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Preparation Date: June 24, 2003 File Name: Integrated.Fuel.Cycle.HiltonHead.June14.2003 Due Date: July 25, 2003

Paper Prepared for Advances in Nuclear Fuel Cycle Management III Conference Hilton Head Island, South Carolina October 5–8, 2003

> Subject Categories Generation-III Fuel Cycle Design Concepts Generation IV Fuel Cycle Design Concepts

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^{*}Oak Ridge National Laboratory, managed by UT-Battelle, LLC, for the U.S. Department of Energy under contract DE-AC05-00OR22725.

AN INTEGRATED ONCE-THROUGH FUEL CYCLE WITH DEPLETED-URANIUM-DIOXIDE SNF MULTIFUNCTION CASKS

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Keywords: Depleted-uranium dioxide, Spent nuclear fuel, Cask

ABSTRACT

An advanced once-through nuclear fuel cycle is proposed that integrates (1) frontand back-end fuel cycle operations and (2) management of all long-lived radionuclides. The front-end of the fuel cycle remains unchanged except that the depleted uranium (DU) from the enrichment facilities (in the form of DU dioxide—DUO₂) is used to produce multifunction shielded casks. Several years after discharge, spent nuclear fuel (SNF) is placed in multifunction casks at the reactor and the casks are then sealed. The shielded casks are used for SNF at-reactor storage, transport, and disposal without ever being opened, thus minimizing SNF handling. Similar casks are used for disposal of greaterthan-class-C wastes.

The multifunction cask system disposes of all long-lived radionuclides from the fuel cycle—including the DU. The cask (1) provides high levels of SNF protection against assault and diversion and (2) allows the option of relatively easy recovery of SNF (with the DU) from the repository by future generations—if required. The addition of the DU reduces the maximum long-term repository radiation doses to individuals by isotopically diluting the high-alpha uranium isotopes in the SNF and other mechanisms.

1. INTRODUCTION

In the development of nuclear energy, it was initially thought that uranium resources were extremely limited; thus, early fuel cycles assumed recycle of SNF. With the discovery of large low-cost uranium resources, the light-water reactor (LWR) fuel cycle evolved into a once-through fuel cycle. As identified in the international Generation IV

fuel cycle evaluations¹, the potential exists for improved once-through fuel cycles by designing (rather than evolving) a once-through fuel cycle. SNF recycle may not be required for the initial large-scale global deployment of nuclear power. Conventional economic uranium resources may be available for a century or more. Because of these factors, it is appropriate to consider how to improve the once-through fuel cycle for its use in a large global nuclear future. One option is described herein.

2. REQUIREMENTS

There are several requirements for a robust, no-regrets once-through fuel cycle. First, the future is uncertain; thus, DU and SNF should be retrievable to allow (1) SNF and DU recycle if it becomes economic or (2) destruction of long-lived radionuclides in the SNF if a societal decision is made to minimize long-term waste toxicity or weapons-usable fissile inventories. Second, SNF contains (1) fissile materials that can be used to construct nuclear weapons and (2) radioactive materials that can be used to construct radiological weapons. Therefor, the SNF should be handled the minimum number of times to minimize the risks of diversion and should be in a secure system that makes assault or theft extremely difficult. Third, the system should address the disposal of all long-lived radionuclides. This includes SNF, radiation sources used for a variety of applications, reactor components with significant concentrations of long-lived radionuclides, and DU. Last, total costs should be minimized.

3. SYSTEM DESCRIPTION

The proposed system consists of a multifunction cask (constructed of a DUO_2 -steel cermet), a storage overpack, a transportation overpack, and a repository overpack (Fig. 1). The cermet consists of DUO_2 particulates embedded in a steel matrix (see below). The overpacks address conflicting storage and disposal requirements that cannot be easily met by the multifunction cask. For management of SNF, the system consists of the following components.

