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CHARACTERIZATION OF OPTIONS AND THEIR ANALYSIS REQUIREMENTS FOR THE LONG-TERM MANAGEMENT OF DEPLETED URANIUM HEXAFLUORIDE

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ABSTRACT

The Department of Energy (DOE) is examining alternative strategies for the long-term management of depleted uranium hexafluoride (UF_6) currently stored at the gaseous diffusion plants at Portsmouth, Ohio, and Paducah, Kentucky, and on the Oak Ridge Reservation in Oak Ridge, Tennessee. This paper describes the methodology for the comprehensive and ongoing technical analysis of the options being considered. An overview of these options, along with several of the suboptions being considered, is presented. The long-term management strategy alternatives fall into three broad categories: use, storage, or disposal. Conversion of the depleted UF_6 to another form such as oxide or metal is needed to implement most of these alternatives. Likewise, transportation of materials is an integral part of constructing the complete pathway between the current storage condition and ultimate disposition. The analysis of options includes development of pre-conceptual designs; estimates of effluents, wastes, and emissions; specification of resource requirements; and preliminary hazards assessments. The results of this analysis will assist DOE in selecting a strategy by providing the engineering information necessary to evaluate the environmental impacts and costs of implementing the management strategy alternatives.

INTRODUCTION

BACKGROUND

With the publication of a Request for Recommendations and Advance Notice of Intent in the November 10, 1994 *Federal Register* (59 FR 56324), DOE initiated a program to assess alternative strategies for the long-term management or use of depleted UF_6. The basic alternatives to continuing the current management plan for the disposition of depleted UF_6 are: (1) use, (2) storage, and (3) disposal. The current management plan entails handling, inspection, monitoring, and maintenance activities to ensure safe storage of the depleted UF_6 cylinders until the year 2020 when conversion to an oxide would begin. The alternatives will be analyzed in an Environmental Impact Statement (EIS) for their impacts on the natural environment and human health. In addition, an accompanying Cost Analysis Report will be developed to provide comparative economic and cost data. The EIS and the Cost Analysis Report will be utilized by DOE in the decision-making process, which will ultimately result in a Record of Decision in early 1998. This Record of Decision will complete the first phase of the Program, management strategy selection. During the second phase, site specific and technology specific considerations will be determined.

The Engineering Analysis Project for the Depleted UF_6 Management Program consists of a technology and engineering assessment of the proposed uses and alternative management strategies. Technology Assessment is the first component of the program. In the November 10, 1995, *Federal Register* notice, DOE requested recommendations from interested individuals, industry, and other government agencies for potential uses and technologies that could facilitate the long-term management of depleted UF_6. A total of 57 responses containing about 70 recommendations were received. These recommendations included five options that DOE was already considering, but which were not suggested in any of the other responses. Recommendations received before the submission deadline were evaluated by five independent technical reviewers. Responses received after the submission deadline were evaluated by the Independent Technical Reviewers as time allowed, or by Lawrence Livermore National Laboratory and Science Applications International Corporation staff. The findings were reported in a Technology Assessment Report (TAR), published June 30, 1995 [Lawrence Livermore National Laboratory, UCRL-AR-120372 Volume I, 600 p., Volume II-Appendices, 400 p.].

Engineering data (pre-conceptual) are being gathered on various methods of material management and use under the Engineering Analysis Project. This generic, non-site specific data will be documented in the Engineering Analysis Report, which is scheduled to be completed by April 1, 1996. The Engineering Analysis Project is analyzing the alternative strategies by their components (modules). Figure 1, the Work Breakdown Structure, summarizes the modules and the top level options that are the building blocks for any alternative. Conversion of the depleted UF_6 to another form, such as the oxide or metal, is needed to implement most of the alternatives. A number of technologies are possible for each of these conversion options and, likewise, there are multiple possibilities under each of the other module options. The next level of detail in the Work Breakdown Structure, shown in Figure 2, is referred to as suboptions. These suboptions provide the necessary definition for the engineering analysis and determination of environmental risks.

