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

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ABSTRACT

The use of depleted uranium (DU) dioxide (DUO_2) particles is being investigated for use in repository waste packages (WPs) containing light-water reactor spent nuclear fuel (SNF). The DUO_2 may be incorporated into the WP (1) as a particulate fill of all void spaces including the SNF coolant channels and (2) as a component of the WP structure. The use of DUO_2 may (1) reduce repository criticality concerns, (2) reduce radionuclide release rates from the repository, and (3) dispose of excess DU. The quantities of DUO_2 that could be used in the WPs are defined for alternative WP designs. The alternatives use 2.5 to 8 t of DU per ton of SNF on a uranium-metal basis. If the only change to the WP is filling all the voids inside the WP with DUO_2 particulates, about 3.5 t of DU are used per ton of SNF. This beneficial use of DU could potentially use the entire inventory of DU.

DESCRIPTION OF CONCEPT

The waste package (WP) with depleted uranium (DU) dioxide (DUO_2) fill would be similar in design to that of the proposed Yucca Mountain (YM) repository WP. The WP would be first filled with spent nuclear fuel (SNF) and then filled with DUO_2 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 (Fig. 1). 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_2). Canadian large-scale experiments using dummy SNF assemblies and full-scale WPs have demonstrated the filling technology. Added DUO_2 can be used as part of the WP wall structure. The sealed WPs are placed in the repository, and then backfill is placed between the WPs and the tunnel wall.

For repository applications, DUO_2 is the preferred form DU. In addition to the advantages of DU in the chemical form of DUO_2, as described below, there are three other considerations. First, anything added to a repository must not compromise its performance. SNF is primarily UO_2; thus, everything in the repository is designed to be compatible with UO_2. This is not true of many other chemical forms of DU. Second, the massive studies of the behavior of SNF UO_2 in a repository environment are applicable to DUO_2 repository studies. Third, DU oxides are a preferred form of uranium if the uranium is to be disposed of.

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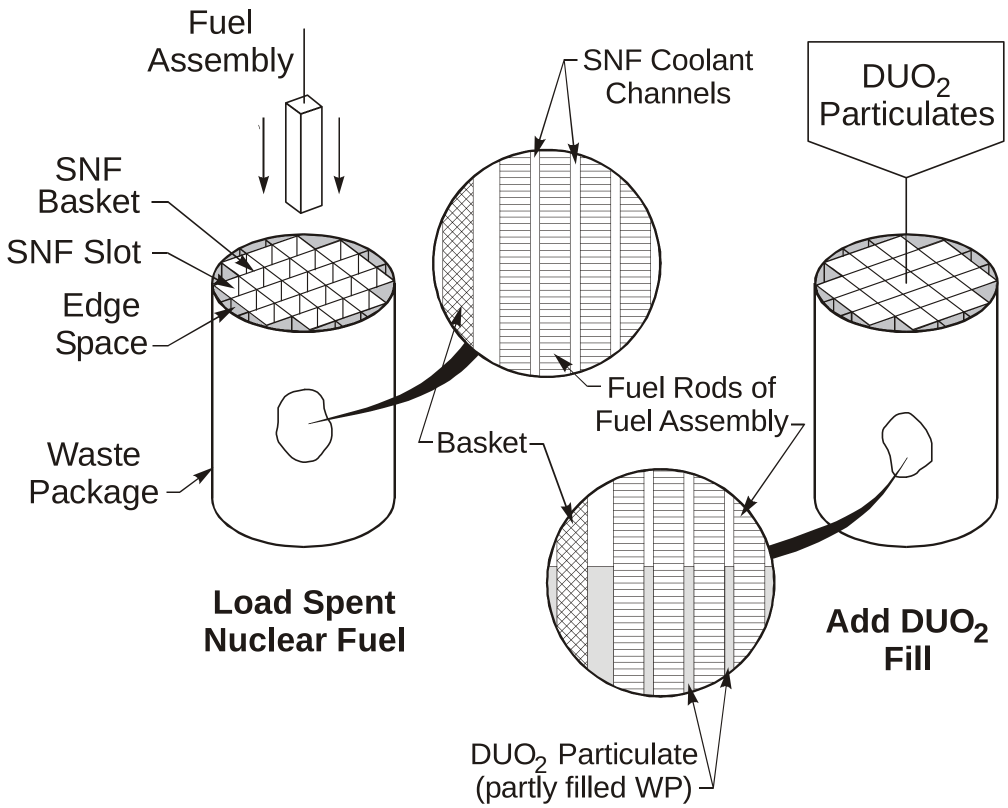

