DEPLETED URANIUM HEXAFLUORIDE MANAGEMENT PROGRAM

The Engineering Analysis Report for the Long-Term Management of Depleted Uranium Hexafluoride

J.W. Dubrin, J.N. Zoller, and L. Rahm-Crites
Lawrence Livermore National Laboratory

T.H. Bertell, Jr., S. Cavanaugh, M. Coflin, S. Coflin, C.J. Crockett, G. DeMoss, M. F
C. Herrington, P. Hogroian, C.L. Johnson, R.A. Kelly, T. Miller, D.C. Riley
Science Applications International Corporation

T. Breitmayer, R. A. Kaiser, B. Marais, M. Pong, D.R. Tiojanco, and D.B. Warren
Bechtel

T. Joseph, D. Milewski, and J.N. Sumner
Lockheed Martin Energy Systems

May 1997

Lawrence Livermore National Laboratory
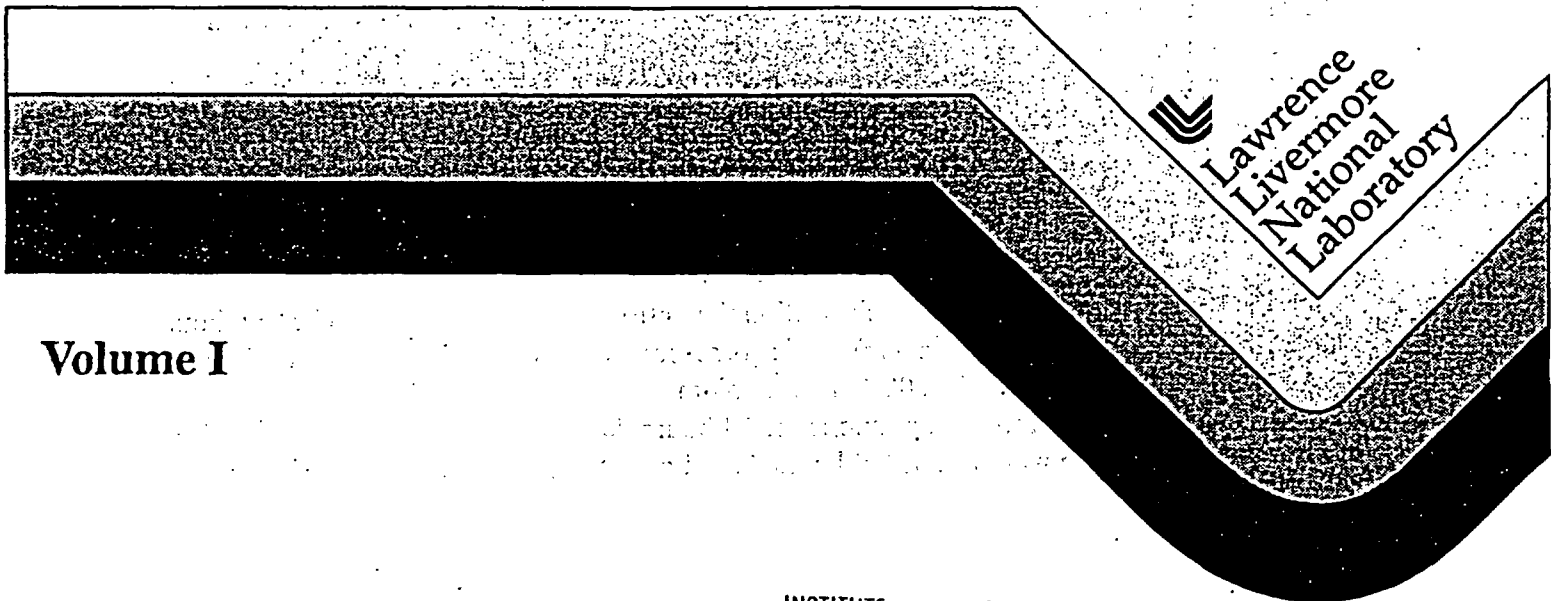
Volume I

2.5 Assumptions

For the purpose of developing engineering data, assumptions were made regarding throughput, isotopic composition, bulk density, operational availability, scheduling, packaging of materials for transportation, and lag storage of materials. These assumptions are stated in each Engineering Data Input Report and are somewhat dependent upon the option or suboption being analyzed. Following are some of the major assumptions used in the reports.

- The depleted uranium is assumed to be chemically pure, with an average isotopic composition of 0.001 percent U-234, 0.25 percent U-235, and 99.75 percent U-238 and a corresponding specific activity (alpha) of 4×10^{-7} curies per gram (Ci/g) depleted uranium (one curie equals 3.7×10^{10} nuclear disintegrations per second). In the filled UF_6 cylinders, the short-lived daughter products of U-238, thorium (Th)-234 and protactinium (Pa)-234, are in the same equilibrium with the U-238; therefore, these beta emitters each have the same activity as U-238 (i.e., 3.3×10^{-7} Ci/g).

- It is assumed that the depleted UF_6 will be transported in 14-ton cylinders like those currently used for storage and that emptied cylinders will be shipped off site for treatment, disposal, or use. Facilities provide three months' onsite storage for outgoing emptied cylinders (to allow for the decay of radioactive daughter products in the heel).

- U_3O_8 will have a final bulk density of 3.0 g/cubic centimeter (cc) and will be transported in 55-gallon (208-liter) drums.

- UO_2, in sintered pellets (0.82 in. x 0.82 in.) or microspheres (1200 micron and 300 micron), will have a final density of about 10 g/cc and will be packaged for transportation in 30-gallon (113-liter) drums.

- Metal derbies from the batch reduction process will be about 20 in. in diameter and 6.7 in. high and will be packaged in wooden boxes.

- Metal billets from the continuous reduction process will be 2 in. x 3 in. x 20 in. and will be packaged in boxes.

