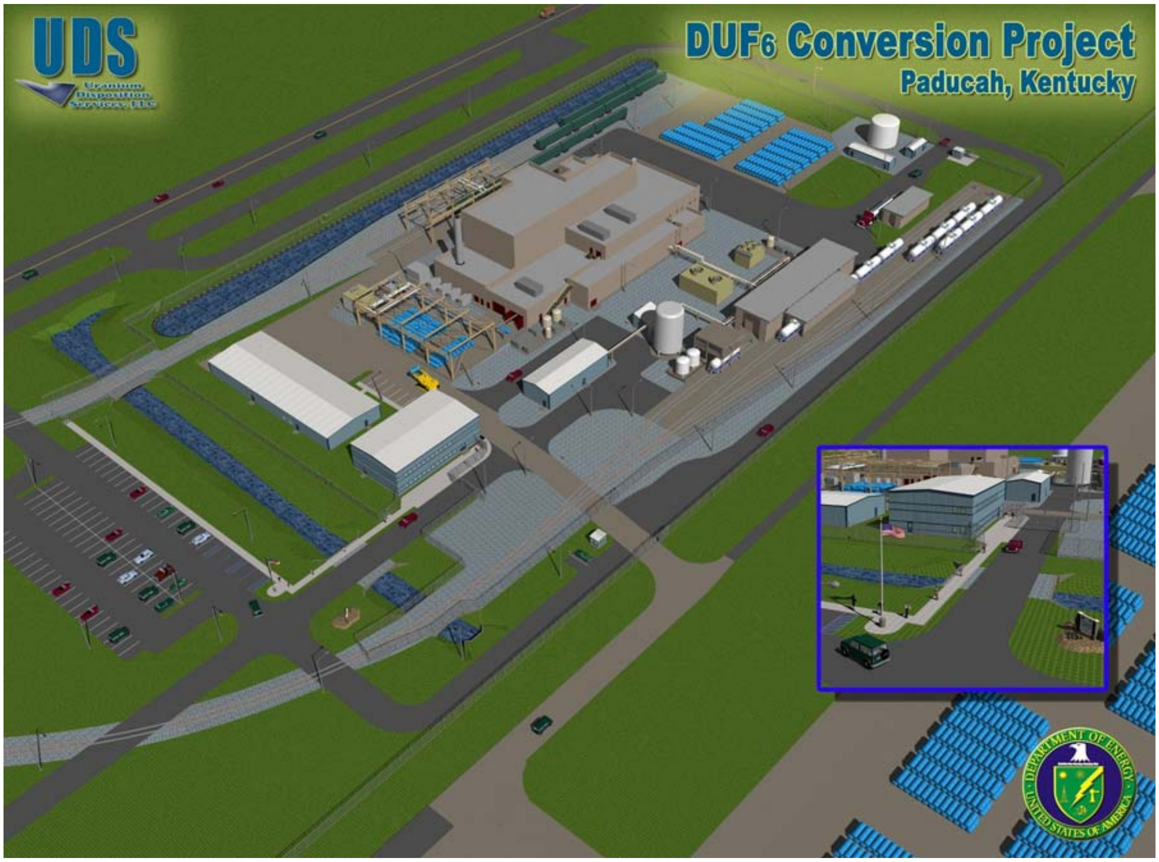

Fig. 2. Paducah Conversion Facility

The DUF_6 Project Schedule is shown in Table I.

Table I. Project Schedule

Contract Award	08/29/02
Start Preliminary Design	08/03
Start Site Work	07/04
Start Conversion Facility Construction	07/05
Physical Construction Complete	12/06
Start of Operations	05/07

DUF_6 CONVERSION PROCESS

A simplified overview of the DUF_6 conversion process is shown in Figure 2. DUF_6 cylinders are heated in autoclaves to vaporize the DUF_6. The DUF_6 along with steam and hydrogen are metered to the conversion unit where the DUF_6 is reacted at elevated temperatures with steam to form uranyl fluoride (UO_2F_2) and hydrogen fluoride (HF) vapor. The UO_2F_2 is then defluorinated and densified in the fluidized bed in a reducing atmosphere to convert the DUF_6 to uranium oxides.

