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ABSTRACT

Multipurpose transport, aging, and disposal casks are needed for the management of spent nuclear fuel (SNF). Self-shielded cermet casks can outperform current SNF casks because of the superior properties of cermets, which consist of encapsulated hard ceramic particulates dispersed in a continuous ductile metal matrix to produce a strong high-integrity, high-thermal-conductivity cask.

A multiyear, multinational development and testing program has been developing cermet SNF casks made of steel, depleted uranium dioxide, and other materials. Because cermets are the traditional material of construction for armor, cermet casks can provide superior protection against assault. For disposal, cermet waste packages (WPs) with appropriate metals and ceramics can buffer the local geochemical environment to (1) slow degradation of SNF, (2) reduce water flow though the degraded WP, (3) sorb neptunium and other radionuclides that determine the ultimate radiation dose to the public from the repository, and (4) contribute to long-term nuclear criticality control. Finally, new cermet cask fabrication methods have been partly developed to manufacture the casks with the appropriate properties. The results of this work are summarized with references to the detailed reports.

INTRODUCTION

Spent nuclear fuel (SNF) casks are needed for storage, transport, and disposal. These casks may be designed for single or for multiple applications [1]. The functional requirements for SNF casks include (1) radiation shielding; (2) cooling of the SNF to limit its peak cladding temperatures; (3) physical protection; and (4) for disposal waste packages (WPs), delay of the radionuclide migration for long periods of time. Such casks may contain the individual SNF assemblies or canisters that contain multiple SNF assemblies. For example, in the United States, repository planning includes the development of a transportation, aging, and disposal (TAD) canister system. In such a system, a cermet-shielded cask might contain a TAD canister. Alternatively, the cask itself may be considered a shielded TAD canister, requiring minimal recycled overpacks to meet the specific requirements of storage, transport, and disposal.

Overall cask performance is ultimately determined by the interactions and collective performances of cask materials of construction with the SNF and the storage, transport, and disposal environments. Shielded cermet casks have the potential to outperform all other SNF casks because cermets enable the incorporation of brittle ceramics with highly desirable properties within a high-thermal-conductivity strong ductile metal matrix. Figure 1 shows cermet cask construction. With advanced cermet manufacturing technologies, the cermet properties can be varied through the thickness of the cask wall to optimize material properties.

Cermet casks have become a viable cask option because of several technical developments: (1) new methods to fabricate cermet casks, (2) increased emphasis on physical protection against terrorist attack, (3) the availability of depleted uranium dioxide (DUO_2) for use in casks, and (4) an understanding of what the material composition of a WP should be in order to create local geochemical conditions that minimize the long-term radionuclide migration. A multiyear multinational program [2] has been investigating the design, construction, and performance of cermet casks. A summary of results and references to the appropriate papers are reported herein.

MANUFACTURE

Because of their hardness and durability, cermets are materials of choice for use in drill bits, armor, brake disks, and other uses in extreme environments. The properties that make cermets useful also make them difficult to fabricate; cermets are difficult to machine and very difficult to weld in thick sections. Because an SNF cask is larger than other cermet products, the commercial viability of a cermet cask strongly depends upon developing methods of cask fabrication. Two methods of powder-metallurgy fabrication and one method to cast DUO_2 casks have been partly developed to fabricate large cermet casks without requiring machining or welding of the cermet. All of the methods avoid welding of cermets.

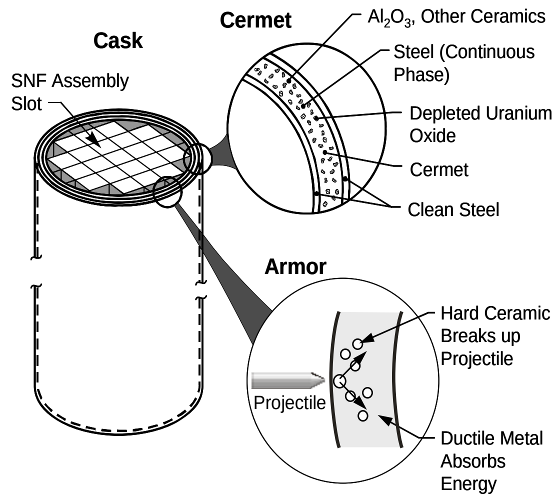

Figure 1. Cermet cask construction.