ENGINEERING ANALYSIS METHODOLOGY

This paper presents an overview of the set of options being analyzed in depth for the Engineering Analysis Report. Individual Engineering Data Reports are being generated under each option which include process flowsheets, top-level facility layouts, resource requirements, emission and waste data, and preliminary hazards assessments. The basis for these options are the responses to the Request for Recommendations. Figure 2 shows the options/suboptions that are being analyzed.

The Environmental Impact Statement is the first level of an assessment containing two tiers. The potential impacts of broad management strategy alternatives will be analyzed in the first tier. There were a significant number of promising conversion technologies recommended in the responses to the Request for Recommendations that, with minor exception, are less technically mature, but potentially offer unique features in the areas of environmental and cost benefits. Because these recommendations are either in the early stages of conceptualization or development, or else contain key design aspects that are proprietary, Engineering Data Reports are not being generated for these options and/or suboptions. They are, nonetheless, preserved for later decision making during the second tier of the assessment process, when more narrowly focused issues such as specific siting, technology, and transportation issues would be analyzed.

For facility sizing purposes, a period of 20 years is assumed for all module options to disposition the entire depleted uranium stockpile (560,000 tonnes UF_6 in about 46,400 cylinders). This corresponds to an annual rate of 28,000 tonnes of UF_6 or about 19,000 tonnes of uranium.

DISCUSSION OF THE ENGINEERING ANALYSIS

TRANSPORTATION MODULE

This critical element refers to the preparation of the depleted UF_6 cylinders at their current storage sites for transportation to an off-site facility for