- *Multifunction cask.* The cask performs several functions: (1) serving as a handling package for SNF from the reactor to the repository and during any future retrieval operations, (2) providing primary radiation shielding, (3) offering physical protection against assault and accidents, and (4) functioning as a sealed safeguards package.
- *Dry storage overpack.* Conflicting design requirements apply to high-capacity cask storage of short-cooled SNF and disposal of SNF. With storage of short-cooled SNF, the primary design constraint is the need to avoid high temperatures that would degrade the SNF. Storage casks require a high ratio of surface area to volume (small casks or fins) to dissipate heat from short-cooled SNF. For disposal, the primary design constraint is to ensure long-term waste-package (WP) integrity. (Decay heat

levels are lower.) The WP should have a low ratio of surface area to volume to minimize both (1) the interactions between groundwater and the WP and (2) the cost of expensive corrosion-resistant materials in the repository overpack. This requirement implies a bare cylinder with smooth surfaces. The use of a removable overpack with heat removal features (fins) during storage can resolve these conflicting performance requirements.² The overpack is used at the reactor but removed before shipment of the SNF. Because the overpack is removed before shipping, the at-reactor package size and weight constraints are limited only by the reactor site constraints, not by transport or disposal requirements. The overpack may also be used to provide added shielding for the short-cooled SNF with its higher radiation levels that decrease with time. Some commercial SNF casks use bolts and other mechanisms to fasten fins to casks; thus, the technology for removable overpacks is available.

- *Transportation overpack.* The overpack provides any additional protection required for the specific challenges of transport.
- *Disposal overpack.* The proposed Yucca Mountain (YM) WP has an inner container for the SNF and an outer container of a corrosion-resistant alloy. The current inner container provides the structural support for the corrosion-resistant overpack but does not provide full radiation shielding. In this system, a multifunction cask replaces the inner container. The overpack is placed over the multifunction cask at the repository.

The disposal of DUO_2 is achieved by its use as part of the cask material of construction. For greater-than-class C wastes and other wastes for which repository disposal is preferred, the same type of cask would be used. Depending upon the characteristics of these wastes, the repository overpacks used for SNF may or may not be required.

4. CERMET CASKS

The multifunction casks are constructed of a DUO_2 -steel cermet (Fig. 2) consisting of DUO_2 ceramic particulates and other (graphite, silicon carbide, etc.) particulates, if needed, embedded in a steel matrix between two clean layers of steel. Half or more of the volume of the cask wall would be DUO_2 . A nominal 21-pressurized-water-reactor (PWR) SNF cask would contain ~10 tons of SNF and 50 tons of DU. The enrichment process produces 5 to 7 tons of DU for every ton of enriched uranium SNF; thus, most of the DU would be used in construction of the casks. (Current applications such as munitions and specialized shielding consume very small quantities of DU compared to the growing U.S. inventory that exceeds 500,000 tons.)





Fig. 1. DUO₂-steel cermet multifunction cask system.

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Fig. 2. Cermet cask cross section.

Cermet SNF casks have the potential for high performance compared with casks constructed with traditional materials because of the fundamental characteristic of a cermet: the encapsulation of different ceramic particulates into a continuous highintegrity metal matrix. Ceramic components have outstanding properties to address specific cask requirements. The DU is a high-performance gamma-radiation shielding material. The oxygen, carbon, silicon, and other additives provide enhanced neutron shielding. Hard ceramics, such as aluminum oxide, provide for very high resistance to assault. However, ceramics have poor physical properties. The traditional weaknesses (low thermal conductivity, low ductility) of ceramics are avoided in the cermet by use of the metal matrix.

For given size or weight constraints, maximizing shielding performance maximizes the quantities of SNF that can be placed within a cask. The DUO_2 -steel cermets may be the highest-performance shielding material³ that can be used in a multipurpose cask. Cermets provide better gamma shielding than steel because DUO_2 (10.9 g/cm³) has a higher density than steel (7.8 g/cm³). Because of the high oxygen content associated with the DUO_2 , which moderates neutrons, cermets also have better neutron shielding capabilities than steel. The cermet would include a neutron absorber such as gadolinium oxide for absorption of thermal neutrons. More-efficient cask shielding materials are prohibited by one or more of the storage, transport, or disposal criteria: (1) no chemically reactive materials, such as uranium metal; (2) no materials that may adversely impact repository performance, such as concretes or organics; (3) no high-cost materials, such as tungsten; and (4) no Resource Conservation and Recovery Act (RCRA) metals, such as lead.

The traditional applications for cermets include (1) military tank and vault armor, (2) vehicle brake shoes, (3) cutting tools, and (4) nuclear fuel in some test reactors. As these applications demonstrate, cermets can be made to survive extreme environments. Consequently, the option exists to design the cermet to create a super cask⁴ capable of withstanding extreme events compared to casks constructed of traditional materials.