In the powder metallurgy processes, a mixture of iron and ceramic powders is ultimately converted into a high-integrity SNF cask. The Forge Cermet Cylinder (FCC) process [3] produces the cylindrical cask body in a near-final form as a single piece. The FCC process minimizes the number of processing steps and maximizes cermet performance; however, large equipment is required for cask fabrication. The Cermet Extrusion Section (CES) process [4] produces extruded cermet pieces that are then assembled into the cask body. Compared with the FCC process, the CES process offers several advantages: (1) it requires less research and development, (2) the front-end capital investments are smaller, and (3) it is a more flexible manufacturing process. The potential disadvantage is that this fabrication process results in a lower-ceramic-content cask. Preliminary economic assessments are favorable [4].

The cast cermet processes [5] have potentially lower fabrication costs. Casting processes are more dependent upon the specific properties of the ceramic particulates that are used, and it is more difficult to produce variable-composition cermets.

PERFORMANCE

A DUO_2–steel cermet has better radiation shielding characteristics than traditional steel casks [6, 7] because the DUO_2 has a higher density than steel and because of some neutron moderation by the high oxygen content in the DUO_2, which aids neutron capture. The cermet can include neutron absorbers such as Gd_2O_3. The higher densities reduce the wall thicknesses and increase the number of SNF assemblies per cask, based on a given total gross cask weight limit, or based on a maximum allowable diameter. The costs of cask loading, transport, and unloading are relatively insensitive to the amount of SNF in each cask. Therefore, there are strong economic incentives to maximize cask loading.

ARMOR AGAINST ASSAULT

Since 9/11, security requirements for SNF casks [8] have been reevaluated and the ability of casks to withstand assault has been analyzed [9–10]. Cermets are a traditional material used in tank armor to stop high-speed projectiles or shaped charges. Cermets contain hard ceramics such as aluminum oxide to break up the projectiles or explosive charge and spread their energy over a larger area. The ductile metal then absorbs the remaining energy. For armor, the cermet should have the highest concentration of the hardest ceramics near the outside surface to force an early breakup of the projectile or blast. Cermet SNF casks that are designed for assault resistance have the potential to be more resistant [11] than most mobile military equipment. A rail cask weighs ~100 tons compared with ~70 tons for a main battle tank; however, a cylindrical SNF cask is much smaller, with a relatively small curved surface area to protect. The new fabrication methods [3–5] have created the option to build very large cermet objects, such as SNF casks.

WASTE ISOLATION

Cermet casks can be used as engineered barriers in enhanced WPs to improve geological repository performance. The proposed Yucca Mountain (YM) WP consists of a relatively thin, unshielded internal metal container for structural strength and a thin external high-nickel C-22 shell to provide long-term corrosion protection. A cermet WP could retain the exterior C-22 shell and replace the inner shell with a thicker, heavier shielded cermet cask. The increased masses of reducing iron and DUO_2 in the cermet will improve repository performance and slow the release of SNF radionuclides for a long period of time after the outer C-22 shell has failed. This protective barrier is a result of several mechanisms.

Optimized geochemical environment. In a cermet WP, the ratio of DUO_2, iron, and other components can be adjusted to maximize WP performance [12-13] by (1) slowing the degradation of the UO_2 in the SNF and thus the release of radionuclides from the SNF, (2) slowing the movement of groundwater through the degraded WP after initial failure, and (3) creating local conditions that trap radionuclides after SNF degradation. The iron and DUO_2 are chemical reducing agents that create locally strong reducing conditions. SNF UO_2 and most other components in SNF are highly insoluble under chemically reducing conditions. Both iron and DUO_2 expand upon oxidation, plug flow channels, and slow water movement. With high affinities for nuclides, the oxidation of iron produces cohesive hydrated iron oxides that trap most heavy metal nuclides; this creates gel-like deposits that also slow water flow through the degraded WP. Iron-based scavenger precipitation processes are effectively used to purify drinking water by these mechanisms. Examples of the geochemical/physical stability of hydrated iron oxides include the intact piles of iron nails that were hastily buried in shallow pits during the times of the Roman Empire. The iron oxide gels formed on the outer surfaces of the piles have protected the inside of the nail piles from corrosion for several millennia.

