

## **SUMMARY**

### **DEPLETED URANIUM DIOXIDE AS SNF WASTE PACKAGE FILL: A DISPOSAL OPTION**

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## INTRODUCTION

The use of depleted uranium (DU) dioxide (DUO<sub>2</sub>) particles is being investigated (Forsberg, August 2000; Forsberg, Nov. 3, 1998) as fill material inside repository waste packages (WPs) containing light-water reactor (LWR) spent nuclear fuel (SNF). The use of DUO<sub>2</sub> fill may (1) eliminate repository criticality concerns, (2) reduce radionuclide release rates from the repository, and (3) dispose of excess DU. A description of the concept is provided. The advantages of this use of excess DU as a disposal method are discussed. The potential quantities of DUO<sub>2</sub> that could be used for this application are defined for alternative WP designs.

## PHYSICAL DESCRIPTION OF CONCEPT

The WP with DUO<sub>2</sub> fill would be similar in design to that of the proposed Yucca Mountain (YM) repository WP. The WP would be first filled with SNF and then filled with DUO<sub>2</sub> particles ranging in size from 0.5 to 1 mm. The particles fill void spaces in the WP and the coolant channels within each SNF assembly. Particle size is chosen to allow efficient filling of the coolant channels. The proposed Canadian SNF WP uses a particulate fill material (but not DUO<sub>2</sub>). Canadian large-scale experiments (Johnson, April 1994) using dummy SNF assemblies and full-scale WPs have demonstrated the filling technology. The sealed WPs are placed in the repository, and then backfill is placed between the WPs and the tunnel wall.

## REPOSITORY BENEFITS

There are two types of potential benefits to the repository in using DUO<sub>2</sub> as a fill material.

### CRITICALITY CONTROL

The DU minimizes the potential for nuclear criticality. The average fissile content of LWR SNF is somewhat less than 1.6 wt % <sup>235</sup>U equivalent. This assumes that the plutonium is equivalent to <sup>235</sup>U. Coolant channels occupy most of the volume in SNF. Assuming a 65% DUO<sub>2</sub> theoretical fill density, a WP can accept ~3.3 tons of DU per ton of uranium in the SNF. If the DU has an assay of 0.2 wt % <sup>235</sup>U in <sup>238</sup>U, the average fissile content of the WP with fill will become ~0.53 wt % <sup>235</sup>U equivalent. This low fissile assay minimizes the potential for nuclear criticality (Forsberg, August 2000).

### REDUCED RADIONUCLIDE REPOSITORY RELEASE RATE

The goal of a geological repository is to contain radionuclides until the most hazardous ones decay to nonradioactive elements. The dominant failure mode of a repository is WP failure, which would then be followed by dissolution of SNF radionuclides in groundwater and ensuing movement of the groundwater to the open environment. The radionuclides are primarily incorporated into the SNF UO<sub>2</sub> pellets and cannot be released until the SNF UO<sub>2</sub> degrades. The DUO<sub>2</sub> fill material, which is in the same chemical form as is the uranium in the SNF, acts as a sacrificial material to delay the disintegration of the SNF UO<sub>2</sub>. Because the DUO<sub>2</sub> is in particulate form and the SNF UO<sub>2</sub> is partly protected by clad, the DUO<sub>2</sub> preferentially reacts with groundwater. There are four SNF protective mechanisms:

- *Chemically reducing conditions.* SNF UO<sub>2</sub> does not degrade under chemically reducing conditions. DU in the form of UO<sub>2</sub> helps maintain chemically reducing conditions in the WP.

Oxygen in groundwater reacts with  $\text{DUO}_2$  to form a mixture of  $\text{U}_3\text{O}_8$  and  $\text{UO}_3 \cdot x\text{H}_2\text{O}$ . Removal of the oxygen creates chemically reducing conditions.

- *Reduction of groundwater flow.* Under the expected YM groundwater conditions, the  $\text{DUO}_2$  fill will slowly react with the oxidized groundwater to form lower-density uranium oxides. This swelling fills void spaces and reduces the groundwater flow through the WP. The oxidation of  $\text{UO}_2$  to  $\text{U}_3\text{O}_8$  results in a 36 vol % increase and a corresponding decrease in density (McEachern, 1998). Once a low-permeability zone is created, the water is expected to flow around the WP—not through it. Simplified calculations indicate the potential to delay and reduce radionuclide releases over several million years.
- *Removal of radionuclides from groundwater.* Hydrated uranium oxides behave as inorganic ion exchangers. As radionuclides escape the SNF, some of them are absorbed onto the DU oxide fill material. Recent SNF leaching experiments show certain long-lived radionuclides such as neptunium are retained (Buck 1998) by degraded SNF. The selective radionuclide retention indicates that the uranium oxide fill will also retard radionuclide migration from the WP.
- *Saturation of the WP water with uranium.* The DU saturates water entering a failed WP with DU. This reduces dissolution of SNF uranium with accompanying releases of hazardous radionuclides.

