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DEPLETED URANIUM HEXAFLUORIDE: WASTE OR RESOURCE?

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Abstract:

The U.S. Department of Energy is evaluating technologies for the storage, disposal, or re-use of depleted uranium hexafluoride (UF_6). This paper discusses the following options, and provides a technology assessment for each one: (1) conversion to UO_2 for use as mixed oxide fuel, (2) conversion to UO_2 to make DUCRETE for a multi-purpose storage container, (3) conversion to depleted uranium metal for use as shielding, and (4) conversion to uranium carbide for use as high-temperature gas-cooled reactor (HTGR) fuel. In addition, conversion to U_3O_8 as an option for long-term storage is discussed.

Introduction:

In the United States, uranium enrichment is currently accomplished through a process known as gaseous diffusion. In this process, gaseous uranium hexafluoride (UF_6) is separated into two streams - one enriched in Uranium-235 (U-235) and the other depleted in U-235. The U.S. has produced enriched uranium on a large scale using gaseous diffusion technology since the 1940s. Until very recently¹, the U. S. Department of Energy (the Department) was responsible for the uranium enrichment enterprise in the United States.

A major consequence of the gaseous diffusion process is the accumulation of a significant amount of depleted UF_6 . Although ratios may vary in practice, producing one pound of UF_6 enriched to 3.0 percent U-235 will typically result in 5.5 pounds of depleted UF_6 at 0.3 percent U-235. This depleted UF_6 is stored as a solid in a partial vacuum in 10- to 14-ton steel cylinders with 5/16 inch- (0.794 cm-) thick walls. The majority of the cylinders are approximately 12 feet (3.65 m) long and 4 feet (1.22 m) in diameter. Currently, this inventory occupies a total of about 47,000 cylinders containing approximately 560,000 metric tons of UF_6 .

Development of a Management Strategy for Depleted Uranium Hexafluoride:

The unique properties of depleted UF_6 , as well as the large volumes in storage, suggest that a careful evaluation and analysis of potential management strategies for the long term disposition of this material may result in the application of technologies and/or end-uses not previously considered. In an effort to develop a cost-effective, energy-efficient and environmentally safe

¹ In October 1992, the Energy Policy Act of 1992 created the United States Enrichment Corporation (USEC), and required the Department of Energy to lease the Portsmouth and Paducah Gaseous Diffusion Plants to the USEC effective July 1, 1993. The Department retains responsibility for the depleted UF_6 produced prior to July 1, 1993.

management strategy for the disposition of this material, the Department has initiated an innovative program to include the public and private industry in the recommendation and evaluation of various technologies for the potential storage, disposal, or re-use of this material. The technology assessment portion of this program was completed in June 1995. This paper will discuss the elements of the technology assessment, including the Department's request for recommendations, the evaluation criteria used for the assessment, and an analysis of various end-uses applicable to the nuclear fuel cycle.

Request for Recommendations and Technology Assessment:

On November 10, 1994, the Department published a notice in the *Federal Register* (FR 56324), asking individuals, industry, and other government agencies to submit suggestions for potential uses for depleted UF₆, as well as for technologies that could facilitate the long-term management of the material. The Department specifically requested recommendations for the following: (1) uses or applications of products or materials that include any form of depleted uranium and (2) technologies that could facilitate the long-term management of depleted uranium. The uses or applications could be for depleted uranium in its current chemical form (UF₆); for any of its individual components; for either the uranium or the fluorine in some other chemical or physical form; or for products made from any form or compound of depleted UF₆, including alloys, cements, or other materials. The deadline for submittal of recommendations was January 9, 1995. Fifty-seven responses containing 70 recommendations were received. Lawrence Livermore National Laboratory (LLNL) was chosen by the Department to conduct the technology assessment of these recommendations. LLNL assembled a group of Independent Technical Reviewers (ITRs) to assist in the assessment. The ITRs were selected based on experience in areas such as process technology, uranium processing and fabrication, engineering finance/economics, environmental engineering and waste management, hazards analysis, and environmental regulations. Using the evaluation factors described below and their own individual expertise, the reviewers assessed the technical feasibility of each recommendation.