Two changes⁵ in cermet technology have created the potential for low-cost multipurpose casks: (1) a new lower-cost fabrication method has been invented to fabricate 100-ton cermet casks, and (2) the massive growth in use of powder metallurgy techniques for manufacture of auto transmissions and other products has drastically reduced the costs of the powdered steel required for cermet manufacture.

5. REPOSITORY OPERATIONS AND PERFORMANCE

5.1 Repository Operations

Multifunction casks simplify repository surface operations. To ensure long-term performance of the WP, there are limits on the peak temperatures within each WP. The peak temperature occurs after underground placement of the WP. Heat transfer from the WP through the earth to the atmosphere is poor. The peak temperatures are controlled by imposing decay-heat limits on each WP. WP heat loads at the proposed YM repository are to be controlled by selection of high-decay-heat and low-decay-heat SNF for each WP, which involves sorting and handling of the SNF. In a multifunction cask system, casks with high heat loads would be stored until the decay heat in each cask has decreased to the allowable repository WP limits. Thus storage, not SNF sorting and handling, would be used to control heat loads of WPs sent underground.

The shielded multipurpose cask simplifies underground repository WP placement.⁶ All operations are contact handled. The DU and SNF are also collocated to ease recovery if needed by future generations. If WP recovery is initiated, the shielded cask avoids both the need for remote-operated equipment and the need to build a storage facility for recovered SNF. The WP itself is the storage cask.

5.2 Disposal of Non-SNF Wastes

The recent U.S. National Academy of Sciences study⁷ identifies the uncertainties associated with the requirements for disposal of large quantities of DU and greater-thanclass-C wastes. The U.S. Nuclear Regulatory Commission⁸ has recommended deep mine or equivalent disposal for DU. While uncertainties remain about the minimum requirements for disposal of these wastes, all of the studies indicate that geological repository disposal would meet or exceed disposal requirements for these wastes.

5.3 Repository Performance

The objective of a repository is to delay and retard the releases of radionuclides to the environment until most have decayed to non-radioactive elements and present no significant hazard to the public. The WP can slow or accelerate the long-term release of radionuclides from the repository. Consequently, repository performance of the WP is a key issue in the adoption of any multi-function cask. Figure 3 shows a recent estimate⁹ of expected radiation doses to an individual at the site boundary (maximum radiation exposure) versus time for the proposed YM repository and the radionuclides of most concern. WPs of DUO₂ cermets can potentially reduce radiation exposures by several mechanisms.

Maintaining SNF Integrity. Most radionuclides in SNF are trapped in the SNF UO_2 sintered-crystalline matrix. The radionuclides cannot escape until the SNF UO_2 is oxidized, the radionuclides are exposed to groundwater, and the radionuclides are transported by groundwater. There are two sources of oxygen to destroy the UO_2 crystal structure.

- *External to the WP*. The planned YM repository has an oxidizing groundwater and atmosphere (the repository level is above the water table) that can provide the oxygen to rapidly oxidize SNF after WP failure. In the proposed YM repository, oxygen from the groundwater and atmosphere control the rate of oxidation of SNF after WP failure.
- *Internal to the WP*. After WP failure, groundwater will enter the WP. Radiolysis of water generates hydrogen and oxygen. Almost all the oxygen and hydrogen recombine back into water. However, hydrogen diffuses more rapidly away from the SNF than the oxygen and thus can leave excess oxygen to oxidize the SNF.



Fig. 3. Projected peak radiation exposures from YM vs time.

The long-term degradation of the cermet [7 to 10 tons of cermet (iron and DUO_2) per ton of SNF] WP by groundwater (1) helps maintain a chemically reducing environment around the SNF UO_2 and thus prevents its oxidation and (2) tends to form impermeable layers that slow oxygen transport from air and groundwater through the outer rind of the sacrificially degrading WP to the SNF. As the iron in the cermet is oxidized, it removes the oxygen from the local environment. The iron is preferentially oxidized before DUO_2 . After oxidation of the iron in the cermet, the smaller DUO_2 particulates tend to be preferentially oxidized before the SNF because of their high surface area. Evaluation of

natural analogs and theoretical calculations indicate¹⁰ the potential to delay radionuclide releases from a failed WPs for hundreds-of-thousands to millions of years from the oxidizing groundwater and air in the repository. This is similar to the estimated times for oxidation of the SNF by oxygen from radiolysis of water¹¹ from inside the WP. This may allow time for (1) decay of most of the ⁹⁹Tc ($T_{1/2} = 2 \times 10^5$ y) and (2) partial decay of ¹²⁹I and ²³⁷Np. Furthermore, the sequestration of these radionuclides in the WP degradation products will retard and spread the release of these radionuclides over long periods of time, such that it is expected to significantly reduce peak radiation exposures. Ongoing work is under way to better quantify this effect.