Geochemical trap. Until the SNF's crystalline UO_2 is oxidized and solubilized, the radionuclides are trapped and are immobile within the sintered SNF pellets. In this context, the DUO_2 plays a unique role. Surrounding the SNF, it has the same chemical behavior as the UO_2 in SNF pellets. Therefore, if the SNF is wrapped in the DUO_2 and the DUO_2 saturates the groundwater, the cermet's DUO_2 acts as a sacrificial material to preserve the SNF UO_2 and to delay the release of nuclides from the SNF. With the same chemical behavior as SNF UO_2, cermet DUO_2 effectively protects SNF UO_2 against all external chemical degradation mechanisms until its significant chemical-buffer capacity is exhausted. While other sacrificial materials can also protect against a specific degradation mechanisms, DUO_2 is the only compound that can protect SNF UO_2 against all types of external chemical degradation mechanisms until it is consumed.

Neptunium and technetium sorption. Neptunium, technetium, and iodine are the isotopes in the performance assessments for the YM repository that control the long-term dose to the public from the repository. Performance can be improved if the WP delays the release of these radionuclides to the environment. Experimental data [14–16] indicate that various hydrated uranium oxides sorb neptunium and may reduce the peak neptunium dose by several orders of magnitude. In addition, iron also assists in reducing neptunium releases [16]. Limited data [15] indicate that hydrated uranium oxides also slow the migration of technetium from the repository; however, these data are very preliminary.

Nuclear criticality. Repositories have large inventories of fissile materials. Most of the fissile isotopes of plutonium and the higher actinides decay to ^{233}U or ^{235}U before SNF dissolution and nuclide migration from the WP occurs. The normal geochemical processes that create uranium ore bodies by reduction and precipitation may effect separation of the uranium from various neutron absorbers that were included in the WP to prevent nuclear criticality. If the fissile content in the SNF is above ~1.3% ^{235}U equivalent without sufficient neutron absorbers, nuclear criticality can occur with the creation of new fission products and actinides. The existence of natural nuclear reactors, such as Oklo, several billion years ago demonstrates how this could occur [17]. Therefore, a nuclear criticality could potentially increase the releases of radionuclides to the environment.

In the cermet WP, depleted uranium (DU) is a neutron absorber with the same chemical properties as the uranium in SNF. Addition of DU to the cermet in the WP prevents long-term criticality by lowering the overall fissile assay of the uranium by isotopic dilution [17–18]. As the SNF uranium and cermet uranium dissolve and migrate together, isotopic mixing occurs and eliminates the potential for nuclear criticality by lowering the fissile content of the uranium.

DISPOSAL OF DEPLETED URANIUM

DU is a by-product of the enrichment of natural uranium to produce enriched uranium for light-water reactors and approximately a million tons of DUO_2 currently exists in storage worldwide, with no identified large-scale reuse. It is a toxic heavy metal similar to lead and cadmium with an alpha activity of several hundred nanocuries per gram. The use of DU in cermet WPs would be an acceptable method for disposal of this waste in YM while being part of the engineered barrier system. Assessments of YM for SNF [14] and separate studies on disposal of DU as a waste in YM [19] indicate that the doses from DU disposal are orders of magnitude below regulatory limits.

If cermet casks are used for SNF storage and transport but not for disposal, there are other cask disposal options exist, such as potential disposal at the neighboring Nevada Test Site low-level burial ground. In such a case, the used casks would likely be used as disposal casks for other radioactive wastes. The Nevada shallow-land burial site has excellent disposal characteristics, including one very unique characteristic—it is a self-burying site. Wind erosion patterns result in the buildup of soil over the burial ground over time and thus provide more isolation with time.