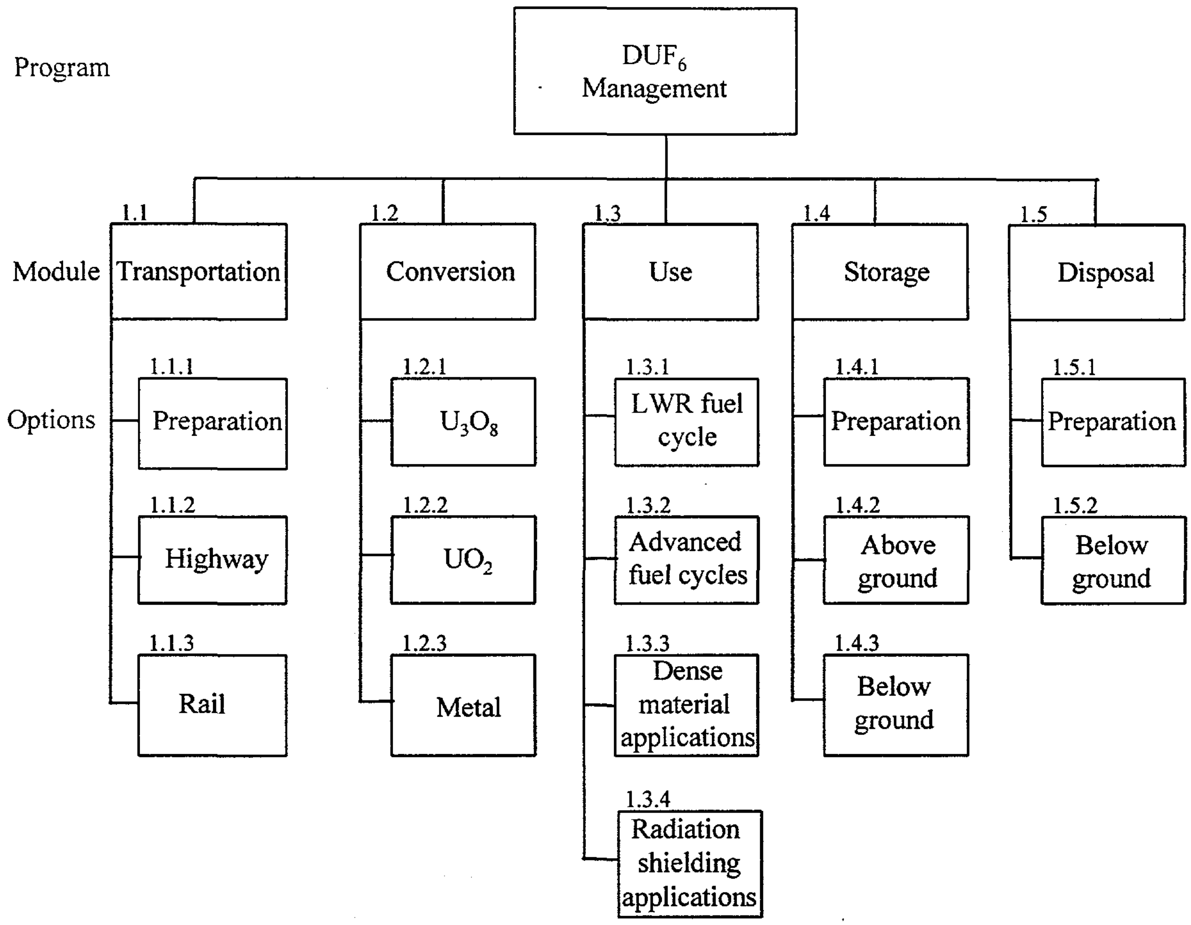

Figure 1. Depleted UF_6 Management Program work breakdown structure.

Conversion (in general). A number of the cylinders currently do not meet Department of Transportation (DOT) requirements for off-site shipment. The cylinder problems are of three types: (1) overfilled cylinders, (2) overpressurized cylinders, and (3) substandard cylinders (e.g., cylinders with below minimum value wall thickness or other nonconforming characteristics). Preliminary estimates of the numbers of cylinders with each of these problems have been made, but they are very rough and are associated with many uncertainties. Although overfilled cylinder records are fairly reliable, overpressurized cylinder predictions are based on limited inspections and are less reliable. Predictions of the future number of substandard cylinders is highly uncertain. In analyzing the Cylinder Preparation Option, two distinct suboptions are being evaluated to address problem cylinders.

Cylinder Preparation Option

The first suboption utilizes a protective overpack approach thereby addressing all three problems. The technology option being analyzed involves placing the UF_6 cylinder in a horizontal "clamshell" vessel for shipment. The overpack would be suitable to contain, transport, and store the cylinder contents regardless of cylinder condition, and could be designed as a pressure vessel enabling liquefaction of the depleted UF_6 for transfer out of the cylinder. Thermal design

Transportation module		Conversion module		Use		Storage module		Disposal module	
Options	Suboptions	Options	Suboptions	Options	Suboptions	Options	Suboptions	Options	Suboptions
Preparation	• Overpack	U_3O_8	• Dry process with AHF** by-product • Dry process with HF neutralization	LWR fuel cycle	• Re-enrichment	Above ground	• Building - U_3O_8 - UF_6 - UO_2	Below ground	• Trench - U_3O_8 cemented - U_3O_8 - UO_2 cemented - UO_2
	• Transfer Facility	UO_2	• Dry process with AHF by-product • Dry process with HF neutralization • Wet process with AHF by-product	Advanced reactor fuel cycles	• Breeder and other fast neutron spectrum reactors				• Vault - U_3O_8 cemented - U_3O_8 - UO_2 cemented - UO_2
Highway	• Truck	U	• Batch metallo-thermic process with AHF by-product • Continuous metallo-thermic process with AHF by-product	Dense material applications	• Existing applications: munitions, armor, counter-weights, and ballasts • New applications	Below ground	• Vault - U_3O_8 - UO_2 • Mined cavity - U_3O_8 - UF_6 - UO_2		• Mined cavity - U_3O_8 cemented - U_3O_8 - UO_2 cemented - UO_2
Rail	• Flatcar			Radiation shielding	• U-metal shielding for spent nuclear fuel • UO_2 shielding for spent nuclear fuel				

* Shaded areas include options/suboptions considered but not analyzed in depth

** Anhydrous hydrogen fluoride (HF)

Figure 2. Depleted UF_6 Management Program options and suboptions being analyzed.

analyses are required to establish heat transfer rates for volatilization. Wall thickness and other design details would be determined during detailed conceptual design.