Evaluation Factors

Evaluation factors were developed by Lawrence Livermore National Laboratory with input from the public to serve as guidelines in the conduct of the technology assessment. Six evaluation factors were considered in the assessments:

A. Environment, Safety and Health. This factor considers issues of concern to workers, the public and the environment such as: issues that may arise as a result of operations, transportation, handling, storage, and disposal, including effluents and emissions; issues that may restrict site choices when constructing or operating a facility that employs a specific technology or application; and design configurations, specifications, or operational requirements that pose problems of nuclear, chemical, or other safety issues involving workers or the public.

B. Waste Management. While related to Factor A, waste management was evaluated separately due to its potential significance. This factor included radiological, nonradiological, hazardous, toxic, mixed, or solid waste streams and waste volumes, or residual material that may pose problems of storage, transportation, treatment, or disposal; the potential for waste minimization in use or manufacture; and potential for recycling.

C. Costs. Consideration was given to costs which are associated with the development or use of a specific technology or with the use of a product, or which could preclude consideration of a recommendation. These include: capital costs, both initial, including research and development (R & D) and continuing; annual operating and maintenance costs; decontamination and decommissioning costs; value of any product or facility salvage; and cost avoidance through the sale of any byproducts.

D. Technical Maturity. For new technologies, issues such as time to availability and probability of success were considered by evaluating the following developmental stages: design - conceptual or detailed; bench or small scale; developed but untested on a large scale; tested or used on a large scale, but not standard industrial practice; or standard industrial practice.

E. Socioeconomic. Consideration was given to the effect of recommendations on socioeconomic indicators such as employment, public acceptance, and local or regional economic development.

F. Other. This factor included any other information believed pertinent to the feasibility of a submission.

Evaluation of Four Nuclear Fuel Cycle End-uses:

Table 1 summarizes several options for the use of depleted UF_6 . From the many recommendations submitted to the Department, four potential fuel cycle end-uses were chosen as examples for this paper: (1) conversion to UO_2 for use as mixed oxide fuel, (2) conversion to UO_2 to make DUCRETE for a multi-purpose storage container, (3) conversion to depleted uranium (DU) metal for use as shielding, and (4) conversion to uranium carbide (UC) for use as high-temperature gas-cooled reactor (HTGR) fuel. Each of these alternatives requires conversion to another form such as an oxide or metal. In addition, the conversion to U_3O_8 is discussed as an option for long-term storage. The following section will generally discuss the uranium processing techniques and provide an assessment of each of these end uses based on the previously described evaluation factors and input from the ITRs.

Conversion to UO_2 :

The conversion of UF_6 to ceramic UO_2 is industrially practiced in the fuel fabrication industry. Either by a "wet" or "dry" process, the UF_6 is converted to a UO_2 powder under carefully controlled conditions to assure suitable powder morphology. The pellet is then pressed under high pressure, and finally sintered to yield a solid which is typically 95% of the theoretical density. UO_2 in the ceramic form as pellets or small particles has a density several times that of normally compacted UO_2 or U_3O_8 powders.

There are two conventional wet processes commonly used for conversion of UF_6 to UO_2 . The ammonium diuranate (ADU) process involves the hydrolysis of UF_6 to UO_2F_2 followed by the addition of ammonium hydroxide to precipitate the uranium and ammonium diuranate, $(NH_4)_2U_2O_7$. After centrifuge separation from the liquid, the ADU slurry is dried. The ADU is calcined to uranium trioxide, and then reduced with hydrogen to UO_2 .