- It is assumed that wastes will be compacted and shipped off site for treatment and disposal. Hazardous wastes will be hauled to a commercial waste facility for treatment and disposal according to Environmental Protection Agency (EPA) Resource Conservation and Recovery Act (RCRA) guidelines.

- Depleted uranium, including depleted uranium waste forms, is subject to regulation by the Atomic Energy Act (AEA), rather than by RCRA.

- A period of 20 years is assumed to disposition the entire depleted uranium stockpile (about 560,000 MT UF_6 in 46,422 cylinders). This corresponds to an annual throughput rate of 28,000 MT of UF_6 or about 19,000 MT of uranium.

- Operations are assumed to be continuous for 24 hours/day, seven days/week, 52 weeks/year; annual operating time would be 7,000 hours, based on a plant availability factor of 0.8.

do not currently meet Department of Transportation (DOT) requirements for offsite shipment. Options for preparing cylinders for shipment are analyzed in Sections 6.1 and 6.2 of the EAR. Preparation of nonconforming cylinders for shipment is also addressed.

2.8.1.2 Use

Like conversion facilities, manufacturing facilities could potentially have to comply with RCRA storage and permitting requirements if adequate offsite treatment and disposal options for hazardous or mixed waste are not available.

2.8.1.3 Storage

Licensing under the AEA for private, long-term storage facilities is unprecedented and inherently controversial. Convincing regulators that storage options do not constitute disposal, especially in the case of the below ground vault or mine options, could be a major regulatory compliance issue. This would require extensive negotiations and demonstration that there is a defined term for storage and likely use of the material at the end of the storage period.

NEPA compliance represents a potential major issue because of the likelihood of controversy. Previous DOE NEPA documents on long-term storage facilities have often taken much longer than anticipated and have sometimes resulted in litigation. Options involving vault and mine storage may be perceived by both regulators and the public as disposal. Site-specific EISs for these options would take longer to develop (between three and six years) than a typical EIS.

Like the conversion options, storage as depleted UF_6 could present major regulatory compliance issues related to the shipment of existing cylinders. Many of these cylinders currently do not meet DOT requirements for offsite shipment.

2.8.1.4 Disposal

The licensing of new low-level waste (LLW) disposal facilities under the AEA would be a major compliance issue. Licensing under the AEA by NRC or authorized states may be difficult due to the extensive regulatory requirements and the inherently controversial nature of the subject. Approvals under the AEA by DOE for new LLW disposal facilities may be difficult due to extensive performance assessment requirements. Disposal facilities could potentially be required to comply with RCRA storage and permitting requirements if offsite treatment and disposal options for mixed waste continue to be limited.

3.0 Summary of Options Analyzed in Depth

As stated in Section 1.4.1, the Engineering Analysis Project developed the engineering data for representative options which were determined to be feasible in *The Technology Assessment Report for the Long-Term Management of Depleted Uranium Hexafluoride*. The feasible recommendations fell into four broad categories—conversion, use, storage, and disposal—which, along with transportation, comprise the five "modules" or building blocks for constructing management strategy alternatives. The options that were analyzed in depth are summarized here. The complete data for these options are contained in Section 6, where the individual Engineering Data Input Reports are found. Other options which were considered but not analyzed in depth are summarized in Section 4.

3.1 Transportation Module

This element includes options for cylinder preparation, emptied cylinder disposition, and transport. Transport of all forms of depleted uranium by both truck and rail is included in the individual Engineering Data Input Reports for the various conversion, use, storage, and disposal options. No specific transportation technologies were described in the responses to the Request for Recommendations.

3.1.1 Cylinder Preparation Option

This element refers to the preparation of the depleted UF_6 cylinders at their current storage sites for transportation to an offsite facility, generally for conversion. A number of the cylinders currently do not meet Department of Transportation (DOT) requirements for offsite shipment. The cylinder problems are of three types: (1) overfilled cylinders, (2) overpressured cylinders, and (3) substandard cylinders (e.g., cylinders with below the minimum value wall thickness or other characteristics that render them unsafe or unserviceable according to ANSI N14.1).[19] There are no definitive data on the number of cylinders affected by any of these problems, so the basis for the engineering analysis is empirical data provided by site personnel. It is anticipated that these estimates may be revised as the issues are further examined, including additional cylinder data. It should be noted that these cylinder conditions are problems only for offsite transportation and do not restrict onsite transport or storage.
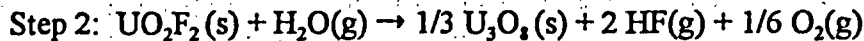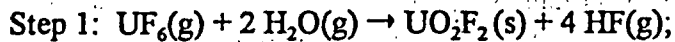
In accordance with the 49 CFR 173.420(a)(4) transportation requirements for UF_6, the volume of solid depleted uranium hexafluoride at 20°C (68°F) may not exceed 62 percent of the certified volumetric capacity of the packaging. Overfilled cylinders are those in which the amount of

---

[19]American National Standards Institute. ANSI N14.1-1995, *American National Standard for Nuclear Materials - Uranium Hexafluoride - Packaging for Transport*. December 1, 1995.
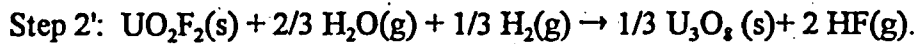
scale industrial basis by Cogema in France for the defluorination of depleted UF_6. The conversion process involves two steps. In the first, exothermic, step, the gaseous UF_6 is hydrolyzed with steam to produce solid uranyl fluoride (UO_2F_2) and HF. In the second, highly endothermic, step, the UO_2F_2 is pyrohydrolyzed with superheated steam (optionally containing H_2) to U_3O_8 and additional HF.

