

## **Depleted Uranium Disposition: Disposal in a High-Level-Waste Geologic Repository**

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## Depleted Uranium Disposition: Disposal in a High-Level-Waste Geologic Repository

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### INTRODUCTION

The objective of this paper is to discuss depleted uranium (DU) management options and some of the consequences of placing DU in a high-level-waste geologic repository.

The DU residues (i.e., “tails”) from enriching uranium in the United States have been stored in steel cylinders at uranium enrichment sites since World War II, first at Oak Ridge, Tennessee, and later at Paducah, Kentucky, and Portsmouth, Ohio. Many of these cylinders are beyond their expected lifetime and are in danger of leaking UF<sub>6</sub>, producing radioactive and toxic materials that could be released into the atmosphere.<sup>1</sup> At present, there are 704,000 metric tonnes of DUF<sub>6</sub> stored in 14-tonne-capacity cylinders at the three U.S. uranium enrichment gaseous diffusion plants.<sup>2</sup> The United States Enrichment Corporation (USEC) could transfer an additional 23,300 tonnes to the Paducah plant between 2002 and 2006. It is imperative that action be taken to manage this DU problem in the immediate future.

The problem of managing the DU belongs to the U.S. Department of Energy (DOE). A number of options are available. Some of the larger-scale options are interim uses of the DU in a wide variety of applications ranging from military uses in shells and armor, to uses in the nuclear industry in the fabrication of radioactive material storage containers, to fabrication of radioactive material shipping and disposal casks, to ballast in ships and aircraft. Small-scale, but potentially valuable, uses include several promising electronic applications such as in the manufacture of semiconductor devices.

The current DOE plan is to dispose of the DU as low-level-waste (LLW) in commercial LLW disposal sites such as the one operated at Clive, Utah, by Envirocare; the Nevada Test Site (NTS); or the repository in Hanford, Washington.<sup>3</sup>

Because of its chemical reactivity, DUF<sub>6</sub> cannot be disposed of directly as LLW. Consequently, two conversion plants are to be constructed, one at Paducah and the other at Portsmouth, to convert the DUF<sub>6</sub> to much more stable uranium oxides, primarily U<sub>3</sub>O<sub>8</sub>. The plants are being constructed by Uranium Disposition Services (UDS).<sup>4</sup> Ground was broken at the Paducah and Portsmouth sites during July 2004 under a \$558 million contract with DOE that runs from August 2002 until August 2010.

In addition to the management options mentioned above, the converted DU could be placed in a geologic repository. In this option, the DU need not be disposed of as LLW. It could be beneficially used in geologic spent nuclear fuel repositories in any of a number of forms. For example, the DU could be used as a material of construction for waste packages (WPs), as backfill in the form of a Richards barrier, as packing around the spent fuel elements, or in the “invert”.<sup>5</sup> In these applications the chemical, nuclear, and geochemical characteristics of DU oxide are used to improve repository performance. There are important factors to be considered in connection with this option. Some of these are presented below as observations relating to placing DU in Yucca Mountain (YM), followed by comments on the observations.

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<sup>1</sup> Upon contacting moist air, the DUF<sub>6</sub> reacts chemically to form solid UO<sub>2</sub>F<sub>2</sub> and gaseous HF.

<sup>2</sup> The DUF<sub>6</sub> is distributed as follows: 56,000 tonnes at the East Tennessee Technology Park (formerly the K-25 Plant) at Oak Ridge, Tennessee; 198,000 tonnes at the Portsmouth, Ohio, plant; and 450,000 tonnes at the Paducah, Kentucky, plant.

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<sup>3</sup> Congress has determined that an effort be made to establish beneficial uses of the DU to defray the costs of disposal as LLW. A small research and development program has been established at Oak Ridge National Laboratory to identify such uses.

<sup>4</sup> UDS is a consortium of Burns and Roe, Duratek Federal Services, and Framatome ANP.

<sup>5</sup> “Invert” is the name DOE gives to the support structure for the WPs in the YM drifts.

## OBSERVATIONS AND COMMENTS

### 1. *Observation*

DOE has announced its intention to submit a license application to the U.S. Nuclear Regulatory Commission (NRC) in December 2004 to build a repository at YM. A change, such as introducing large amounts of DU into the repository, could put DOE's date for docketing the application at risk, either from requests from the NRC for additional analyses of the effects of the DU or in the form of intervenors questioning the adequacy of the repository license application.

#### *Comment*

Unlike nuclear reactors, repositories are constructed progressively over the lifetime of the facility. Consequently, it is expected that the design of the repository will change as new technologies become available and that licensing amendments will reflect those changes.

### 2. *Observation*

Space in YM is limited and may in fact be inadequate to meet storage needs for commercial spent nuclear fuel. Addition of DU would exacerbate the problem.