Fig. 1. Waste package loading sequence.

REPOSITORY BENEFITS

There are several potential benefits[1] to the repository in using DUO_2 as a fill material.

Criticality Control

The DU minimizes the potential for nuclear criticality. The average fissile content of light-water reactor (LWR) SNF is somewhat <1.6 wt % ^{235}U equivalent. This assumes that the plutonium is equivalent to ^{235}U. Assuming that 65 vol % of the void space is filled with solid DUO_2 and the remaining 35 vol % is composed of the spaces between individual particulates, a WP can accept -3.5 t of DU per ton of uranium in the SNF. If the DU has an assay of 0.2 wt % ^{235}U in ^{238}U, the average fissile content of the WP with DUO_2 fill will become -0.5 wt % ^{235}U equivalent. If significant mixing of the depleted and SNF uranium occurs as the WP contents degrade, this low fissile assay would eliminate the potential for nuclear criticality.

Reduced Radionuclide Repository Release Rate

The goal of a geological repository is to contain radionuclides until the most hazardous ones decay to nonradioactive isotopes. The dominant failure mode of a repository is WP and SNF failure, which would then be followed by the dissolution of SNF radionuclides in groundwater and the ensuing movement of the groundwater to the accessible environment. The radionuclides are primarily incorporated into the SNF UO_2 pellets and cannot be released until the SNF UO_2 degrades. The DUO_2 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_2. Because the DUO_2 is in particulate form and the SNF UO_2 is partly protected by the fuel pin clad, the DUO_2 should preferentially react with groundwater. There are four radionuclide isolation mechanisms[1].

- *Chemically reducing conditions*. Under oxidizing conditions, SNF UO_2 reacts with oxygen in air and groundwater to form U_3O_8 and/or $UO_3 \cdot xH_2O$. The oxidation process releases many radionuclides to the groundwater. Under chemically reducing conditions, SNF UO_2 is thermodynamically stable and the radionuclides are trapped in the UO_2 matrix. The DUO_2 particulates preferentially react with oxygen in air and groundwater. Removal of the oxygen in the WP creates chemically reducing conditions for extended times.

- *Reduction of groundwater flow*. The oxidation of UO_2 to U_3O_8, as described above, results in a 36 vol % expansion[2]. This swelling fills inter-particulate void spaces and reduces the groundwater flow through the WP. Once a low-permeability zone is created, the water is expected to flow around the WP—not through it.

- *Saturation of the WP water with uranium*. The DU saturates water entering a failed WP with DU. This reduces dissolution of SNF uranium with the accompanying reduction in release of hazardous radionuclides.

- *Removal of radionuclides from groundwater*. The fill provides (1) filtering to slow the escape of radionuclide colloids from the SNF and (2) absorption of selected radionuclides from the groundwater.

Based on the known behavior of natural uranium ore deposits, the use of DUO_2 has the potential to improve WP performance by several orders of magnitude. Field studies show that parts of natural uranium ore bodies have remained intact with oxidizing groundwater conditions nearby for geological periods of time. The outer parts of the deposits protect the masses of UO_2 inside the deposits by the mechanisms described earlier. Such natural analogs indicate the potential for excellent waste isolation using DUO_2 fill. There are significant uncertainties to be addressed.