The second suboption utilizes a facility to transfer the depleted UF_6 from problem cylinders into new cylinders. Construction and operation of a Transfer Facility for all problem cylinders would appear to bound potential environmental impacts. The Transfer Facility could also be used to develop a long term Storage Alternative for storing the depleted UF_6 in new cylinders.

CONVERSION MODULE

Conversion of the depleted UF_6 to another chemical form is required for most alternative management strategies. The three principal uranium forms of interest are: (1) triuranium octaoxide (U_3O_8), (2) uranium dioxide (UO_2), and (3) uranium metal (U). Due to their high chemical stability and low solubility, uranium oxides in general are presently the preferred form for the Storage and Disposal alternatives. High density UO_2 and U metal are the preferred forms for spent nuclear fuel radiation shielding applications due to their efficacy in gamma ray attenuation. Uranium metal is the required form for most dense material applications, where high density and high kinetic energy are the determining factors. All conversion processes start with the volatilization of UF_6 and all processes being analyzed in depth involve the processing of major quantities of hydrogen fluoride (HF). Uranium hexafluoride and hydrogen fluoride represent the major chemical hazards to the environment and the worker.

U_3O_8 Option

The conversion of uranium hexafluoride to U_3O_8 by steam is often referred to as defluorination. This "dry" process is well established and is utilized on a large scale industrial basis 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 uranium oxyfluoride (UO_2F_2) and HF. In the second, highly endothermic step, the UO_2F_2 is pyrohydrolyzed with superheated steam to U_3O_8 and additional HF. Due to the large excess steam requirements for the second step, concentrated hydrofluoric acid (typically 70% HF - 30% H_2O) is the direct process by-product. The U_3O_8 would be compacted to achieve a bulk density of about 3 grams/cc prior to storage or disposal.

Two suboptions are being developed in the Engineering Analysis Project for the dry conversion of UF_6 to U_3O_8. In the first process, the concentrated HF is upgraded to anhydrous HF (AHF, $< 1\%$ H_2O) for sale with unrestricted usage. The second process neutralizes the HF with CaO to form calcium fluoride (CaF_2) for disposal.

Defluorination with AHF production is preferred over defluorination with HF neutralization in terms of waste avoidance and by-product value. This is because there is a considerable market for AHF in North America, while the market for HF is somewhat limited. It is anticipated that the AHF will contain only trace amounts of depleted uranium (less than 1 part per million, or 0.4 picocuries/gram). As generally recommended in the responses to the Request for Recommendations, the hydrofluoric acid is upgraded to AHF by distillation.

In the unlikely event that the AHF cannot be marketed due to the presence of trace amounts of uranium – either for unrestricted use, or recycled in the nuclear fuel industry for the conversion of yellowcake (concentrated U_3O_8) to natural UF_6 – the acid would be neutralized with lime and disposed of in an ordinary landfill. Alternatively, in the absence of regulatory constraints, the CaF_2 could be sold as a fluorspar substitute for the commercial production of AHF. This would avoid the hazards associated with the handling, storage, and transportation of large quantities of AHF.

In addition to the multiple responses recommending dry conversion with upgrading to AHF, there were several recommendations on emerging technologies or new concepts that offer unique features in the areas of environmental and cost benefits. One such case is a molten metal catalyzed process for conversion to uranium oxides. This single step conversion offers a more compact process, avoids the HF distillation step, minimizes the post processing steps to increase the bulk density, and offers a broad degree of by-product flexibility.

High Density UO_2 Option

High density UO_2 refers to uranium dioxide having a particle density greater than or equal to 90% of its theoretical density (10.8 grams/cc). Depending on the particle shape, size, and size distribution, the bulk density of UO_2 will generally be 2-3 times that of compacted U_3O_8 powder. This higher density translates into substantially reduced space requirements for the Storage and Disposal alternatives. It also enables use in radiation shielding applications, where the oxide is substituted for the course aggregate material in conventional concrete.