The reactions are as follows:

Step 1: $UF_6(g) + 2 H_2O(g) \rightarrow UO_2F_2(s) + 4 HF(g)$;

Step 2: $UO_2F_2(s) + H_2O(g) \rightarrow 1/3\ U_3O_8(s) + 2 HF(g) + 1/6\ O_2(g)$

or

Step 2': $UO_2F_2(s) + 2/3\ H_2O(g) + 1/3\ H_2(g) \rightarrow 1/3\ U_3O_8(s) + 2 HF(g)$.

Due to the large excess steam requirements for the second step, concentrated HF (typically 70 percent HF - 30 percent H_2O) is the direct process by-product. The U_3O_8 would be compacted to achieve a bulk density of about 3.0 g/cc prior to storage or disposal.

As indicated, the technology feasibility for the large scale conversion of UF_6 to U_3O_8 is well established. For the engineering analysis, there are scaling uncertainties, including residency times, associated with the conversion reactors. These and the uncertainties in materials of construction and the optimal operating conditions would be resolved in a subsequent engineering development phase of the Program. Although anhydrous HF is not produced as the by-product from the Cogema facility, distillation (the assumed process to upgrade the aqueous HF) is well established. Again, any uncertainties with the specific distillation process and its integration assumed for the engineering analysis (see 3.2.1.1) would be addressed in a subsequent engineering development phase of the Program.

Two suboptions were developed in the Engineering Analysis Project for the dry conversion of UF_6 to U_3O_8. The first process upgrades the concentrated HF to anhydrous HF (AHF < 1 percent H_2O) for sale with unrestricted usage, based on the very low uranium contamination level. The second process neutralizes the HF to calcium fluoride (CaF_2) for sale or disposal. In addition to several technologies recommended by industry, the U.S. Nuclear Regulatory Commission and the State of Tennessee Department of Environment and Conservation made general recommendations for conversion to U_3O_8.

It is considered unlikely that the presence of trace amounts of uranium would prevent the AHF from being made available for unrestricted use, and even more unlikely that this would prevent its being recycled in the nuclear fuel industry for the conversion of yellowcake (concentrated U_3O_8)

endothermic reaction to UO_2 powder in the second reactor by a mixture of steam and a stoichiometric quantity of hydrogen. The reactions are as follows:

Step 1: $UF_6(g) + 2H_2O(g) \rightarrow UO_2F_2(s) + 4HF(g)$;

Step 2: $UO_2F_2(s) + H_2(g) \rightarrow UO_2(s) + 2HF(g)$.
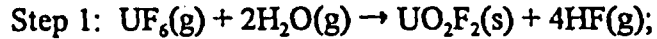
After standard physical treatment operations (milling, compacting, and screening) and the addition of a dry lubricant, the UO_2 powder is pressed into pellets with a density of about 50 percent of theoretical. The pellets are sintered in furnaces with a hydrogen-reducing atmosphere to achieve an assumed density of about 90 percent of the theoretical density. The HF is then upgraded to AHF as described in Section 3.2.1.1.

Due to the fact that the oxide throughput is an order of magnitude higher than that for nuclear fuel fabrication plants, the preconceptual design assumes much larger sintering furnaces than those used in commercial fuel fabrication plants. Furnaces of this size and with these performance specifications are not presently available, but furnaces with one or two of the features (high capacity, high temperature, and special gas atmosphere) are common. It is believed that sintering furnaces combining all of these features can be engineered and fabricated with moderate risks.

The Engineering Data Input Report for this suboption is located in Section 6.6.

3.2.2.2 Dry Process with HF Neutralization Suboption

The only difference between this suboption and the one described in Section 3.2.2.1 is the neutralization of the HF acid by-product. The neutralization step is the same as that described in Section 3.2.1.2.

Due to the fact that the oxide throughput is an order of magnitude higher than that for nuclear fuel fabrication plants, the preconceptual design assumes much larger sintering furnaces than those used in commercial fuel fabrication plants. Furnaces of this size and with these performance specifications are not presently available, but furnaces with one or two of the features (high capacity, high temperature, and special gas atmosphere) are common. It is believed that sintering furnaces combining all of these features can be engineered and fabricated with moderate risks.

The Engineering Data Input Report for this suboption is located in Section 6.7.

4.2   HF DISTILLATION

The hot off-gas and vapor from the reactors flow to a distillation column for HF purification and recovery. The system is shown in Figure 4-5.

The vapor is first quenched with distillation column bottoms to cool the superheated vapor to its saturation temperature. The saturated vapor is fed to the column, which produces an anhydrous HF overhead product at 67°F containing about 200 ppm water, and a 45 wt% HF bottoms stream at 230°F that is recycled to Reactor No. 1.

The overhead product is condensed in a chilled water condenser at 40°F, collected and sampled. Upon satisfactory analysis, the HF is pumped through an underground pipeline to storage tanks in the HF storage building. The HF is then loaded into railroad tank cars or tank trucks for shipment to customers.

The noncondensible gases, primarily oxygen, pass through a -20°F refrigerated condenser to recover additional HF. The gases then flow to the HF scrubbing system.

Major process equipment for the HF distillation process includes a 4 ft-six in diameter by 23 ft high Monel distillation column, equipped with a 2 ft diameter by 6 ft high shell and tube reboiler, a 4 ft diameter by 12 ft long 3,500 ft^2 40°F condenser, a 2 ft diameter by 6 ft long 150 ft^2 -20°F condenser, and two 6 ft diameter by 14 ft long 3,000 gallon hold tanks equipped with cooling coils, and six 12 ft diameter by 45 ft long 38,000 gallon storage tanks.

4.3   HF SCRUBBING SYSTEM

Off-gas from HF Distillation is treated in a scrubber to reduce atmospheric releases of HF to acceptable levels. The HF is recovered and converted to solid CaF_2 product for sale. The system is shown is Figure 4-6.