Radiation Shielding

DU can be used to reduce the radiation levels from the WP. In a repository environment, there are two different radiation shielding options.

- *Improve repository performance*. High radiation fields can react with water and rock external to the WP to produce chemical species that degrade the WP and accelerate radionuclide migration. As a consequence, typical WPs contain some shielding to minimize these effects. This shielding is not sufficient such as to allow contact-handling of the WP.

- *Simplify operations.* The WP can contain sufficient shielding such as to allow contact-handling of the WP. This simplifies interim storage of WP until underground placement and simplifies underground operations.

DISPOSAL OF EXCESS DU

DU is a byproduct of the production of enriched uranium for commercial power reactors and defense applications. Worldwide, about 47,000 t are produced annually. Currently, DU consumption is at somewhat <1,000 t/year. About one-million metric tons are in storage with no identified uses. About 40% of that inventory is in the United States.

Most of the DU from the commercial nuclear power industry is from the manufacturing of LWR fuel. Natural uranium with a ^{235}U content of 0.711 wt % is separated into a DU fraction and an enriched uranium fraction. The enriched uranium (typically 3–5% ^{235}U) is fabricated into fuel. Table I shows the quantities of DU produced per ton of enriched uranium fuel for different product and DU assays. Typically, 4 to 6 t of DU with a fissile content of 0.20–0.35% ^{235}U are produced per ton of enriched uranium nuclear fuel. The DU ^{235}U assay depends upon the price of uranium, the price of enrichment services, and other factors.

Table I. Ratio of DU produced per unit of enriched uranium produced

	Enriched product assay (wt % of ^{235}U)			
DU assay (wt % of ^{235}U)	**2**	**3**	**4**	**5**
0.1	2.110	3.746	5.383	7.020
0.2	2.523	4.479	6.436	8.393
0.3	3.136	5.569	8.002	10.436
0.4	4.145	7.360	10.576	13.791

As a method of disposal, the beneficial use of DU in the repository has several advantages.

- *Meets disposal requirements*. Various facilities have been evaluated[3-5] for the disposal of DU. The assessments indicate that if DU is considered a waste, the proposed YM repository would meet all requirements for disposal[4-5]. Repository beneficial use of DU is consistent with disposal requirements.

- *Total use of DU.* This application can beneficially use some or all the DU, depending upon the WP design that is selected.

- *No recycle issues.* DUO_2 fill is a consumptive end use of DU that avoids potential end-of-product-life disposal issues in a changing regulatory climate.

- *Consistent nuclear futures.* From a long-term perspective, the world will either develop new energy sources (e.g., fusion) or deploy breeder reactors. If breeder reactors are fully deployed, there will ultimately be no disposal of SNF or DU. The SNF will be processed to obtain fissile material for the breeder reactors, and the DU will be used as a fertile material. If the DU is with the SNF, both can be recovered simultaneously—if needed. If breeder reactors are not deployed, it will be necessary to dispose of the SNF and DU.

  Technical characteristics of this system support this approach. First, WPs are designed to last thousands of years and thus allow simplified recovery for similar periods of time. Second, Canadian experiments on non-DUO_2 fill indicate that fill materials can be separated from the SNF without serious damage to the SNF (Forsberg 1997). SNF and DUO_2 are separable.

WP DESIGNS

Based on the functional uses of DUO_2, several preconceptual WP designs have been developed. These designs are to (1) define options and (2) determine how much DU could be reasonably consumed by this application.

The YM repository will use several types of WPs. The most common WP[6] is designed for 21 pressurized-water reactor (PWR) fuel assemblies (Fig. 2) and is used as a starting point for the concepts described herein. The WP capacity is limited by the maximum allowable decay heat per package—not mass. If the decay heat is excessive, the resulting higher temperatures might degrade repository performance. A heavier WP with DUO_2 fill could be deployed. This WP with SNF has a gross weight of 42.28 t. The repository is currently planning to accept WPs with gross weights up to -75 t.