SUMMARY AND CONCLUSIONS

The research programs have progressed sufficiently to show the technical viability of cermet casks. Concurrently, recent institutional developments are beginning to create the environment for the commercialization and deployment of cermet casks. The potential for major economic savings comes from integrating SNF storage, SNF transport, SNF disposal, and DU disposal. With the development of more integrated SNF management systems (such as the proposed DOE/RW TAD canister system), with the requirement for disposal of excess DU, and with new security requirements, new incentives emerge for highly integrated multipurpose cask systems, such as cermet casks. Significant development work remains.

These casks could unify the disposal of the two long-lived wastes (SNF and DU) from the once-through fuel cycle. If, in the future, the decision is made to go to a closed fuel cycle, the two nuclear materials required for this type of fuel cycle (SNF and DU) will be collocated and copackaged to enable joint recovery of both materials.

REFERENCES

1. C. W. Forsberg and L. R. Dole in *Proc. Advances in Nuclear Fuel Cycle Management III* (American Nuclear Society, La Grange Park, IL, 2003).
2. M. J. Haire and V. I. Shapovalov in *Proc. 10th International Conference on Environmental Remediation and Radioactive Waste Management* (Glasgow, Scotland, 2005), ICEM05-1387.
3. C. W. Forsberg, T. N. Tiegs, and V. K. Sikka in *Proc.10th International Conference on Environmental Remediation and Radioactive Waste Management* (Glasgow, Scotland, 2005), ICEM05-1350.
4. C. W. Forsberg and T. N. Tiegs in *Proc. 2006 International High-Level Radioactive Waste Management Conference* (American Nuclear Society, La Grange Park, IL, 2006), pp. 1292–1298.
5. V. T. Gotovchikov et al. in *Proc. 2006 International High-Level Radioactive Waste Management Conference* (American Nuclear Society, La Grange Park, IL, 2006), pp. 886–870.
6. C. W. Forsberg, P. M. Swaney, and T. N. Tiegs in *Proc. 14th International Symposium on the Packaging and Transportation of Radioactive Materials, PATRAM-2004* (Berlin, Germany, 2004).

7. V. I. Shapovalov et al. in *Proc. 2006 International High-Level Radioactive Waste Management Conference* (American Nuclear Society, La Grange Park, IL, 2006), pp. 885–889.
8. National Research Council, *Safety and Security of Commercial Spent Nuclear Fuel Storage* (Washington, DC, 2005).
9. F. Lange, E. Hormann, and W. Koch in *Proc. 13th International Symposium on the Packaging and Transportation of Radioactive Materials, PATRAM-2001* (Chicago, IL, 2001).
10. O. G. Alekseev et al. in *Proc. 2006 International High-Level Radioactive Waste Management Conference* (American Nuclear Society, La Grange Park, IL, 2006), pp. 1211–1216.
11. C. W. Forsberg and R. L. Landingham in *Trans. American Nuclear Society 2002 Winter Meeting* (Washington DC, 2002), p. 313.
12. C. W. Forsberg and L. R. Dole in Mater. Res. Soc. Symp. Proc. **757** (Pittsburgh, PA, 2003), pp. 677–684.
13. L. Longcheng and I. Neretnieks, *Nuclear Technol.* **137**, 228-240 (March 2002).
14. M. W. Kozak et al., "An Independent Total Systems Performance Assessment of Yucca Mountain", Nucl. Technol. (in press).
15. T. V. Kazakovskaya et al. in *Proc. 2006 International High-Level Radioactive Waste Management Conference* (American Nuclear Society, La Grange Park, IL, 2006), pp. 379–384.
16. J. H. Kessler, M. W. Kozak, M. J. Apted, W. Zhou, and G. Mungov in *Proc. 2006 International High-Level Radioactive Waste Management Conference* (American Nuclear Society, La Grange Park, IL, 2006), pp. 990–995.
17. C. W. Forsberg, *Nucl. Technol.* **131** (3), 337–353 (September 2000).
18. T. W. Hicks, *Review of the Use of Depleted Uranium for Criticality Control*, 0525-1 (Galson Sciences LTD, Rutland, England, 2005).
19. D. C. Sassani, *Disposal of Depleted Uranium at Yucca Mountain: An Assessment of Costs and Direct Impacts to Potential Repository Performance* (Yucca Mountain Project, Las Vegas, Nevada, 1999).