The ammonium uranyl carbonate (AUC) process is also a precipitation process, and was developed to reduce the number of steps associated with the ADU process. The UF_6 is

hydrolyzed to uranium fluoride, and, through the addition of ammonia and carbon dioxide, the uranyl fluoride is precipitated as ammonium uranyl carbonate $(\text{NH}_4)_4\text{U}_2\text{O}_7\text{CO}_3$. After filtration and drying, the AUC is calcined in the presence of hydrogen to UO_2 . The oxide is subsequently pressed and sintered.

In recent years, fuel fabricators are increasingly turning to dry routes when replacing or expanding capacity. The dry route has substantially fewer steps and fewer waste management issues than the wet routes. As in the case with the wet processes, there are several variations of dry processes. British Nuclear Fuels Limited uses a technique called the Integrated Dry Route, in which uranium hexafluoride vapor and steam react at one end of a rotary kiln to produce uranyl fluoride. The uranyl fluoride is then converted to UO_2 powder in the main body of the kiln by a mixture of hydrogen and steam introduced at the opposite end of the kiln. The reaction byproducts are hydrogen fluoride and water.

Mixed Oxide Fuel Application:

One use for UO_2 resulting from the conversion of depleted UF_6 is for blending with plutonium dioxide (PuO_2) or highly enriched UO_2 for the production of mixed oxide fuels (MOX) for light water reactors (LWRs). Currently, mixed oxide fuels are used in Europe, where reprocessing of spent LWR fuels yields considerable plutonium and slightly enriched uranium which can be recycled. Japan is also pursuing plutonium recycle and MOX fuels. The materials used (in Western Europe) to fabricate MOX reactor fuels for LWRs are slightly enriched UF_6 and plutonium recovered from spent nuclear fuel (SNF) recycling. The role for depleted uranium in this system would be as feed to a fast breeder reactor (FBR) cycle. Unlike the LWR cycle, the breeder cycle includes two discrete fuel types: a driver fuel, consisting of 20-30% plutonium in DU, and a blanket fuel, consisting of DU only. Although with the slowed growth of nuclear power there is no economic driver for the fast breeder reactors today, several demonstration FBRs were built in Europe in the 1970s, and the French built two large FBRs (Phenix and Super-Phenix).

If depleted uranium were used in the once-through (no SNF recycling) cycle used in the U.S., it would be as a UO_2 blend with weapons uranium or plutonium in place of enriched uranium. When consideration is given to the quantities of depleted uranium used in either the reprocessing cycle or the once-through LWR cycle, it is clearly not cost-effective to utilize depleted uranium in the production of MOX fuel in the U.S.; however, there may be limited application in blankets of sodium-cooled FBRs.

Conversion of depleted UF_6 to uranium dioxide for storage until the time of future application as MOX fuel has the advantage of retaining the uranium in a more stable and inert form. However, the costs of conversion of a substantial quantity of the depleted UF_6 inventory to UO_2 would be lower than conversion to uranium metal. Operation of fast neutron breeder reactors is technically mature, although mixed oxide (UO_2 and PuO_2) fuels can be more economically fabricated by reprocessing spent light water reactor fuel rods. The safe, long-term storage of UO_2 can be achieved, although public acceptance of a breeder reactor program may be forthcoming only after the fossil fuels are nearly exhausted.

DUCRETE Application:

The Department of Energy is currently developing a multi-purpose container for use in the future storage, transportation, and disposal of spent nuclear fuel. It has been proposed that

UO_2 could be used in concrete as shielding material in these containers. Concrete is generally a mixture of cement, sand (SiO_2), and aggregate (gravel, usually SiO_2 forms). By substituting a uranium oxide in the place of either the sand or the aggregate, a depleted uranium concrete (DUCRETE) can be produced which has a much higher density than standard concrete. Current DUCRETE development has focused on using stabilized dense UO_2 as the aggregate and U_3O_8 as the substitute material for the sand to achieve the desired density.