The WP is a stainless-steel cylinder with an internal diameter of 142.4 cm and an internal length of 458.5 cm. The 5-cm-thick cylinder is covered with a 2-cm corrosion-resistant layer of C-22, a high-nickel alloy. Inside the cylindrical WP body, an egg-crate structure (called a basket) is used to hold the SNF in place. The walls that make up the basket contain multiple layers: (1) carbon steel for structural strength, (2) aluminum plates to conduct heat from the SNF to the cylinder wall, and (3) neutron absorbers to prevent nuclear criticality.

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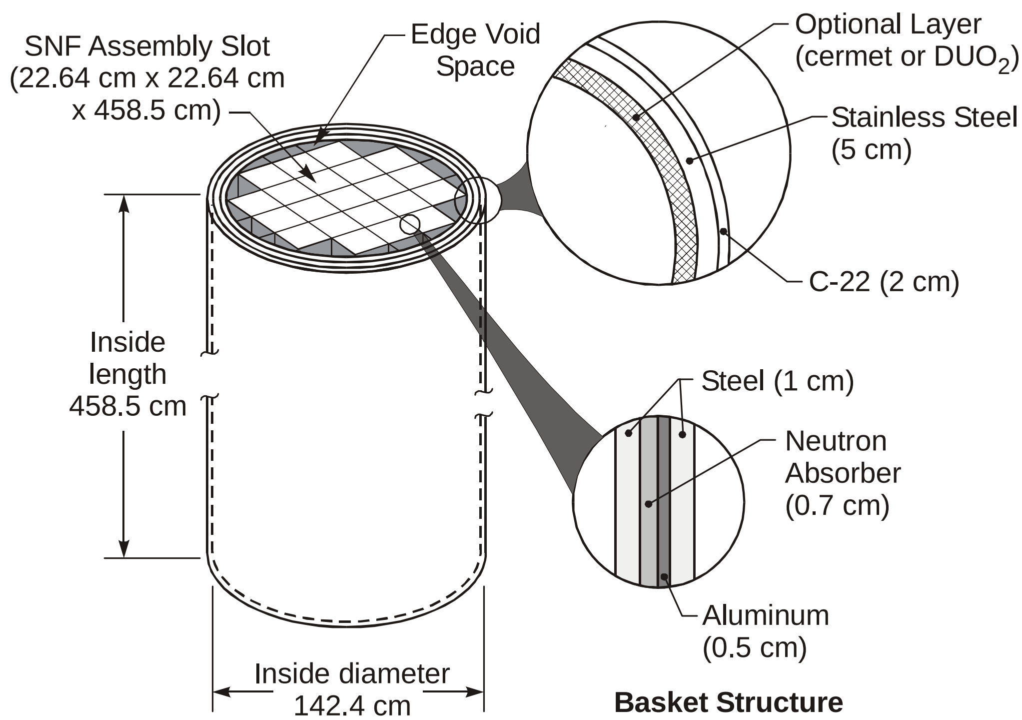

Fig. 2. 21-PWR fuel assembly waste package.

The cylindrical inside volume of the WP is 7.302 m^3. The basket solid volume is 1.119 m^3; thus, the WP void space is 6.183 m^3. This void space is divided between the 21 slots for SNF assemblies (4.935 m^3) and the edge spaces between the square grid structure and the round WP (1.248 m^3). Each basket slot has a volume of 0.235 m^3 —with the 21 slots having a total void volume of 4.935 m^3. Each slot (22.64 cm × 22.64 cm × 458.5 cm) is somewhat larger than the SNF assembly to allow for clearance during the loading of SNF. The slot length allows the WP to be used for SNF assemblies with different heights.