Three suboptions are being developed in the Engineering Analysis Project for the conversion of UF_6 to UO_2. A generic industrial dry process with conversion (similar to that described for U_3O_8), followed by conventional pelletizing and sintering to produce centimeter-sized pellets, is the basis for the first two suboptions. The first suboption upgrades the concentrated HF to AHF for sale with unrestricted usage. The second suboption neutralizes the HF to calcium fluoride (CaF_2) for disposal or sale. The third suboption is a wet process, based on pilot-scale studies, and is referred to as gelation process.

Step one in the dry process is the same as the first step in the U_3O_8 conversion process. Namely, the gaseous UF_6 is hydrolyzed with steam to produce solid UO_2F_2 and HF in an exothermic reaction. The solid UO_2F_2 from the steam hydrolysis is converted in an endothermic reaction to UO_2 powder in the second reactor by a mixture of steam and a stoichiometric quantity of hydrogen. 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% of theoretical. The pellets are sintered in furnaces with a hydrogen reducing atmosphere to achieve an assumed density of about 90% of theoretical. The HF can either be upgraded to AHF or neutralized, as described in the section on U_3O_8 conversion processes.

In the gelation process, dense microspheres of UO_2 (millimeter-sized) are produced by the sintering of ammonium diuranate-like spheres in the presence of hydrogen gas. The initial step in the gelation process is the steam hydrolysis of UF_6 to form UO_2F_2 and HF. Since only a minor amount of excess of steam is required for this exothermic step, the process should essentially produce AHF. The gelation process intrinsically avoids the pelletizing step and, compared to the conventional route, results in a higher throughput while sintering at a lower temperature. The smaller sized particles afforded by the gelation process enable higher bulk densities.

U Metal Option

Two metallothermic reduction routes (batch and continuous) are being analyzed in depth for the production of uranium metal. Both processes utilize the same chemical reaction; namely, the magnesium (Mg) metal reduction of uranium tetrafluoride (UF_4) to produce U metal and a magnesium fluoride (MgF_2) by-product slag. The UF_4 required for either process would be generated by the hydrogen (H_2) reduction of depleted UF_6 (a standard industrial process), producing AHF as the by-product.

The standard industrial process for approximately 50 years has been the batch metallothermic reduction process. The MgF_2 by-product slag resulting from this process is contaminated with appreciable quantities of uranium. Without further treatment, the slag must be disposed of as a low-level waste (LLW). With the rising cost for LLW disposal, the disposal cost has become a significant fraction of the total cost for producing metal. For the batch metallothermic suboption, an acid leaching step to reduce the uranium content in the slag and enable its disposal in a sanitary landfill is being analyzed. An exemption would be required for slag disposal in a sanitary landfill since the uranium activity in the treated slag would still be large compared to that in typical soils.

The other suboption being analyzed in depth is the continuous metallothermic reduction process, currently under development. Here, the reactants are continuously fed into the top of a heated vertical reactor. Due to density differences, the molten uranium product settles to the bottom of the reactor while the molten MgF_2 by-product floats on top. The uranium and MgF_2 are separately withdrawn from the reactor on a continuous basis. The continuous metallothermic reduction process offers three primary advantages: (1) higher throughput for a comparable size reactor, (2) a much lower level of uranium contamination of the by-product slag, and (3) a liquid uranium product stream for direct casting into the end-product form, i.e., avoidance of a remelting step. The current continuous metallothermic reduction design produces a uranium alloy product containing a small percentage of iron. This alloy is judged to be acceptable for the primary use of interest, radiation shielding. The initial expectation is that the level of uranium contamination in the MgF_2 by-product will be sufficiently low that post-treatment would not be necessary. Again, an exemption for disposal in a sanitary landfill would be required.

The central issue for metallothermic reduction processes in general is the disposition of the by-product slag. Increasingly stringent requirements for non-LLW disposal may require alternate or additional treatment processes. There were several responses to the Request for Recommendations which specifically addressed the treatment of the MgF_2 by-product slag. These advanced MgF_2 treatment technologies, which are at an early stage of development, offer key waste minimization and economic benefits. In addition to alternate technologies for improved decontamination, these recommendations integrally addressed the recovery and beneficial use of the by-product constituents.