The off-gas enters a packed column, where it is contacted with a potassium hydroxide (KOH) scrub solution. The HF is removed by the reaction $HF + KOH \longrightarrow KF + H_2O$. The spent scrub solution is collected in a precipitation tank, where hydrated lime is added to remove the fluoride and regenerate the KOH by the reaction $2KF + Ca(OH)_2 \longrightarrow CaF_2 + 2KOH$. A minimum level of KF is maintained in the scrub solution by adding less then the stoichiometric quantity of lime. This ensures all the lime reacts, which keeps solid lime out of the CaF_2 product and the packed bed scrubber.

The scrub solution slurry is filtered in a rotary drum vacuum filter to remove the solid CaF_2 precipitate. The CaF_2 is washed with water to remove impurities and dried in a steam-heated rotary dryer. After cooling, the CaF_2 is packaged in drums and sent to the warehouse for storage. The KOH filtrate and spent wash water are collected, and a side stream is withdrawn and evaporated to remove the water formed by the scrubber chemical reaction and

Table 7-1, Annual Air Emissions During Operation

Pollutants	Principal Release Point	Annual Emissions (lb)
CRITERIA POLLUTANTS		Boiler/Other[1]
Sulfur Dioxide	Boiler Stack / Grade	70/50
Nitrogen Dioxide	Boiler Stack / Grade	9,600/380
Hydrocarbons	Boiler Stack / Grade	200/320
Carbon Monoxide	Boiler Stack / Grade	4,800/2,500
Particulate Matter PM-10	Boiler Stack / Grade	360/80
OTHER POLLUTANTS		
HF	Process Bldg. Stack	900
U_3O_8	Process Bldg. Stack	3.3
Copolymers	Cooling Tower	1,100
Phosphonates	Cooling Tower	110
Phosphates	Cooling Tower	110
Calcium	Cooling Tower	1,900
Magnesium	Cooling Tower	500
Sodium and Potassium	Cooling Tower	200
Chloride	Cooling Tower	350
Dissolved Solids	Cooling Tower	10,500

[1] Other sources are diesel generator and vehicles

Table 7-2, Annual Radiological Emissions During Operation

Radiological Isotope	Principal Release Point	Release Rate (Ci/yr)[1]
Depleted Uranium in Gaseous Effluent	Process Bldg. Stack	5.0×10^{-4}
Depleted Uranium in Liquid Effluent Stream	Effluent Outfall	1.0×10^{-3}

[1] Based on an assumed activity of 4×10^{-7} Ci/g of depleted uranium - see Section 1.2.1 for isotopic composition

a heat exchanger for cooling and is recirculated to the top of the absorber column. A portion of the circulating liquid is continuously withdrawn and discharged to the HF neutralization system.

The vapor leaving the first column flows to the second column for additional HF removal. Fresh water is added to the second column to make up for the liquid that was discharged to the first column.

Major process equipment for the HF absorption process includes a 4 ft diameter by 21 ft high Monel absorption column, a 3 ft diameter by 8 ft long 1,200 ft^2 Monel cooler, a 2 ft-6 in. diameter by 19 ft high Monel absorption column, a 1 ft-6 in. diameter by 8 ft long 300 ft^2 Monel cooler, and associated circulation pumps.

4.3   HF SCRUBBING SYSTEM

Off-gas from HF Absorption is treated in a scrubber to reduce atmospheric releases of HF to acceptable levels. The system is shown in Figure 4-6.

The off-gas enters a packed column, where it is contacted with a potassium hydroxide (KOH) scrub solution. The HF is removed by the reaction HF + KOH ---> KF + H_2O. The spent scrub solution is collected in a precipitation tank, where hydrated lime is added to remove the fluoride and regenerate the KOH by the reaction $2KF + Ca(OH)_2$ ---> $CaF_2 + 2KOH$. A minimum level of KF is maintained in the scrub solution by adding less than the stoichiometric quantity of lime. This ensures all the lime reacts, which keeps solid lime out of the CaF_2 product and the packed bed scrubber.

The scrub solution slurry is filtered in a rotary drum vacuum filter or pressure filter to remove the solid CaF_2 precipitate. The CaF_2 is washed with water to remove impurities, and transferred to the CaF_2 dryer in the HF neutralization system.

The KOH filtrate and spent wash water are collected, and a side stream is withdrawn and evaporated to remove the water formed by the scrubber chemical reaction and the water added for CaF_2 washing. The scrub solution is then cooled and pumped back to the scrubber. The treated off-gas is filtered and discharged to atmosphere.

Major equipment includes a 2 ft diameter by 10 ft high Monel HF scrubber with plastic packing, 7 ft diameter by 7 ft high 2,000 gallon Monel precipitation and filtrate tanks, a 6 ft diameter by 8 ft long Monel rotary drum filter, a 1 ft-6 in. diameter by 4 ft Monel evaporator/condenser unit, and associated tanks and pumps.

4.4   HF NEUTRALIZATION SYSTEM

The 20% HF solution from absorption is neutralized with slaked lime to form CaF_2. The system is shown in Figure 4-7.

Pebble lime (CaO) is mixed with water and milled in a vertical attritor to

Table 7-1, Annual Air Emissions During Operation

Pollutants	Principal Release Point	Annual Emissions (lb)
CRITERIA POLLUTANTS		Boiler/Other[1]
Sulfur Dioxide	Boiler Stack / Grade	60/50
Nitrogen Dioxide	Boiler Stack / Grade	8,300/380
Hydrocarbons	Boiler Stack / Grade	180/310
Carbon Monoxide	Boiler Stack / Grade	4,100/2,500
Particulate Matter PM-10	Boiler Stack / Grade	310/80
OTHER POLLUTANTS		
HF	Process Bldg. Stack	300
U_3O_8	Process Bldg. Stack	3.3
Copolymers	Cooling Tower	1,600
Phosphonates	Cooling Tower	150
Phosphates	Cooling Tower	150
Calcium	Cooling Tower	2,700
Magnesium	Cooling Tower	700
Sodium and Potassium	Cooling Tower	280
Chloride	Cooling Tower	500
Dissolved Solids	Cooling Tower	14,900

