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Characteristics and Fabrication of Cermet Spent Nuclear Fuel Casks: Ceramic Particles Embedded in Steel

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Cermets are being investigated as an advanced material of construction for casks that can be used for storage, transport, or disposal of spent nuclear fuel (SNF). Cermets, which consist of ceramic particles embedded in steel, are a method to incorporate brittle ceramics with highly desirable properties into a strong ductile metal matrix with a high thermal conductivity, thus combining the best properties of both materials. Traditional applications of cermets include tank armor, vault armor, drill bits, and nuclear test-reactor fuel. Cermets with different ceramics (DUO_2 , Al_2O_3 , Gd_2O_3 , etc.) are being investigated for the manufacture of SNF casks.

Cermet casks offer four potential benefits: greater capacity (more SNF assemblies) for the same gross weight cask, greater capacity (more SNF assemblies) for the same external dimensions, improved resistance to assault, and superior repository performance. These benefits are achieved by varying the composition, volume fraction, and particulate size of the ceramic particles in the cermet with position in the cask body. Addition of depleted uranium dioxide (DUO_2) to the cermet increases shielding density, improves shielding effectiveness, and increases cask capacity for a given cask weight or size. Addition of low-density aluminium oxide (Al_2O_3) to the outer top and bottom sections of the cermet cask, where the radiation levels are lower, can lower cask weight without compromising shielding. The use of Al_2O_3 and other oxides, in appropriate locations, can increase resistance to assault. Repository performance may be improved by compositional control of the cask body to (1) create a local geochemical environment that slows the long-term degradation of the SNF and (2) enables the use of DUO_2 for long-term criticality control.

While the benefits of using cermets follow directly from their known properties, the primary challenge is to develop low-cost methods to fabricate casks with variable cermet compositions as a function of position in the cask body. Two fabrication methods are being developed: a casting method and a new (patent pending) powder metallurgy method. The powder metallurgy method minimizes manufacturing operations, produces a near-final-form cask, and enables construction of variable-composition cermet casks. The characteristics of the cermet casks and the new fabrication methods are described.

1. Introduction

SNF casks are required for storage, transport, and disposal. The casks may be designed for a single application or for multiple applications [1]. The functional requirements for an SNF cask include a handling package for the SNF, radiation shielding, cooling the SNF to limit its peak temperatures, physical protection, and—for waste packages (WPs)—delaying the degradation of SNF over long periods of time. Meeting these requirements economically is complicated by other constraints. Handling facilities at the reactor restrict the weight of the cask to ~ 100 tons. The physical size is limited by facility constraints at the reactor and by rail shipping requirements. Changing requirements (such as better physical protection, higher burnup SNF, longer-term storage, and repository disposal) have created strong incentives for better casks.

SNF cask performance is ultimately limited by the performance of cask materials of construction. Preliminary investigations of the characteristics of SNF casks made of cermets show the potential for superior performance compared with casks constructed of other materials. Because cermets are used in numerous applications, the properties of many cermets are either known or can be estimated from prior experience; thus, the characteristics of cermet casks can be evaluated. The outstanding performance of cermets follows from their intrinsic characteristic: the encapsulation of variable quantities of different ceramic particulates (including brittle ceramics) into a strong continuous high-integrity high-thermal-conductivity ductile metal matrix. The mixture is optimized to meet specific requirements.

Figure 1 shows a cermet cask design. Steel is selected as the continuous metal phase in the cermet because of its low cost, high strength, and good thermal conductivity. As will be discussed later, the cermet is clad in steel; thus, the cask exterior is steel. The use of cermet casks was not previously investigated because it was not evident how such casks could be fabricated economically. However, new fabrication techniques may prove economical; thus, there is a strong incentive to examine cermet casks today. This paper addresses the three key issues for a cermet cask: (1) What is the optimum cermet composition? (2) How can cermet casks be cooled? and (3) How can variable-composition cermet casks be manufactured?