A manufacturing site for DUCRETE production would have to handle the oxide source material. The primary health and safety concerns arise from internal radiation exposure due to inhalation of airborne oxides or from the chemical toxicity of the uranium as a heavy metal due to ingestion. Therefore, a concrete factory producing DUCRETE would need to be fully enclosed and equipped with air filtering, pressure control, radiation detection, etc. Disposal of DUCRETE after container use would present additional waste management issues, although this issue may be mitigated in the case of deep geological disposal of spent nuclear fuel. Breaking up large DUCRETE structures would entail airborne particulate hazards similar to those produced during manufacture. The costs for manufacture and disposal of DUCRETE would exceed those of concrete. The technology for large-scale production of DUCRETE has not been developed. Use of DUCRETE as shielding material for on-site storage of spent nuclear fuel or in shipping containers appears to be a reasonable option, although life-cycle costs could be somewhat high, particularly when decontamination and decommissioning costs of such facilities are considered.

Conversion to DU Metal:

Depleted uranium metal has been produced for many years, primarily for defense purposes. The standard industrial process in the U.S. has been the batch metallothermic reduction of uranium tetrafluoride with magnesium metal (Ames process). This process generates a magnesium fluoride byproduct slag which is contaminated with appreciable quantities of uranium in various forms. Without further treatment, the slag (about 0.5 kg/kg-U) must be disposed of as low-level waste. There are a variety of options to decontaminate the slag, including options which also recover the fluorine value for recycle.

Another option is continuous metallothermic reduction, which offers higher throughput than the currently practiced batch process, and a MgF_2 byproduct with a much lower level of uranium contamination. A fundamentally different option is the plasma dissociation of UF_6 gas. In the presence of a hydrogen quench, the end products are uranium metal and anhydrous hydrogen fluoride (AHF).

Metal Shielding Application:

The beneficial re-use of depleted uranium metal for radiation shielding of commercial SNF or vitrified high-level waste (HLW) containers has been considered by the Office of Technology Development. Uranium provides an effective gamma shield, and depleted uranium metal could be utilized in containers for storage and transportation of vitrified HLW (Yoshimura 1993), in metal shielded casks for on-site dry storage and subsequent shipment of SNF (Hertzler and Nishimoto 1994), or incorporated in the shield plug for multi-purpose container designs. It was concluded that these applications could possibly use the entire inventory of depleted uranium. This particular application addresses two major concerns: SNF/HLW shielding and depleted uranium disposition.

Conversion of depleted UF₆ to uranium metal has been accomplished for decades by one of several technically mature industrial processes. The conventional Ames reduction process produces an amount of solid waste, mostly in the form of MgF₂, approximately equivalent to that of the uranium metal produced. This solid waste would have to be disposed of as low level waste or else processed to remove nearly all of the uranium in order to dispose of the waste in a sanitary landfill. Use of uranium as shielding for spent fuel rods in various casks designed for storage and transportation requires that the uranium be protected against oxidation during manufacture and storage. Utilization of uranium metal for shielding in SNF and HLW canisters appears to be a viable option for the re-use of the depleted UF₆.

Conversion to Uranium Carbide:

Uranium carbides (UC) are usually manufactured in spherical shapes and then assembled into the desired form. To date, most uses for uranium carbide have been in nuclear fuel applications. Depleted UF₆ can be converted to UO₂ by various methods previously described. The UO₂ then becomes the starting material for microsphere production (Benedict, Pigford, and Levi 1981). In a commonly used process, the UO₂ powder is mixed with carbon flour and an ethylene binder to form a slurry, which is oven-dried and milled to sand-sized particles. The oxides are converted to carbides in a vacuum heating step. Subsequently, coatings are applied to the microspheres in a fluidized bed furnace to isolate the UC from the environment at the microscopic level (GA Technologies 1982). The coated spheres are then assembled into fuel rods for reactor use. In some applications, other types of fuel materials (e.g., thorium) may be incorporated into the fuel with the spheres.