[1] Other sources are diesel generator and vehicles

Table 7-2, Annual Radiological Emissions During Operation

Radiological Isotope	Principal Release Point	Release Rate (Ci/yr)[1]
Depleted Uranium in Gaseous Effluent	Process Bldg. Stack	5.0×10^{-4}
Depleted Uranium in Liquid Effluent Stream	Effluent Outfall	1.0×10^{-3}

[1] Based on an assumed activity of 4×10^{-7} Ci/g of depleted uranium - see Section 1.2.1 for isotopic composition

Solid UO_2F_2 from Reactor No. 1 flows into the rotary kiln, where it reacts with hydrogen and steam to form solid UO_2 and gaseous HF. The reaction is endothermic, and the kiln is electrically-heated to maintain the temperature at about 1200°F. Solid UO_2 is discharged from the reactor, cooled, and conveyed to the granulation area. There is also a drum loading station to provide interim storage of UO_2 in drums as necessary.

The HF and water vapor flow through a cyclone and sintered metal filter to remove UO_2 particulates, which are discharged back into the reactor. The off-gas vapor flows to the HF distillation column.

Hydrogen is provided from a packaged ammonia dissociator unit. The chemical reaction is $2NH_3 \longrightarrow N_2 + 3H_2$. Liquid ammonia is vaporized and fed to the dissociator, which decomposes the ammonia at 1600°F in a catalyst bed. The hydrogen/nitrogen mixture is fed to Reactor No. 2.

Preliminary major equipment descriptions include two 6 ft by 30 ft long Inconel rotary kilns, two 12 in. by 10 ft long screw conveyor/coolers, and a drum loading station.

4.3 HF DISTILLATION

The hot off-gas and vapor from the reactors flow to a distillation column for HF recovery and purification. The system is shown in Figure 4-6.

The vapor is first quenched with distillation column bottoms to cool the superheated vapor to its saturation temperature. The saturated vapor is fed to the column, which produces an anhydrous HF overhead product at 67°F containing about 200 ppm water, and a 49 wt% HF bottoms stream at 233°F that is recycled to Reactor No. 1.

The overhead product is condensed in a chilled water condenser at 40°F, collected and sampled. Upon satisfactory analysis, the HF is pumped through an underground pipeline to storage tanks in the HF storage building. The HF is then loaded into railroad tank cars or tank trucks for shipment to customers.

The noncondensible gases, primarily nitrogen and hydrogen, pass through a -70°F refrigerated condenser to recover additional HF. The gases then flow to the HF scrubbing system.

Major process equipment for the HF distillation process includes a 4 ft-six in diameter by 23 ft high Monel distillation column, equipped with a 2 ft diameter by 6 ft high shell and tube reboiler, a 4 ft diameter by 12 ft long 3,500 ft^2 40°F condenser, a 2 ft diameter by 6 ft long 150 ft^2 -70°F condenser, and two 6 ft diameter by 14 ft long 3000 gallon hold tanks equipped with cooling coils, and six 12 ft diameter by 45 ft long 38,000 gallon storage tanks.

4.4 HF SCRUBBING SYSTEM

Off-gas from HF distillation is treated in a scrubber to reduce atmospheric releases of HF to acceptable levels. The system is shown in Figure 4-7.

The off-gas enters a packed column, where it is contacted with a potassium hydroxide (KOH) scrub solution. The HF is removed by the reaction $HF + KOH \longrightarrow KF + H_2O$. The treated off-gas is filtered, mixed with ventilation exhaust air to dilute the hydrogen to a safe concentration, and discharged to atmosphere. The spent scrub solution is collected in a precipitation tank, where hydrated lime is added to remove the fluoride and regenerate the KOH by the reaction $2KF + Ca(OH)_2 \longrightarrow CaF_2 + 2KOH$. A minimum level of KF is maintained in the scrub solution by adding less than the stoichiometric quantity of lime. This ensures all the lime reacts, which keeps solid lime out of the CaF_2 product and the packed bed scrubber.

The scrub solution slurry is filtered in a rotary drum vacuum filter or a pressure filter to remove the solid CaF_2 precipitate. The CaF_2 is washed with water to remove impurities, and dried in a steam-heated rotary dryer. After cooling, the CaF_2 is packaged in drums and sent to the storage building.

The KOH and wash water filtrate are collected, and a side stream is withdrawn and evaporated to remove the water formed by the scrubber chemical reaction and the water added for CaF_2 washing. The filtrate is then cooled and pumped back to the scrubber as scrub solution.

Major equipment includes a 1 ft diameter by 10 ft high Monel HF scrubber with plastic packing, 4 ft diameter by 5 ft high 450 gallon Monel precipitation and filtrate tanks, a 2 ft diameter by 6 ft long Monel drum filter, a 2 ft diameter by 4 ft high Monel evaporator/ condenser unit, a 1 ft-6 in diameter by 5 ft long rotary dryer and associated tanks and pumps.

4.5   UO$_2$ GRANULATION

The UO$_2$ powder is processed to form free-flowing granules that enable precise and reproducible feeding of the pellet presses. This system is shown in Figure 4-8.

The raw UO$_2$ powder is fed to a hammer mill to eliminate any lumps or agglomerates. The milled powder is fed to a roller compactor, which compresses the powder to about 40% of theoretical density. The compacted powder sheets are then size-reduced in a granulator.

The UO$_2$ granules are then screened, with oversize particles recycled to the granulator and undersize particles recycled to the compactor. The product granules, with a particle size between -20 and +60 mesh, are blended with a dry lubricant in a double-cone mixer or ribbon mixer to produce granules containing 0.5 wt% Sterotex lubricant (powdered hydrogenated cottonseed oil by Capital City Products). The granules are conveyed to the pelletizing area. There is also a drum loading station to provide interim storage of UO$_2$ granules in drums as necessary.