#### *Comment*

There is room in YM for expansion by digging additional drifts. YM is heat limited, not volume limited. Consequently, low-heat wastes can be disposed of between, above, or below existing spent nuclear fuel disposal drifts. The need for expanded spacing of WPs containing spent fuel caused by decay heat does not exist for DU. Consequently, the DU containers could be much more closely packed than spent fuel WPs. Assuming a 50% packing fraction for the DU oxide packed in boxes, the WPs would occupy  $8.46 \times 10^4 \text{ m}^3$ . This would require excavation of eight additional drifts [ref. 1].

### 3. *Observation*

The possibility has been raised that an unacceptable radiation dose might be present at the YM site boundary because of DU in the repository.

#### *Comment*

Within a 10,000-year time frame, the peak dose rate from DU at the accessible YM environment (20 km distance from YM) is conservatively estimated as about  $3 \times 10^{-7}$  mrem/year. For 50,000 years the calculated dose rate is nearly constant at about 0.2 mrem/year, about a factor of ten lower than the

present U.S. Environmental Protection Agency (EPA)-mandated dose rate for drinking water [ref. 1]. Figure 1 compares the dose rate from DU with that from other radionuclides. The dose rate at the site boundary from DU reaches a maximum at about 30,000 years; the dose rate from all other radionuclides in the spent fuel approaches a maximum at about 500,000 years. In neither case does the dose rate exceed the EPA limit of 4 mrem/year for drinking water and 25 mrem for other types of exposure at the YM site boundary.

### 4. *Observation*

The possibility exists that an unknown but small number of DUF<sub>6</sub> cylinders contain radionuclides other than uranium isotopes and that portions of these will carry over to the DU oxides in the conversion process. In particular, there is the potential for Tc and transuranium elements, notably Np and Pu, to be present. (These radionuclides come from reenriched uranium recovered from irradiated spent nuclear fuel.) If present in the DU oxides in sufficient amounts, these radionuclides could contribute to the dose at the site boundary. The maximum total quantities of Tc and transuranium elements in the DUF<sub>6</sub> inventory are given in Table I.

#### *Comment*

It is expected that neither Tc nor transuranium elements present in the cylinders will transfer in significant amounts to the feed to the conversion plants. Filters between the cylinders and the conversion facility are expected to trap whatever (small) fraction of these elements leave the cylinders. The bulk of the radionuclides are expected to be solid compounds that remain in the cylinders.

### 5. *Observation*

There is a possibility that chemical conditions in the repository could change the composition of the oxide product of conversion, which will be primarily U<sub>3</sub>O<sub>8</sub>.<sup>6</sup> For example, because the repository will have an oxidizing atmosphere, the surfaces of the DU<sub>3</sub>O<sub>8</sub> and DUO<sub>2</sub> could oxidize. If U(VI) forms, then in the presence of moist air, the very stable and highly soluble uranyl tricarbonate complex could form. This could significantly and adversely change the amount of DU reaching the site boundary and thus the radiation dose there. Furthermore, the presence of chloride ion in the repository could lead to the formation of soluble uranium chloride complexes.

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<sup>6</sup> Based on experience, it is anticipated that the conversion facility product will be about 80% U<sub>3</sub>O<sub>8</sub> and 20% UO<sub>2</sub>.