The offgas from the conversion unit containing HF vapor, excess steam, nitrogen, and hydrogen is cooled and scrubbed to recover HF acid. The condensed HF acid is transferred from HF storage/loadout tanks to vendor railcars for shipment to existing markets.

The uranium oxide product from the conversion unit is transferred to the uranium oxide hoppers. The hoppers are used to fill modified DUF_6 cylinders with uranium oxides, and then the cylinders are loaded onto railcars and transported to a long-term storage/disposal site.

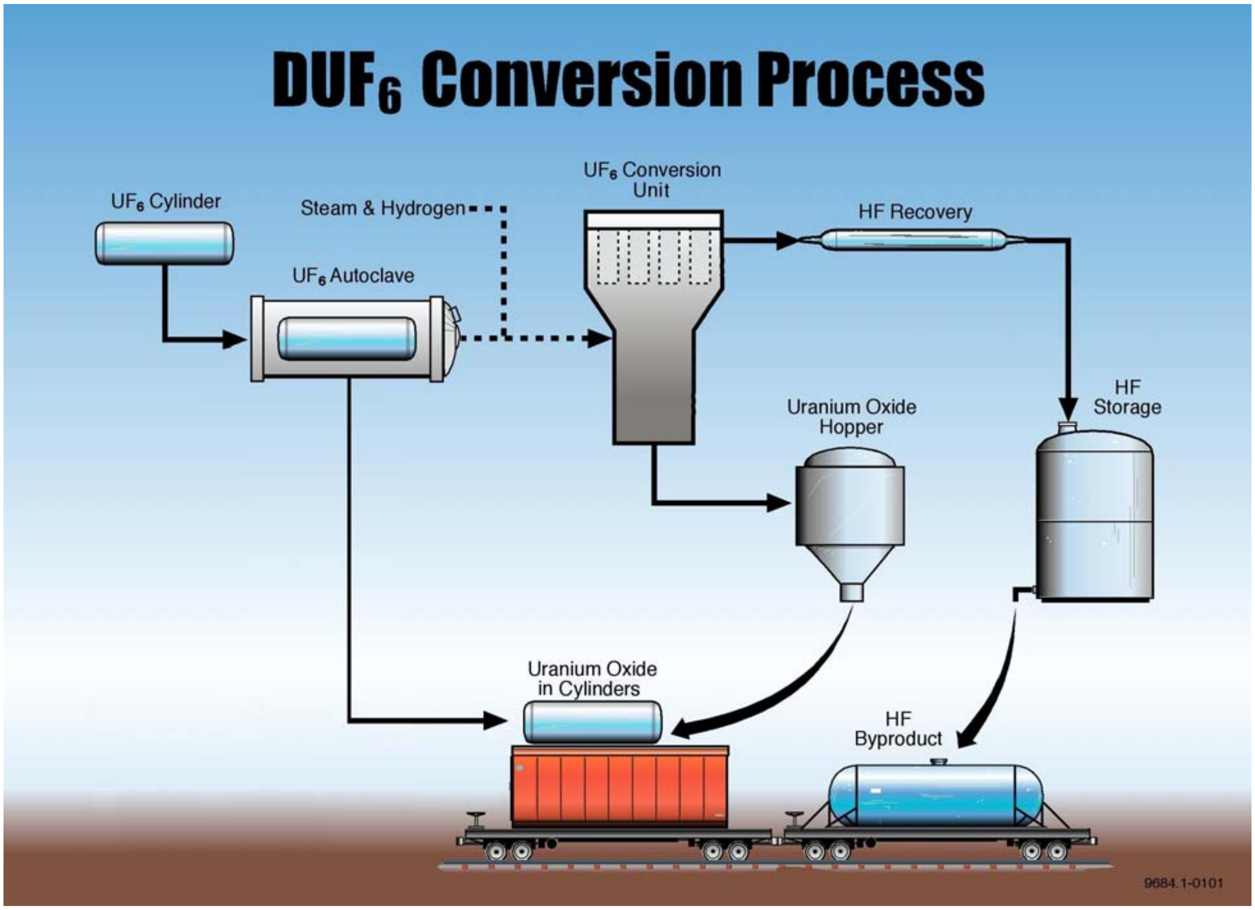

Fig. 3. DUF_6 Conversion Process Overview

CONVERSION FACILITY

The facility is designed to convert DUF_6 to uranium oxides and to recover the HF acid byproduct. The nominal design capacity for the Paducah facility is 18,000 MT/yr of DUF_6 operating 24 hours per day, 7 days per week. High integrity process equipment and piping will provide primary containment. The facility will be a concrete building (walls and roof) that has a controlled ventilation system and utilizes airlocks. The heating, ventilation, and air conditioning (HVAC) system will process all the building and process offgas ventilation through prefilters and high efficiency particulate air (HEPA) filters prior to venting to the facility vent stack. The stack is monitored for uranium and HF emissions.

The facility contains identical conversion lines (modules) that include vaporization, conversion, HF recovery, and process offgas scrubbing systems. In addition, oxide powder handling systems, cylinder stabilization, and a cylinder modification system are housed in the building (see Figure 3).

HF storage and potassium hydroxide (KOH) regeneration systems are also provided but are not contained in the Conversion Facility. Utility support systems required by the process include steam, nitrogen, hydrogen, and cooling water.