Major process equipment includes a hammer mill, roller compactor, granulator, a vibrating screen separator, two 15 ft^3 double-cone blenders, a drum loading station, and a dust collection system.

Table 7-1, Annual Air Emissions During Operation

Pollutants	Principal Release Point	Annual Emissions (lb)
CRITERIA POLLUTANTS		Boiler/Other[1]
Sulfur Dioxide	Boiler Stack / Grade	70/54
Nitrogen Dioxide	Boiler Stack / Grade	9,300/460
Hydrocarbons	Boiler Stack / Grade	200/410
Carbon Monoxide	Boiler Stack / Grade	4,600/2,700
Particulate Matter PM-10	Boiler Stack / Grade	350/92
OTHER POLLUTANTS		
HF	Process Bldg. Stack	900
UO_2	Process Bldg. Stack	12
Copolymers	Cooling Tower	1,500
Phosphonates	Cooling Tower	150
Phosphates	Cooling Tower	150
Calcium	Cooling Tower	2,600
Magnesium	Cooling Tower	700
Sodium and Potassium	Cooling Tower	300
Chloride	Cooling Tower	500
Dissolved Solids	Cooling Tower	14,300

[1] Other sources are diesel generator and vehicles

Table 7-2, Annual Radiological Emissions During Operation

Radiological Isotope	Principal Release Point	Release Rate (Ci/yr)[1]
Depleted Uranium in Gaseous Effluent	Process Bldg. Stack	2×10^{-3}
Depleted Uranium in Liquid Effluent Stream	Effluent Outfall	2×10^{-3}

[1] Based on an assumed activity of 4×10^{-7} Ci/g of depleted uranium - see
Section 1.2.1 for isotopic composition

The sintered pellets have a density of about 9.8 g/cc (90% of theoretical), and are about 0.82 in diameter by 0.82 in long. The boats and carts exit the furnace through a second airlock, and the sintered pellets are removed from the boats and loaded into 30 gallon drums in a drum filling station. The drum is sealed, cleaned and transported to the storage building.

Major process equipment includes two 365 pellets/min rotary powder compacting presses, three 6 ft wide by 6 ft high by 70 ft long 400 kw sintering furnaces, an off-gas treatment system, a dust collection system, a drum loading station, and conveyors for the pellet boats.

4.4 HF ABSORPTION

The hot off-gas from the reactors, containing HF, water vapor, nitrogen and hydrogen, flows to a series of two absorption columns for HF recovery. The system is shown in Figure 4-7.

The vapor enters the first column, where it is contacted with aqueous HF solution. The HF and water condense, which increases the solution temperature by the heat of condensation and heat of solution. The liquid drains to the bottom of the column, where it mixes with liquid from the second absorber column. The resulting 20% HF solution is pumped through a heat exchanger for cooling and is recirculated to the top of the absorber column. A portion of the circulating liquid is continuously withdrawn and discharged to the HF neutralization system.

The vapor leaving the first column flows to the second column for additional HF removal. Fresh water is added to the second column to make up for the liquid that was discharged to the first column.

Major process equipment for the HF absorption process includes a 3 ft-6 in diameter by 20 ft high Monel absorption column, a 2 ft-6 in diameter by 10 ft long 1050 ft^2 Monel cooler, a 2 ft diameter by 19 ft high Monel absorption column, a 1 ft-6 in diameter by 7 ft long 250 ft^2 Monel cooler, and associated circulation pumps.

4.5 HF SCRUBBING SYSTEM

Off-gas from HF absorption is treated in a scrubber to reduce atmospheric releases of HF to acceptable levels. The system is shown in Figure 4-8.

The off-gas enters a packed column, where it is contacted with a potassium hydroxide (KOH) scrub solution. The HF is removed by the reaction HF + KOH ---> KF + H_2O. The treated off-gas is filtered, mixed with ventilation exhaust air to dilute the hydrogen to a safe concentration, and discharged to atmosphere. The spent scrub solution is collected in a precipitation tank, where hydrated lime is added to remove the fluoride and regenerate the KOH by the reaction 2KF + $Ca(OH)_2$ ---> CaF_2 + 2KOH. A minimum level of KF is maintained in the scrub solution by adding less than the stoichiometric quantity of lime. This ensures all the lime reacts, which keeps solid lime out

of the CaF_2 product and the packed bed scrubber.

The scrub solution slurry is filtered in a rotary drum vacuum filter or a pressure filter to remove the solid CaF_2 precipitate. The CaF_2 is washed with water to remove impurities, and transferred to the CaF_2 dryer in the HF neutralization system.

The KOH and wash water filtrate are collected, and a side stream is withdrawn and evaporated to remove the water formed by the scrubber chemical reaction and the water added for CaF_2 washing. The filtrate is then cooled and pumped back to the scrubber as scrub solution.

Major equipment includes a 1 ft diameter by 10 ft high Monel HF scrubber with plastic packing, 4 ft diameter by 6 ft high 550 gallon Monel precipitation and filtrate tanks, a 6 ft diameter by 4 ft long Monel rotary drum filter, a 1 ft-6 in diameter by 4 ft high Monel evaporator/condenser unit, and associated tanks and pumps.

4.6   HF NEUTRALIZATION SYSTEM

The 20% HF solution from absorption is neutralized with slaked lime to form CaF_2. The system is shown in Figure 4-9.

Pebble lime (CaO) is mixed with water in a vertical stirred mill to form a 25 wt% $Ca(OH)_2$ slaked lime slurry. The exothermic chemical reaction is $CaO + H_2O \longrightarrow Ca(OH)_2$. The slurry is collected in a feed tank with cooling coils, which cools the hot slurry to near ambient temperature.