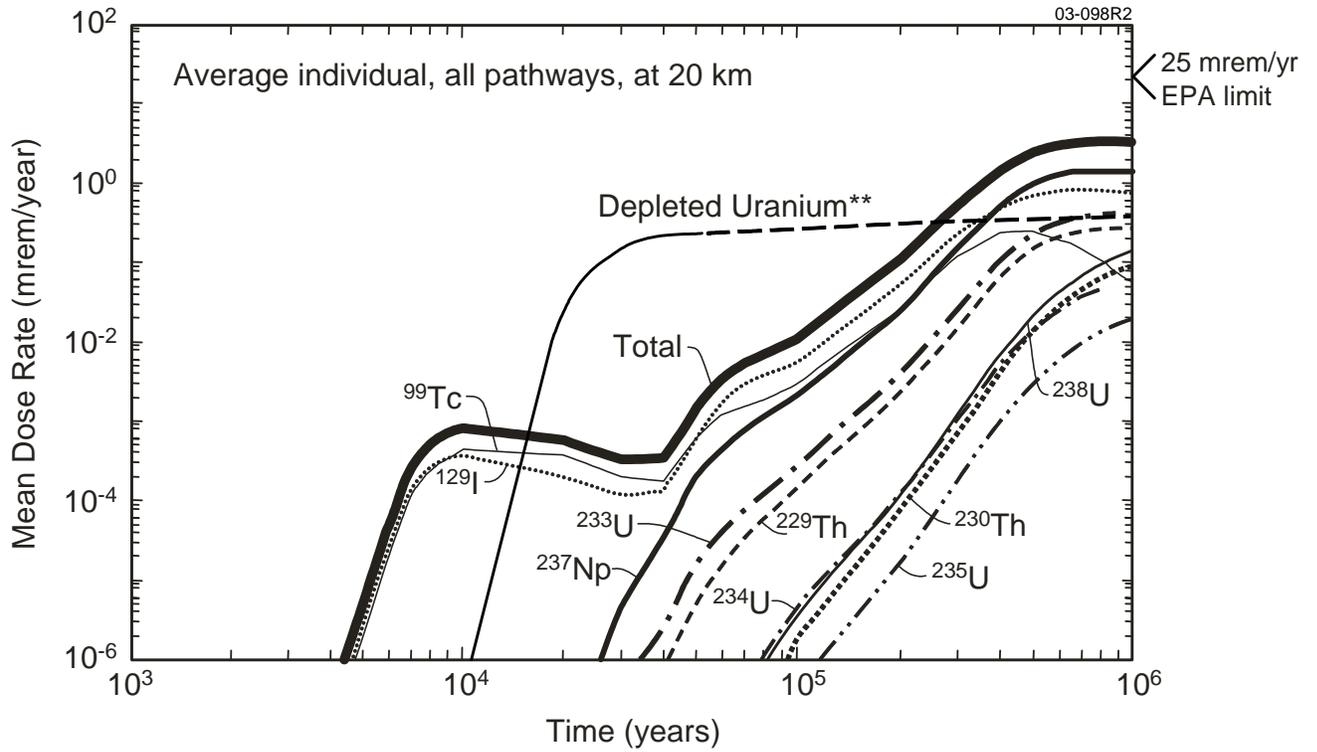


Fig. 1. Dose rate projections for Yucca Mountain Repository from spent nuclear fuel [ref. 2] and Depleted Uranium [ref. 1]

Table I. Maximum Total Amounts of Technetium and Transuranium Elements in the  $\text{DUF}_6$  Inventory

Radionuclide	Maximum Amount, g
Pu	24
Np	17,800
Tc	804,000

\*Source: [Ref. 3].

### Comment

Laboratory experiments performed with  $\text{UO}_2$  in a variety of synthetic and natural groundwaters and across a range of pH and redox conditions, as well as in the presence of a large range of chloride ion concentrations and low concentrations of carbonate ions, have shown that measured solubilities were larger than would be expected under reducing conditions. However, under pH conditions near 7 (neutral) the concentrations of total dissolved uranium species were only about  $10^{-7}$  to  $10^{-8}$  mol/dm<sup>3</sup> [ref. 4]. Within 50,000 years, the dissolved uranium concentration in YM from  $\text{DUO}_2$  is expected to be about  $10^{-8}$  mol/dm<sup>3</sup> [ref. 1]. Thus, there is no apparent reason to expect chemical species prevalent in YM to change the radiation dose at the site boundary.

### 6. Observation

Unevaluated and potentially large costs are associated with disposal of DU in YM. It might not be cost-effective to dispose of DU in YM, both because of the costs associated with changing the license application to include DU and because of the DU emplacement costs. Furthermore, YM is intended for the disposal of commercial spent nuclear fuel and irradiated fuel generated by DOE and its predecessor agencies. Changes that would be required to the YM authorizing legislation could lead to costly delays and potentially expensive lawsuits and fines related to perceived “mispending” of the electric utility surcharge of 1 mil/kWh for waste management, which is meant to provide for disposing of commercial spent nuclear fuel.

### Comment

The direct costs estimated for disposing of DU in YM are relatively small and are commensurate with the costs of disposing of the DU as LLW.<sup>7</sup> A total DOE system-wide analysis that evaluates and compares both costs and cost savings to DOE (and consequently to the U.S. taxpayer) of managing the DU in the several ways that have been mentioned in this report is highly desirable. Only in this way can the real costs be determined.

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<sup>7</sup> According to Sassani [ref. 1], the total estimated additional cost at YM for providing for DU disposal is \$205 million. This is to be compared with a potential estimated cost of disposal as LLW at the Envirocare LLW disposal facility of ~\$100 million.

## CONCLUSION

A fairly large number of options exist for managing the DU tails from uranium enrichment plants. Some of the options are interim, producing useful and valuable products but requiring ultimate disposal of the DU at a later time, perhaps many decades in the future. Other options, for example, those involving disposal of DU as LLW in commercial burial grounds or in YM, are final disposal options.

Some of the reasons that might be proposed for not disposing of large amounts of DU in YM do not appear to be well founded. Costs are the greatest area of uncertainty, and because there are many millions of dollars at stake, a system-wide analysis of potential incurred costs to DOE of the various DU management options should be performed.

## REFERENCES

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