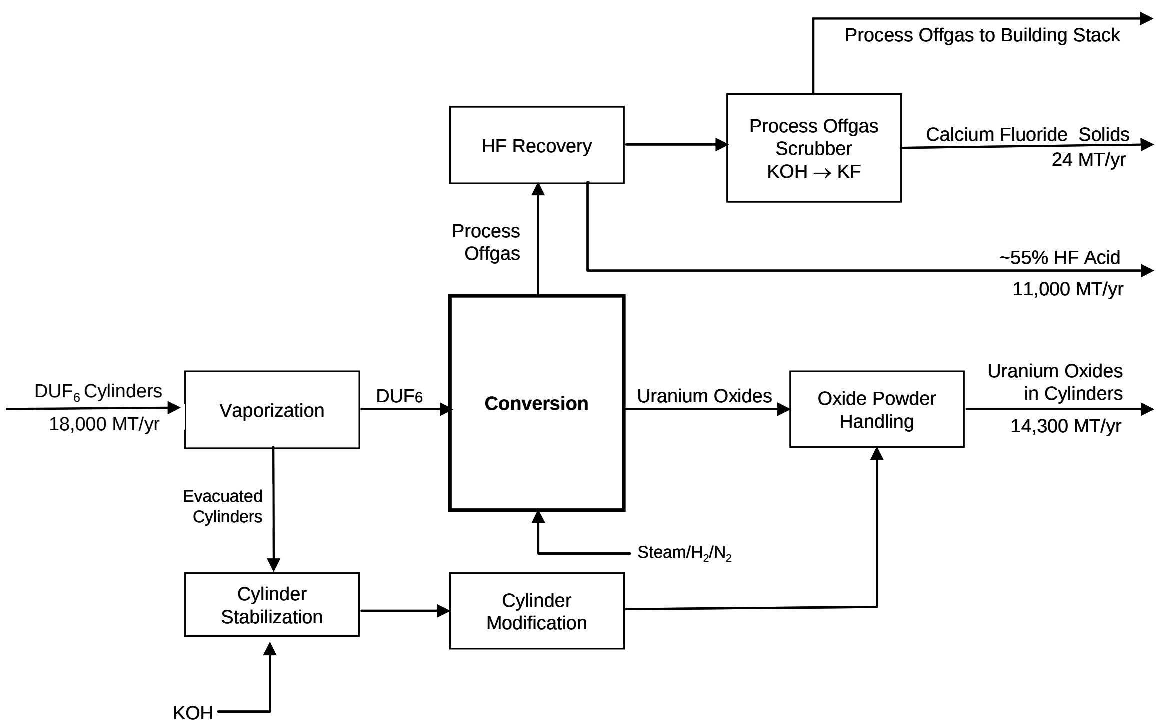

Fig. 4. Paducah Conversion Facility Material Flow Diagram

DUF_6 Vaporization

Each DUF_6 cylinder will undergo nondestructive analysis verification (to verify that it is not enriched material) before the cylinders are moved into the Conversion Facility and placed into an autoclave. There are two autoclaves per conversion line, plus two additional autoclaves for special handling of cylinders that may require special processing conditions (not suitable for normal vaporization temperatures). One autoclave will feed the conversion process, while the other autoclave is being emptied, loaded, cooled down, or heated up. All autoclaves are designed to handle 48- and 30-inch cylinders and have identical safety features. Connections are made between the cylinder and the DUF_6 piping within the autoclave. The autoclave door is then shut and sealed. The cylinder valve is opened remotely and the cylinder is heated until all of the solid DUF_6 has been liquefied (DUF_6 is a solid a room temperature). DUF_6 vapor is drawn through a flow control manifold (hotbox) and directed to two conversion units. The autoclave serves a dual purpose: 1) it heats the cylinder to temperature, and 2) it provides secondary containment if the primary containment (cylinder) is breached. A central control system monitors temperature, pressure, and weight of the DUF_6 cylinder, and automatically takes steps to stop DUF_6 flow in case of any abnormal conditions. The control system also shuts off the heat in case of elevated temperature and/or pressure.

The autoclaves are American Society of Mechanical Engineers (ASME) standard pressure vessels designed to provide containment of DUF_6 and HF from a ruptured DUF_6 cylinder.

A hotbox is used to provide secondary containment where flanged connections are required (DUF_6 flow control valves and flow elements). The hotbox is vented to a HEPA filter. The vent line includes an in-line monitor to detect DUF_6 leaks. Any leaks to the secondary containment or to the room will activate safety interlocks to stop DUF_6 flow from the autoclave and minimize releases to the room or the ventilation system. In the event of a power or instrument air failure, all valving and the control system default to the safest state.

DUF_6 to Uranium Oxide Conversion