Slowing Neptunium Migration. Current assessments indicate that neptunium is the single most important radionuclide in terms of long-term radiation dose to the public from the repository. There is experimental evidence that neptunium will sorb onto the surfaces of various hydrated uranium oxides. Thus, the addition of large quantities of DU would be expected to slow neptunium releases to the environment. However, many uncertainties remain. Therefore, a scientific program lead by Oak Ridge National Laboratory and the Institute of Chemical Physics of the Russian National Academy of Sciences has been initiated to quantify these effects.

Limiting Radiation Doses from High-Alpha Uranium Isotopes. In the long term, the SNF uranium and associated decay products can become significant contributors to the radiation dose to the public. The alpha activity (Table 1) of uranium isotopes such as ²³³U and ²³⁴U per unit mass are more than four orders of magnitude greater than ²³⁸U, the primary uranium isotope in DU. While the concentrations of these isotopes are low in natural and DU, significant concentrations of these higher activity uranium isotopes exist in SNF (Table 2). For example, ²³⁸U decays to ²³⁴U; thus in natural uranium the ²³⁴U is in sequar equilibrium with ²³⁸U. There are equal number of curies of both isotopes per ton of uranium. However, in SNF the peak radioactivity of ²³⁴U (at ~1,000 years) is approximately a factor of ten greater than ²³⁸U. This elevated concentration is a consequence of the enrichment process that selectively enriches the fresh LWR fuel with ²³⁴U and the decay of ²³⁸Pu and other isotopes in the SNF to ²³⁴U. The concentration of ²³⁴U initially increases after SNF discharge from the reactor. The actinides (such as plutonium, americium, and curium) decay to uranium isotopes. SNF uranium is much more radioactive than natural or DU.

Isotope	Half life (years)	Alpha activity (nCi/g)
U-233ª	1.62×10^{5}	9.48×10^6
U-234 ^b	2.48×10^5	$6.17 imes 10^6$
U-235	$7.13 imes 10^8$	2.14×10^{3}
U-236	2.39×10^{7}	6.35×10^4
U-238	4.48×10^9	3.35×10^{2}

Table 1. Uranium isotopic characteristics

^aPrimarily from decay of ²³⁷Np

^bWith higher-burnup SNF, primarily from decay of ²³⁸Pu

Time (years)	U-233 (curies)	U-234 (curies)	U-235 (curies)	U-236 (curies)	U-238 (curies)
0	2.991 × 10 ⁻⁵	1.223	1.900 × 10 ⁻²	4.024×10^{-1}	3.093×10^{-1}
10	5.440×10^{-5}	1.400	1.900×10^{-2}	4.026×10^{-1}	3.093×10^{-1}
30	1.037×10^{-4}	1.716	1.901 × 10 ⁻²	4.030×10^{-1}	3.093×10^{-1}
100	2.993×10^{-4}	2.499	1.904 × 10 ⁻²	4.043×10^{1}	3.093×10^{-1}
300	1.059×10^{-3}	3.343	1.911 × 10 ⁻²	4.081×10^{1}	3.093×10^{-1}
1,000	7.316×10^{-3}	3.557	1.949×10^{-2}	4.259×10^{1}	3.093×10^{-1}
3,000	2.292×10^{-2}	3.538	2.023×10^{-2}	4.562×10^{-1}	3.093×10^{-1}
10.000	7.790×10^{-2}	3.475	2.250×10^{-2}	5.236×10^{-1}	3.093×10^{-1}
30,000	2.255×10^{-1}	3.302	2.704×10^{-2}	5.774×10^{-1}	3.093×10^{-1}
100,000	6.447×10^{-1}	2.765	3.217×10^{-2}	5.836×10^{-1}	3.094×10^{-1}
300,000	1.279	1.706	3.295×10^{-2}	5.802×10^{1}	3.094×10^{-1}
1,000,000	1.428	5.033×10^{-1}	3.293 × 10 ⁻²	5.683×10^{-1}	3.095×10^{-1}

Table 2. Uranium SNF radioactivity vs time for 1-ton of PWR SNF

*Uranium isotopics for 1 metric ton of 17-by-17 PWR SNF with a burnup of 50,000 MWd/t, capacity factor of 0.85, and initial enrichment of 4.5%.