Plasma dissociation of UF_6 is a fundamentally different technology for metal production, offering a single step conversion process without the generation of MgF_2. This technology was recommended in response to the Request for Recommendations, and bench scale experiments have generated small quantities of uranium metal. Since the plasma process is in the early stages of development and there are design uncertainties, it is not being analyzed in detail.

USE MODULE

There are a variety of options for uses of the conversion products of depleted UF_6. These include light water reactor fuel cycle, advanced reactor fuel cycle, dense material, and radiation shielding applications. Of the various uses proposed in response to the Request for Recommendations, the production of radiation shielding material provides the basis for the two suboptions being analyzed. Other dense material applications are bounded by the more general radiation shielding application. Use in the light water reactor fuel cycle (either in re-enrichment processes or as blending material) or for advanced reactor cycles are not being analyzed in depth in the Engineering Analysis Project. The reason for not analyzing the fuel cycle options in depth is their longer time frame (particularly for advanced reactors). The option to pursue these uses in the future is preserved through the analysis of the storage options being considered.

Radiation Shielding Option

The engineering analysis is considering two principal forms for use of depleted uranium – dense UO_2 and metal – and their shielding manufacturing approaches. The first suboption uses depleted uranium as sintered UO_2 in the manufacture of depleted uranium concrete for shielding in spent nuclear fuel storage containers. This concrete, which substitutes dense UO_2 for the coarse aggregate (typically, silica) in conventional concrete, is known as Ducrete. Ducrete offers size and weight advantages over conventional concrete. Ducrete may also be an appropriate material for overpacks in spent nuclear fuel disposal, although this is more speculative than its use in storage applications. Accordingly, after the period of spent nuclear fuel storage is completed, the engineering analysis assumes the empty Ducrete cask would be disposed of as low-level waste.

The second suboption uses depleted uranium as the metal for the manufacture of annular shields for a Multi-Purpose Unit system. The Multi-Purpose Unit concept is to provide a spent nuclear fuel package that, once loaded at the reactor, provides confinement of spent nuclear fuel assemblies during storage, transportation, and disposal. In this approach, the depleted uranium is disposed of with the spent nuclear fuel.

For both shielding suboptions the shielding material would be enclosed between stainless steel (or equivalent) annular elements (shells) to provide structural integrity and avoid contact with the atmosphere.

STORAGE MODULE

Storage of depleted uranium is predicated on its use at a later date. Storage options are defined by the chemical form of the depleted uranium stored and the type of storage facility. The possible types of storage facilities being analyzed in the Engineering Analysis Project are: (1) above ground buildings, (2) below ground vaults, and (3) below ground mined cavities. The chemical form of the depleted uranium for storage depends partly on which of the use options is considered to be most likely utilized. Storage in the form of UF_6 provides maximum flexibility for future uses, and it is difficult to predict which of the use options would be most likely selected in the longer term. Storage in another form, such as UO_2, would imply a specifically identified future use option. However, U_3O_8, a relatively benign material, is the generally recommended form for disposal. Hence, storage as U_3O_8 is an alternative to storage as UF_6, until a determination is made that all or part of the depleted uranium is no longer needed.

Another consideration in evaluating the chemical form is the storage area required. The storage area is a function of the uranium bulk density, the type of storage containers, and the container storage configuration. The representative bulk densities for UF_6, sintered UO_2 microspheres, sintered UO_2 pellets, and U_3O_8 are 3.1, 9.0, 5.9, and 3.0 gm/cc, respectively. Therefore, all other factors being equal, the sintered oxide microspheres would require significantly less storage area.

Additionally, environmental and cost considerations must be evaluated in assessing storage options. The primary concern for storage of depleted uranium is the integrity of the container to prevent potential releases to the environment as well as to protect the container contents for future use. The cost of the storage facility will be proportional to its size and consequently a strong function of chemical form. However, the overall cost for a particular storage option also includes the costs for conversion, inter-site transportation, and any required repackaging. Storage as UF_6 would have no associated conversion cost. Storage as UO_2 would appear to have a higher associated conversion cost than that of U_3O_8, but the storage volume would be significantly less. Accordingly, there are a variety of cost tradeoffs.