DUF_6 vapor, steam, nitrogen, and hydrogen are metered into electrically heated conversion units where the DUF_6 is converted to uranium oxides and HF vapor. There are two conversion units and two backup filter vessels per process line. The uranium oxides are discharged out the bottom of each conversion unit and are then pneumatically transported to the oxide powder handling system. The offgas is filtered through sintered metal filters at the top of the conversion unit, passed through a second set of sintered metal filters (backup filter vessel), and is then routed to the HF recovery system. Essentially all of the uranium oxide powder is retained in the conversion unit and transferred to the uranium oxide handling system. The backup filter vessel provides a redundant barrier to assure no uranium carryover to the HF recovery system in case of a filter failure in the conversion unit.

Safety features for the conversion process area include primary containment of all process lines/equipment, room UO_2F_2 detectors, and the backup filter vessel. Automatic interlocks are incorporated into the design that will shutdown flow of DUF_6 to the conversion unit (at the autoclave) in case of any upset process conditions or DUF_6 leak detection. Any leaks to the room will activate the safety interlocks to stop the flow of DUF_6 from the autoclave and minimize releases to the room or the ventilation system.

In the event of a power or instrument air failure, all valving and the control system default to the safest state.

Oxide Powder Handling

The oxide powder handling system transfers uranium oxides from the conversion unit, collects it in oxide hoppers, and loads it into modified DUF_6 cylinders. Each of the two oxide hoppers can hold roughly the mass equivalent of two DUF_6 cylinders filled with uranium oxides.

The uranium oxides exit the conversion unit through a rotary valve and are gravity fed to a powder transfer station. The uranium oxides are cooled in a heat exchanger before they are pneumatically transferred (air at slightly below atmospheric pressure) to receiving vessels located above the oxide hopper. The uranium oxides from the receiving vessel are discharged into the oxide hopper. The uranium oxides in the oxide hopper are discharged through a rotary valve into a modified DUF_6 cylinder. The powder transfer station is enclosed within a hood to prevent uranium oxides from escaping the process equipment. An enclosure also surrounds the connection of the process equipment and the modified DUF_6 cylinder. The hoods are vented to a prefilter and a HEPA filter, and then routed to the main HEPA filter bank.