High -Temperature Gas Reactor (HTGR) Fuel Application:

The typical commercial HTGR fuel cycle utilizes high enriched (93%) uranium (HEU) and thorium (Th). Potential HTGR fuel cycles using depleted uranium include a low enriched (LEU) cycle, which would use a mixture of 5-15% enriched UO₂ and depleted UO₂, and an HEU cycle, which could be blended down with depleted uranium or recycled U-233 (from a previous HEU-Th cycle). Although there are no commercial HTGRs currently operating in the United States, several have operated in the past, and advanced HTGR design work is underway.

Typical fuel fabrication plant for HTGRs convert UO₂ and thorium dioxide (ThO₂) into fuel elements. The fuel element for an HTGR consists of a hexagonal block of graphite into which vertical coolant and fuel holes are drilled. The fuel holes are filled with rods consisting of a graphite sleeve containing a column of cylindrical fuel compact. In addition to fuel and coolant channels, fuel elements contain a small amount of boron carbide (BC), formed into rods, to act as a burnable poison. HTGR fuel consists of tiny, spherical, carbon-coated, enriched UC₂ and Th particles blended together and formed into rods by means of a matrix filler and binder. The basic steps in manufacture of HTGR fuel assemblies are: particle production; fuel rod fabrication; and element manufacture. Particle production consists of a fissile particle production process which results in either HEU or LEU uranium carbide fuel particles. The fertile particle production process results in the formation of Th particles or, in the case of a depleted uranium cycle, the formation of depleted uranium fertile fuel particles. The fissile and fertile particles are then fabricated into fuel rods through an injection molding process, and the resulting fuel rods are positioned in their respective channels to form the fuel element.

In order for depleted uranium to be used in the HEU-Th cycle, it must first be heavily re-enriched, which does not prove to be economically feasible. It is more likely that depleted uranium would be consumed through the LEU cycle, with the following assumptions:

- (1) depleted uranium stockpile is re-enriched to 15% for use in the fissile particle production;
- (2) the remaining 0.1% tails are used in the fertile particle production in place of the Th;
- (3) the material weight requirements for the LEU fissile and fertile particle production are the same as in the HEU-Th case. In this scenario, almost 600 metric tons of the depleted UF_6 stockpile could be consumed annually. If the depleted uranium were used in the fertile particle production only, approximately 540 metric tons could be used annually. This use rate assumes a reference facility that produces approximately 96 fuel assemblies per day.

Conversion of depleted UF_6 to uranium carbide requires the initial conversion to UO_2 , producing a CaF_2 waste stream and hydrofluoric acid (HF), followed by formation of UC by either a graphite or gelation method. Additional costs arise from the use of depleted, rather than naturally occurring, UF_6 as the feed material, due to the substantially increased energy costs associated with gaseous diffusion. Relatively small quantities of depleted UF_6 would be expected to be utilized by this process due to the lack of commercial acceptability of the HTGR technology. Public acceptance for the construction and operation of additional nuclear power plants in the U.S. has diminished in the last 15 years. Therefore, the option of using significant quantities of depleted UF_6 as HTGR fuel does not appear to be reasonable at this time.

Conversion of UF_6 to U_3O_8 :

The conversion of UF_6 to U_3O_8 is commonly referred to as defluorination. The by-product of the defluorination process is either HF or anhydrous HF (AHF), depending on the process selected. There is a large market for AHF in North America, but only a limited market for hydrofluoric acid. In Europe, however, there is a large market for concentrated hydrofluoric acid (typically 70% HF). Cogema operates the world's only defluorination facility (France) for depleted UF_6 . Two example processes are provided below for the conversion of UF_6 to U_3O_8 .

The Cogema process for defluorination with hydrofluoric acid by-product is a two-step, vapor-phase process. In the first step, UF_6 vapor is hydrolyzed with steam (at 250°C) to produce solid UO_2F_2 and gaseous hydrogen fluoride and water. The oxyfluoride is then fed to a rotary reactor and pyrohydrolyzed (at 750°C) with hydrogen and superheated steam to give U_3O_8 and additional hydrogen fluoride gas. The HF/steam stream is filtered and condensed to recover concentrated HF.