### **ADVANTAGES OF DU DISPOSAL BY THIS OPTION**

There are several potential advantages of this option for disposal of excess DU.

#### **REGULATORY ACCEPTANCE**

The regulatory requirements for disposing of DU are unclear; however, a geological repository will clearly meet all disposal requirements. Assessments by the U.S. Nuclear Regulatory Commission (Hickey, 1992; NRC, 1994) confirm geological disposal as a desirable disposal option for DU.

#### **ENVIRONMENTALLY SAFE DISPOSAL**

The YM project has conducted a preliminary performance assessment (Owen, 1999) of adding DU as a waste (not as fill material) to the proposed repository. The analysis shows that DU disposal in the repository meets long-term waste isolation requirements.

#### **TOTAL USE OF DU**

Initial assessments (see below) indicate that this application can use some or all the DU depending upon the WP design that is selected.

#### **NO RECYCLE ISSUES**

$\text{DUO}_2$  fill is a consumptive end use of DU. There are no end-of-product-life disposal issues. DU is a heavy metal. The chemical toxicity exceeds the radiological toxicity. For any product that uses a toxic heavy metal in a non-disposal application, there are significant uncertainties on allowable future uses. Changing U.S. environmental laws are continuing to phase out the use of heavy metals in many

applications (lead paint, leaded gasoline, chrome cooling tower corrosion control, etc.). Europe is phasing out heavy metals— except where there are clearly no alternatives, or full recycle is possible (batteries).

## NUCLEAR FUTURES

If there are large quantities of SNF to be disposed of, there will be large quantities of DU that require disposal. From a long-term perspective, the world will either develop new energy sources (e.g., fusion) or deploy breeder nuclear reactors. If breeder reactors are deployed, there will be no SNF or DU to dispose of. The SNF will be processed to obtain fissile material for the breeder reactors, and the DU will be used as a fertile material. If breeder reactors are not deployed, it will be necessary to dispose of the SNF and DU. These strategic considerations suggest advantages in disposing both materials in the same location.

Two technical factors support this perspective. WPs are designed to last thousands of years. If the DU is part of the WP, the DU will be easy to recover. Second, Canadian experiments on non-DUO<sub>2</sub> fill indicate that fill materials can be separated from the SNF without serious damage to the SNF. SNF and DUO<sub>2</sub> are separable.

## CONSUMPTION OF DU

To manufacture LWR fuel, natural uranium with a <sup>235</sup>U content of 0.71 % is separated into a DU fraction and an enriched uranium fraction. The enriched uranium (3–5% <sup>235</sup>U) is fabricated into fuel. Typically, 4 to 6 tons of DU with a fissile content of 0.2–0.35% <sup>235</sup>U are produced per ton of enriched uranium nuclear fuel. Worldwide, about 47,000 tons are produced annually. Currently, DU consumption is at somewhat less than a 1,000 tons/year. About one million tons are in storage, and ~40% of that inventory is in the United States. This use of DUO<sub>2</sub> could beneficially use much (if not all) of the DU. The quantity of DUO<sub>2</sub> required for this application depends upon the design of the fill and the design of the WP. The WP consists of cylindrical metal container with an egg-crate structure within the container to hold the SNF. Four logical quantities of DUO<sub>2</sub> can be added to a WP.

- *Minimum performance benefits.* To obtain the minimum performance benefits, the coolant channels of the SNF must be filled with DUO<sub>2</sub>. If coolant channels are half empty, much of the SNF will not see any benefit from the use of DUO<sub>2</sub> fill.
- *Efficient package utilization.* After filling the SNF channels, all other void spaces in the WP can be filled with DUO<sub>2</sub>. This improves performance without changing the outer diameter of the WP as compared to a WP that does not contain fill material. The WP description above is of this option.
- *Self-shielded package.* In this concept, the package diameter and length are expanded with addition of DUO<sub>2</sub> to the top, bottom, and circumference of the WP. Sufficient DUO<sub>2</sub> is added such as to create a self-shielded WP.
- *Maximum performance benefits.* Using this approach, the WP is expanded until all available DU is used. It maximizes the performance benefits.

## CONCLUSIONS

DUO<sub>2</sub>, as a fill material in repository SNF WPs, may significantly improve the performance of the repository. Beyond repository improvements, this use of DUO<sub>2</sub> is a method for DU disposition that is capable of using the entire existing and future inventory of DU. There remain many technical and economic uncertainties. Research is continuing to reduce these uncertainties.

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## NOTE TO REVIEWER

The final paper will expand several sections. Most of the expansion will be in the last section on consumption of DU with an analysis of the quantities of DU that can be disposed of per ton of SNF for each of the four design options that are described.