A typical Westinghouse, 17 ×17 pin, SNF assembly has an exterior volume (21.4 cm × 21.4 cm × 409.9 cm) of 0.188 m^3, but the solid displacement volume is only 0.07332 m^3; thus, the total solid displacement volume of the 21 fuel assemblies is 1.540 m^3. A fuel assembly is mostly empty space for water-coolant flow in the reactor; consequently, the fuel assembly void fraction is 61 vol %. This particular example of a fuel assembly has a weight of 611 kg and contains 401 kg of uranium. The loaded WP will thus contain 8.42 t of uranium in 21 fuel assemblies.

Several preconceptual designs (Table II) were developed to examine the placement of DUO_2 in the WP. No structural analysis and only limited thermal analysis has been completed. For these beneficial uses, the closer the DUO_2 is to the WP, the more effective it is. Consequently, the analysis starts with DUO_2 fill only in each SNF basket slot. Each additional design concept adds DUO_2 further out from the SNF.

Table II. Characteristics of WPs with different quantities of DU[a]

Case	WP gross wt (t)	DUO_2/WP (t)	Ratio DU to SNF (U metal)
Existing WP	42.28	0.00	0.00
Minimum use	68.97	24.19	2.53
Full void utilization	75.36	33.08	3.46
Cermet WP (self-shielding)			
100 t	100.00	49.10	5.14
125 t	125.00	65.38	6.84
DUO_2 WP (self-shielding)			
100 t	100.00	55.33	5.79
125 t	125.00	77.91	8.15

[a]The SNF in the WP contains 8.42 t initial heavy metal. The conversion factor for DUO_2 to DU is 0.881.

Minimum Use

To obtain the repository benefits, the minimum required usage of DUO_2 is to fill the WP basket slots. This results in DUO_2 particulates in (1) the coolant channels of the SNF, (2) the spaces between the SNF assembly and the slot walls, and (3) the spaces above and below the SNF assembly. The available volume (3.395 m^3) is the volume of the 21 WP slots (4.935 m^3) minus the solid displacement volume of the 21 SNF assemblies (1.540 m^3). Given the density of UO_2 (10.96 g/cm^3) and a typical fill efficiency (65 vol %), the WP will accept 24.19 t of DUO_2. A total of 2.53 t of DU are disposed of per metric ton of SNF on a uranium basis.

For the fill to perform satisfactorily, another fill material is needed for the 1.248 m^3 of void spaces between the square grid structure and the basket structure. This is required so that DUO_2 does not escape from its position next to the SNF and move to these void spaces as the WP degrades. It is assumed that a particulate fill with a density of 2 g/cm^3 is used to eliminate this problem. This adds 2.53 t of material. The total WP weight becomes 68.97 t.

Within the constraints of obtaining performance benefits from using DUO_2, this option maintains the same exterior geometric dimensions while minimizing WP weight.

Full Void Use

This option is identical to the previous option except that all void spaces in the WP are filled with DUO_2. In addition to filling the basket slots, the edge spaces between the square grid structure and the cylindrical WP body are also filled with DUO_2. It maximizes DUO_2 usage within the existing WP geometric envelope. The total void volume of this edge space is 1.248 m^3. It allows the addition of 8.89 t of DUO_2 per WP. A total of 3.46 t of DU are disposed of per ton of SNF on a uranium-metal basis.

Cermet WP

The cermet WP adds a layer of cermet between the basket structure and the exterior WP shell[7]. The WP shell diameter must be expanded to make room for the cermet. The WP shell thickness is assumed to be unchanged. As in the previous case, all void spaces in the basket are filled with DUO_2. The cermet consists of DUO_2 particulates (ρ = 10.96 g/cm^3) embedded in a steel (ρ = 7.86 g/cm^3) matrix. The cermet is assumed to contain 65 vol % UO_2 and 35 vol % steel. Its density would be 9.88 g/cm^3. The ratio of DUO_2 to steel depends upon the design objectives. This high loading is chosen to maximize DUO_2 usage. Lower DUO_2 loadings will increase the strength of the cermet.