The HF solution is mixed with the slaked lime in a continuous neutralization tank. The chemical reaction is $2HF + Ca(OH)_2 \longrightarrow CaF_2 + 2H_2O$. The reaction is exothermic and cooling coils are provided. The slurry then flows to a second neutralization tank for final pH adjustment.

The neutralized slurry, containing about 16 wt% solids, is filtered in a rotary drum vacuum filter to remove the solid CaF_2 precipitate. The CaF_2 is washed with water to remove impurities, and dried in a steam-heated rotary tube dryer. After cooling, the CaF_2 is packaged in drums and sent to the storage building. The filtrate and condensate are collected and reused in HF absorption and lime slaking. Excess water is sent to the industrial waste treatment facility.

Major equipment includes a two 9 ft diameter by 10 ft high 4,500 gal Monel HF neutralization tanks (one tank with cooling coils), a 9 ft diameter by 10 ft high 4,500 gal steel filtrate tank, a 3 ft diameter by 4 ft long steel rotary drum filter, an 11 ft diameter by 12 ft high 8,500 gallon recycle water tank, a 6 ft diameter by 35 ft long rotary steam tube dryer, and associated pumps, conveyors and bins.

4.7   UF6 CYLINDER HANDLING SYSTEMS

Incoming, filled DUF_6 cylinders will be off-loaded from either rail cars or flatbed trucks by a yard crane. The crane will place the cylinder on a cart,

Table 7-1, Annual Air Emissions During Operation

Pollutants	Principal Release Point	Annual Emissions (lb)
CRITERIA POLLUTANTS		Boiler/Other[1]
Sulfur Dioxide	Boiler Stack / Grade	60/60
Nitrogen Dioxide	Boiler Stack / Grade	8,000/470
Hydrocarbons	Boiler Stack / Grade	170/410
Carbon Monoxide	Boiler Stack / Grade	4,000/2,700
Particulate Matter PM-10	Boiler Stack / Grade	300/92
OTHER POLLUTANTS		
HF	Process Bldg. Stack	300
UO_2	Process Bldg. Stack	12
Copolymers	Cooling Tower	1,800
Phosphonates	Cooling Tower	180
Phosphates	Cooling Tower	180
Calcium	Cooling Tower	3,200
Magnesium	Cooling Tower	800
Sodium and Potassium	Cooling Tower	330
Chloride	Cooling Tower	600
Dissolved Solids	Cooling Tower	17,700

[1] Other sources are diesel generator and vehicles

Table 7-2, Annual Radiological Emissions During Operation

Radiological Isotope	Principal Release Point	Release Rate (Ci/yr)[1]
Depleted Uranium in Gaseous Effluent	Process Bldg. Stack	2×10^{-3}
Depleted Uranium in Liquid Effluent Stream	Effluent Outfall	2×10^{-3}

[1] Based on an assumed activity of 4×10^{-7} Ci/g of depleted uranium - see Section 1.2.1 for isotopic composition

DEPLETED URANIUM HEXAFLUORIDE MANAGEMENT PROGRAM

The Engineering Analysis Report
for the Long-Term Management of
Depleted Uranium Hexafluoride

J.W. Dubrin, J.N. Zoller, and L. Rahm-Crites
Lawrence Livermore National Laboratory

T.H. Bertell, Jr., S. Cavanaugh, M. Coflin, S. Coflin, C.J. Crockett, G. DeMoss, M. Feather, C. Herrington, P. Hogroian, C.L. Johnson, R.A. Kelly, T. Miller, D.C. Riley
Science Applications International Corporation

T. Breitmayer, R. A. Kaiser, B. Marais, M. Pong, D.R. Tiojanco, and D.B. Warren
Bechtel

T. Joseph, D. Milewski, and J.N. Sumner
Lockheed Martin Energy Systems

May 1997

Volume II

INS TITTF   RGY &
              NTAI SEARCH
        rei Avenue
      Park, MD
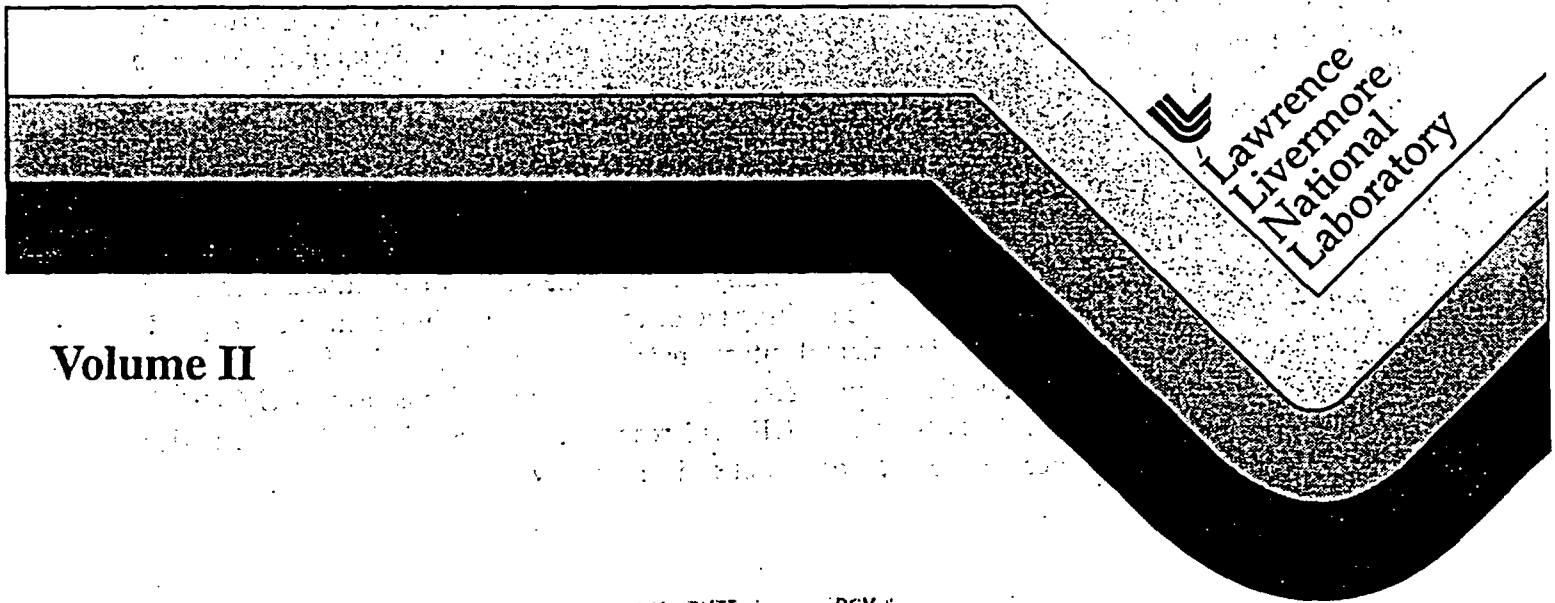
release from the disposal facility; transport to the environment by air, water, and the food chain; and exposure to the public from inhalation, ingestion, and external exposure. The time period over which this analysis is made can be very long (1,000 to 10,000 years). At any time during this period, certain radiological and chemical toxicities cannot be exceeded at the disposal site. For example, the annual radiation dose to a member of the general public cannot exceed 25 mrem/year (from all pathways). A disposal site must also meet the atmospheric pathway limit of 10 mrem/year (Clean Air Act). Chemical toxicity limits for uranium have been proposed by the Occupational Safety and Health Administration (OSHA) for continuous air exposure (0.05 mg/m^3), and by EPA for drinking water limits (60μg/L).