In the U.S., General Atomics has developed a process to produce U_3O_8 with an AHF byproduct. This patented process also involves a two-step reaction sequence to produce U_3O_8 . UF_6 is first reacted with steam to produce a uranyl fluoride intermediate and a gaseous mixture of HF and water. The second step then converts the intermediate by steam to U_3O_8 and a gaseous mixture of HF, water, and oxygen. The gaseous HF/H₂O mixtures from the two reactors are combined and separated in a distillation column to obtain an AHF stream and an aqueous azeotrope stream. The azeotrope stream is vaporized and recycled to the primary reactor as steam feed. Although this process has been successfully demonstrated at a laboratory scale, it has not yet been commercialized at the industrial scale.

Due to the reactive nature of UF_6 , the depleted uranium inventory could be converted to U_3O_8 for interim or long-term storage. The advantages of U_3O_8 are the relatively low chemical reactivity, solubility, and health risks compared to other uranium forms. U_3O_8 is insoluble even in the weak acids and bases typically found in soils and groundwater (Martin Marietta 1990). Storage of the material in the form of U_3O_8 would not preclude the use of this material at a later date.

Conclusion and Discussion:

The overwhelming response to the Request for Recommendations and the assessment of the technologies presented in this paper, as well as others, indicate that there are opportunities to utilize the depleted uranium resulting from the enrichment process in the nuclear fuel cycle. Completion of the Technology Assessment Project ended the first phase in the process of selecting a long-term management strategy for the Department's depleted uranium. A more detailed engineering and cost analysis is ongoing. These analyses will be utilized in the development of an Environmental Impact Statement and the final Record of Decision, which is scheduled to occur in 1998.

The Depleted Uranium Hexafluoride Management Program has provided an opportunity for public involvement in a major federal action and engaged private industry in the development of a cost-effective management strategy for disposition of this material. This program actively encouraged the involvement of the public and private sector in the Department's decisionmaking process. The consideration of re-use applications is important as the U.S. and other countries continue to strive toward a sustainable economy and to minimize wastes associated with energy production and other industrial processes. A program such as the one described here could be used as a model for many other issues currently facing the U.S. Department of Energy and other federal agencies, including the re-use of federal facilities and the disposition of excess government equipment and materials.

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Table 1 - Summary of Options

Conversion (Section 2.0)				Storage (Section 3.0)				Recycle/Reuse (Section 4.0)				Disposal (Section 5.0)			
U ₃ O ₈ (Defluorination Processes)	HF Byproduct	Cogema	Chemical Forms	Facilities	Outside Yards, Buildings, or Vaults	Dense Material Applications	Chemical Forms	Waste Forms	Site Characteristics	U ₃ O ₈	GC	Bulk, containerized, encapsulated, or vitrified	DOE/ commercial, east/west, wet/dry, near-surface/ mined		
	AHF Byproduct	Generic	U ₃ O ₈	Re-enrichment	AVLIS										
UO ₂ (Ceramic)	General Atomics	General Atomics	U ₃ O ₈	Radiation Shielding	Metal										
	Cameco	ADU	UO ₂ (Ceramic)	Oxide	Dense UO ₂										
	Conventional	AUC				Surplus Weapons	LEU Fuel								
	IDR					Fissile Material Disposition	MOX Fuel								
	Advanced	Gelation/ Sol-gel				Advanced Reactor Fuel Cycles	Fast Breeder								
Metal	Improved Ames (Batch metallothermic)	Generic	Metal					Integral Fast Reactor		Uranium Carbide ¹					
	Continuous metallothermic reduction	Cameco						Thermal Reactor							
	Plasma Reduction	INEL/ LANL	Uranium Carbide ¹												

¹ Processes for converting UF₆ to uranium carbide would be involved if carbide options were developed.