HF Recovery

There are two deionized water scrubbers, two HF condensers, and two HF receiver tanks per conversion line for recovery of HF acid.

The HF vapor/steam/nitrogen/hydrogen from the backup filter vessel is vented (slightly below atmospheric pressure) to the HF condenser. As the HF vapor and steam are condensed, the liquid HF acid flows by gravity to the HF receiver tank. Piping and equipment, which is made from corrosion resistance material, provides primary containment of the HF, either gaseous or liquid, and prevents escape to the environment. The temperature of the inlet and outlet of the process offgas is monitored. The HF acid recovered from the system is pumped to HF storage tanks.

Process Offgas Scrubber

The process offgas scrubber system is used to remove the HF that remains in the process offgas after the HF recovery system has recovered greater than 99% of the HF acid. The system is designed to meet environmental discharge requirements. From the HF recovery system, a blower routes the offgas through a KOH scrubber where the residual HF vapor reacts with KOH solution to produce potassium fluoride (KF) and water.

The offgas from the scrubber is routed to the Conversion Facility HVAC final HEPA filter bank and released through the building exhaust. Periodic transfer of the spent scrubber solution batches to the KOH regeneration system and replacement of this solution with regenerated scrubber solution are controlled automatically.

The process offgas scrubber systems are operated below atmospheric pressure and include redundancy for reliability.

KOH Regeneration

The function of the KOH regeneration system is to regenerate spent scrubber solution for the KOH scrubbers in the process offgas scrubber system. The KOH regeneration system removes fluoride from spent scrubber solution and converts it into calcium fluoride (CaF_2) solids. The CaF_2 solids are dewatered for loadout and the regenerated scrubber solution is recycled to the KOH scrubbers.

Cylinder Stabilization

Cylinders that have been processed by the vaporization system are expected to have small quantities of DUF_6 in the heel after being processed. Stabilization is necessary to ensure that cylinder heels do not contain reactive or corrosive materials in excess of waste disposal acceptance criteria. Based on the maximum expected heel quantity, a standard amount of aqueous stabilizing solution consisting of KOH and water is injected into the cylinder. The cylinder is rotated to allow the stabilizing solution to coat the interior. The residual DUF_6, and any remaining reactive intermediate uranium/fluoride compounds, react with the water and KOH in the stabilizing solution to form UO_2F_2 and KF.

Cylinders will be aged until the cylinder activity level (from Thorium-234) has been reduced to an acceptable level. After aging, cylinders are returned to the Conversion Facility for modification in the cylinder modification system.

Cylinder Modification

The DUF_6 cylinders that have been processed through the vaporization system and aged are transferred to the cylinder modification system for modification. Automated equipment is used to remove a circular section of the valve end of the cylinder and to weld a flange to accommodate addition of uranium oxides. The modified cylinder is then transferred to the oxide powder handling system to be filled with uranium oxides.

CONCLUSION

In approximately 50 years of enrichment activities, nearly 700,000 MT of DUF_6 has been produced at three sites (Oak Ridge, Paducah, and Portsmouth). This legacy DUF_6 is volatile and highly reactive posing a safety risk, which DOE is committed to manage. UDS, in accordance with the DOE's management strategy, is building two Conversion Facilities to convert this legacy material to uranium oxides – a much more stable compound for disposition at a long-term storage facility.

The UDS technology is based on currently operating commercial fuel fabrication technologies with innovative strategies to minimize plant waste/effluent. Key examples are stabilizing the heel material in-situ, reuse of the cylinders as packaging for the uranium oxide, and regenerating the offgas treatment chemicals for reuse.

UDS is highly committed to the safety of the DUF_6 conversion facilities. Key elements of safety for DUF_6 processing are high integrity primary containment, secondary containment around flanges and fittings in DUF_6 piping, failsafe valving, instrumentation and control, leak detection and interlocks, HEPA filtration, and radiation monitoring.