Two WP gross weight limits are considered: 100 and 125 t. These weight limits are chosen because there is a large experience base in handling SNF shipping casks of this size. In addition, there is significant experience in handling 100-t packages in some underground disposal facilities such as the Swedish Final Repository for reactor waste facility. There is little experience above these weight limits.

With the 100-t WP, the cermet provides sufficient radiation shielding[8] such as to make the WP a self-shielded WP—except for neutron shielding. An earlier study[9] examined a transport-disposal package for 21 PWR SNF assemblies using a different DU fill. One observation from that study was that for large WPs, the gross weights of optimized self-shielded WPs—with and without DU fill—are close to each other. The use of fill adds weight but reduces the required shield thickness in the WP walls.

With a 100-t weight limit, 24.67 t (100 t - 75.36 t) of material can be added to the WP that already contains DUO_2 fill in all void spaces. This added weight is divided into (a) 22.22 t of cermet (a 9.5-cm-thick layer around the basket structure that contains 16.02 t of UO_2 and 6.20 t of steel) and (b) 2.42 t of additional stainless steel and C-22. Because the cermet layer increases the diameter and height of the WP, additional metal is required for the exterior WP metal shell. A total of 5.14 t of DU are disposed of per ton of SNF on a uranium-metal basis.

With a 125 t weight limit, 49.64 t (125 t - 75.36 t) of material can be added to a WP that already contains DUO_2 fill in all void spaces. This added weight is divided into (a) 44.78 t of cermet (a 19.12-cm-thick layer around the basket structure that contains 32.30 t UO_2 and 12.48 t of steel) and (b) 4.86 t of additional stainless steel and C-22. Because the cermet layer increases the diameter and height of the WP, additional metal is required for the exterior WP metal shell. A total of 6.84 t of DU are disposed of per ton of SNF on a uranium metal basis.

The design herein adds the cermet to an existing WP. There are also options to use the cermet to replace the existing WP structural components. This could include both the WP and the basket structure. Cermets have significant structural strength.

DUO_2 WP

This WP is similar to the cermet WP except that the cermet is replaced by a thick layer of DUO_2 blocks. Compared to the cermet options, the blocks allow higher DUO_2 loadings—the metal component of the cermet is replaced by DUO_2. Because of the unavoidable porosity in UO_2 blocks and spaces between ceramic blocks, an average DUO_2 density (10 g/cm^3) somewhat less than a typical fuel pellet is assumed. There are uncertainties about whether solid DUO_2 could be used as a shield material. The issue of cracking must be addressed. The DUO_2 does not add strength to the WP; thus, there are also structural issues that may require added steel and less DUO_2.

With a 100-t weight limit, an additional 24.64 t of material can be added to a loaded WP that already contains DUO_2 fill in all void spaces. This added weight is divided into (a) 22.25 t of DUO_2 (a 9.4-cm-thick layer around the basket structure) and (b) 2.39 t of additional stainless steel and C-22. Because the DUO_2 layer increases the diameter and height of the WP, additional metal is required for the exterior WP metal shell. A total of 5.79 t of DU are disposed of per ton of SNF on a uranium metal basis.

With a 125-t weight limit, an additional 49.64 t of material can be added to a loaded WP that already contains DO_2 fill in all void spaces. This added weight is divided into (a) 44.83 t of DUO_2 (a 18.9-cm-thick layer around the basket structure) and (b) 4.81 t of additional stainless steel and C-22. Because the DUO_2 layer increases the diameter and height of the WP, additional metal is required for the exterior WP metal shell. A total of 8.15 t of DU are disposed of per ton of SNF on a uranium metal basis.

CONSUMPTION

The DUO_2 consumption for various WP designs can be compared to existing and future DU inventories. For representative LWR fuel cycles, about half the existing and future DU inventory can be used as a fill material in the void spaces in proposed WPs. All of the inventory can be used if added DUO_2 is used in the WP walls.

CONCLUSIONS

DUO_2, as a fill material in repository SNF WPs, may significantly improve the performance of the repository. Beyond repository improvements, this use of DUO_2 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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