The engineering analysis for the Storage module is considering the storage of depleted UF_6 in a building and mined cavity in containers similar to those in which it is currently stored. Storage of sintered UO_2 microspheres and U_3O_8 in 30-gallon and 55-gallon drums, respectively, are being considered in the following suboptions: building, below ground vault, and below ground mined cavity.

DISPOSAL MODULE

Disposal options are defined by the disposal facility and nature of the depleted uranium waste form. The engineering analyses for the disposal module includes three disposal facilities: (1) below ground vault, (2) engineered trench, and (3) below ground mined cavity. Each disposal method is being evaluated for four different waste forms. The base case waste form being analyzed is cemented (grouted) U_3O_8, and the alternatives being evaluated are: bulk U_3O_8 (not cemented), cemented UO_2, and bulk UO_2. The UO_2 suboption is represented by sintered UO_2 microspheres (high density). The spectrum of cases reflects the differences in potential site meteorology and geology factors, and differences in chemical stability, release rates, solubility and friability characteristics. U_3O_8 has high chemical stability and low solubility under most environmental conditions. However, it is difficult to control the particle size distribution of U_3O_8 and, hence, this compound is quite friable. For this reason, and to further reduce its release rate, the base case assumes U_3O_8 mixed with cement to produce a grouted, solid product. Cementation and repackaging would occur at the waste disposal site. Uncemented U_3O_8, sealed in drums, would require a somewhat smaller disposal area thereby avoiding repackaging.

UO_2 is also insoluble, but, at ambient temperature in air, it will slowly convert to U_3O_8. Sintered UO_2 can, however, be stabilized with a density substantially greater than compacted U_3O_8. Therefore, disposal as UO_2 microspheres represents the minimal disposal area requirement for oxides.

Concrete vaults represent an improved means for LLW disposal, particularly in the eastern part of the country. The engineering analysis is evaluating a reinforced concrete vault just below grade, with the excavated material mounded above the vault and original grade as a water intrusion resistant cap. Engineered trenches are also analyzed, but are feasible primarily in the drier parts of the country, such as the western desert area.

SUMMARY

The Department of Energy has identified a broad set of technology and application options toward developing and analyzing alternative strategies in an EIS for the long-term management of depleted UF_6. The long-term management strategy alternatives fall into three broad categories of disposition: (1) use, (2) storage, or (3) disposal.

Transportation is likely to be the first step in implementing any alternative. The preparation of the depleted UF_6 cylinders at their current storage sites for transportation to an off-site facility for chemical conversion or storage is being analyzed. Conversion of the depleted UF_6 to another chemical form would also be required for most alternatives. Uranium oxides are preferred for storage and disposal alternatives, while uranium metal and the sintered (densified) oxide are preferred for applications where higher density is essential. Chemical conversion processes having a significant technical basis are being analyzed in depth (pre-conceptual designs). Radiation shielding for spent nuclear fuel containers represents the use option being analyzed in depth.

The long-term storage options being analyzed are: (1) U_3O_8, sintered UO_2, and UF_6 chemical forms and (2) above ground building storage, below ground vault storage, and below ground mined cavity storage facilities. The disposal options being analyzed are: (1) U_3O_8 (bulk and cemented) and UO_2 (bulk and cemented) forms and (2) below ground vault disposal, engineered trench disposal, and below ground mined cavity disposal facilities. The spectrum of cases reflects the differences in potential site meteorology and geology factors, and differences in chemical stability, solubility, and friability characteristics.

The transportation preparation, conversion, use, storage, and disposal options being analyzed in depth have a sufficient technical basis to allow development of meaningful data. The analysis of these options includes development of pre-conceptual designs; estimates of effluents, wastes, and emissions; specification of resource requirements; and preliminary hazards assessments. The results of this analysis will assist DOE in selecting an alternative management strategy.

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