The radiological hazards associated with depleted uranium are the consequence of its isotopic composition. The typical concentrations of the three isotopes contained in depleted uranium are: 99.80 percent (U-238); 0.20 percent (U-235); and 0.0005 percent (U-234). Depleted uranium produced by gaseous diffusion is essentially free of daughter products from these isotopes. However, subsequent decay of the depleted uranium produces daughter products that lead to an ingrowth of decay products. It has been reported that the quantity of radon-226 produced by this process is insufficient to produce a significant radiological hazard for tens of thousands of years (Hertzler, 1994). Radionuclides with the short half-lives (Th-234, Pa-234, and Th-231) reach their maximum concentration within a few months after the production of depleted uranium, and this concentration then remains constant.

The radiological hazards from depleted uranium are primarily due to alpha particle emissions. Thus, for most cases, the limiting radiological hazard is the internal radiation dose from ingestion or inhalation. Highly insoluble uranium oxides such as UO_2 and U_3O_8 will be retained in the lungs for much longer times than more soluble uranium compounds such as UF_6. Table 1.1 (MMES, 1990) lists the inhalation classifications for the various compounds of depleted uranium. Although the low solubility of U_3O_8 is a positive factor in groundwater migration (and in kidney take up), it is a negative factor in air pathway considerations.

A comparison of the chemical and radiological toxicity of various forms of depleted uranium is given in table 1.2 (Hertzler, 1994). It should be noted that the chemical and radiological toxicities of the various compounds of uranium are generally of the same order of magnitude. Whether chemical or radiological toxicity will be the limiting factor for a given uranium compound depends on the specific situation. For environmental situations where drinking water is the primary concern, chemical toxicity will be the limiting factor for insoluble compounds such as U_3O_8. This is because highly soluble compounds such as UF_6 will reach the very restrictive EPA radiation dose limit for drinking water before exceeding a chemical toxicity limit.

Table 1.2   Chemotoxicity Versus Radiotoxicity for Various Chemical Forms of
Depleted Uranium (Hertzler, 1994)

Chemical Compound	Limiting Air Concentration			Limiting Water Concentration		
	Chemotoxicity[a] (mg/m³)	Radiotoxicity[b]		Chemotoxicity[c] (µg/L)	Radiotoxicity[d]	
		(pCi/m³)	(mg/m³)		(pCi/L)	(µg/L)
U₃O₈	0.68	189	0.47	60	220	550
UO₂	0.68	189	0.47	60	220	550
UF₆	0.07	540	1.35	60	22	55

a. Air concentration at which constant exposure results in a steady state kidney burden of 0.330 mg
(about 1 µg/g of kidney tissue). The OSHA occupational limit for continuous exposure is 0.05
mg/m³ based on chemical toxicity.

b. Air concentration at which constant exposure results in a radiation dose equal to the annual
occupational limit of 5 rem/year. Conversion from pCi/m³ to mg/m³ is based on a depleted
uranium specific activity of 4×10^{-7} Ci/g.

c. Proposed EPA standard for naturally occurring uranium in drinking water based on chemical
toxicity.

d. Drinking water concentration that would result in an annual dose equaling the EPA drinking water
standard of 4 mrem/year. Conversion from pCi/L to µg/L is based on a depleted uranium specific
activity of 4×10^{-7} Ci/g.

Table 1.3   Distribution of Various Wasteforms

	U_3O_8 (55-gallon drums)		UO_2 (30-gallon drums)	
	grouted	ungrouted	grouted	ungrouted
Density	2.88 g/cc (180 lb/ft^3)	3.0 g/cc (187 lb/ft^3)	8.0 g/cc (500 lb/ft^3)	9.0 g/cc (562 lb/ft^3)
Weight of drum contents	0.598 te (1,320 lb)	0.625 te (1,380 lb)	0.907 te (2,000 lb)	1.024 te (2,260 lb)
Amount of U per drum	0.254 te 560 lb	0.530 te 1,170 lb	0.600 te 1,320 lb	0.902 te 1,990 lb
Number of drums	1.494 million	0.715 million	0.632 million	0.420 million