The peak radiation exposures to individuals from uranium isotopes in the repository can be reduced by adding DU to the WP. If DU is added to the WP, the SNF uranium and DU will mix as the WP degrades and ultimately migrate together in the groundwater. Because the migration of uranium from a repository is limited by uranium solubilities in the contacting groundwater within the degraded WP that transports uranium to humans, this isotopic mixing reduces the peak radiation doses to future generations by two mechanisms.

- *Dose dependency isotopics*. The quantity (in grams) of uranium and decay products an individual is exposed to is limited by the uranium solubility in groundwater. However, the ultimate radiation dose depends upon the isotopics of the uranium. If the alpha activity of the uranium is reduced by a factor of 5 via isotopic dilution of SNF uranium by DU, the radiation exposure is also reduced by a factor of 5.
- *Time delays*. In a solubility-limited system, the added DU spreads the migration of uranium over time, thus providing more time for the shorter-lived uranium isotopes to decay before reaching the open environment.

Reducing the potential for long-term nuclear criticality. As the SNF degrades, the concentrations of the fissile isotopes ²³³U and ²³⁵U increase. This is a consequence of decay of higher actinides, such as ²³⁷Np and ²³⁹Pu. With the degradation of the WP and SNF, the uranium migrates with the groundwater. This creates the potential to create deposits of uranium such that nuclear criticality might occur. The addition of DU to the WP adds sufficient ²³⁸U to isotopically dilute uranium isotopes and reduce the potential for long-term nuclear criticality.

6. OTHER CONSIDERATIONS

The cermet multifunction cask system⁴ has the potential to significantly reduce the risks of theft, diversion, and assault. The highest risks associated with SNF are during handling operations. In a multifunction cask system, the SNF is handled only once—during SNF transfer to the multifunction cask, which is then sealed. The weight and size of a 100-ton cask minimize the risk of theft compared with that associated with individual SNF assemblies. The cermet construction of the cask also maximizes resistance to assault. Cermets are the traditional materials used in military armor.

Economic studies are under way to understand the impacts of these fully integrated systems. The potential for major improvements in economics is based on two factors.

• *Multifunction cask system*. A single multipurpose cask system that is used for all long-lived wastes avoids the complexities of designing, siting, licensing, and building multiple systems to dispose of different long-lived wastes (SNF, DU, Greater-than-

class-C wastes). With the management of SNF, the multipurpose cask system (1) avoids constructing SNF storage and handling facilities at both reactors and at the repository and (2) multiple handling of SNF.

• *DUO*₂-*steel cermet*. Construction of the multifunction cask using a DUO₂-steel cermet simultaneously disposes of the DU, improves cask resistance to external events, and increases cask capacity within any given size and weight constraints.

7. CHALLENGES

The viability of the system depends upon successfully addressing three issues.

- *Cask fabrication*. Analysis indicates that the new cask fabrication method (patents filed in January 2003) has the potential for low costs; however, development of the fabrication technology is only beginning. Actual fabrication of casks is required before there will be a high level of confidence in the manufacturing economics.
- *Repository performance*. The initial repository assessments and evaluations of DU have been favorable; however, a detailed repository performance assessment is required. This requires additional data and models to better understand the multiple impacts of DU on repository performance.
- *Institutional structures*. Different organizations are responsible for SNF storage, SNF disposal, DU storage and disposal, and greater-than-class C (radiation sources and highly irradiated hardware) waste storage and disposal. At various times, different organizations have responsibility for SNF safeguards and security. Minimizing the costs of any one component of the system is not the same as minimizing the costs of the total system. To implement such a system requires (1) the cask and system designs be integrated and (2) the organizations purchasing the multipurpose casks receive some of the savings obtained elsewhere in the system.

8. CONCLUSIONS

The current waste management system evolved from a system designed for recycle of SNF into a once-through fuel cycle. The disposal of some waste, such as DU, has not been fully addressed. Security and safeguards requirements for SNF are changing. These factors support the need to consider an integrated once-through fuel cycle to maximize future options, improve safeguards and security, improve waste management, and reduce costs. An integrated DUO₂-steel cermet multipurpose cask system may create such an option. Integrated systems will require both development of appropriate technologies and changes in the corresponding institutional structures.

ACKNOWLEDGMENTS

This work was done under the auspices of the U.S. Department of Energy Depleted Uranium Uses Research and Development Program.

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