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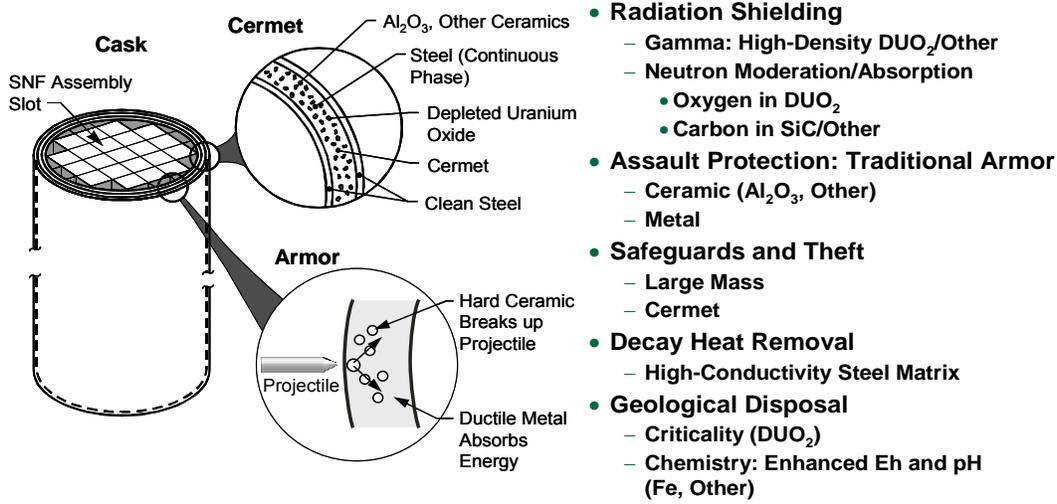


Fig. 1. SNF cask made of cermets (ceramics embedded in steel).

2. Cermet Cask Composition

The new fabrication methods enable the fabrication of variable-composition cermet casks. A series of studies have been undertaken to understand what the optimum cermet compositions should be as a function of location within the cask. Three issues are being investigated: resistance to assault, radiation shielding, and repository performance. The ultimate goal of these investigations is to enable the design of a cask optimized to meet the multiple requirements of real casks. This second integration step is a future activity.

2.1 Armor

Recent security concerns have resulted in increase interest in SNF casks that can withstand extreme events. Many types of armor, such as that used in Russian main battle tanks, are made of cermets in which the ceramic and metal components are selected to address different kinds of threats [2]. Armor made of a single material can be more easily defeated because it is possible to target the weaknesses of the specific material. In traditional cermet armors, the ceramic is a hard material (such as Al₂O₃) near the outside that breaks up the incoming projectile or explosive charge and spreads the forces over a wider area. However, hard materials are generally brittle and do not absorb much energy. The ductile metal then absorbs the energy. The inhomogeneous characteristic of the cermet breaks up shock waves. A main battle tank is lighter and larger than an SNF cask. As a consequence, appropriate designs of SNF cermet casks should be able to withstand substantially more severe events.

2.2 Shielding

SNF cask capacity is usually limited by weight. Reactor limitations, such as cranes, determine the maximum gross cask weight. If the cask weight can be reduced, the number of SNF assemblies per cask can be increased with the same gross (loaded) cask weight. Cask weight is determined primarily by gamma shielding requirements where, to a first approximation, the required shielding can be defined in terms of the mass per unit area (g/cm^2) necessary to stop the gamma radiation. If this were the only consideration, cask weight would be independent of the density of the shielding material. However, two geometric effects are also present.

- *Cask diameter.* The “area” requiring shielding is a variable. Excluding end effects, if the shielding material had no thickness, the area would be defined as follows: πH inner diameter of the cask H cask height. However, shielding materials have various thicknesses. At a distance of 1 cm into the shielding, the area is πH (inner diameter of the cask + 2 cm) H height. The further from the inner cask surface, the more shielding material is required per centimeter of cask thickness, because the cask circumference increases as one moves out from the inner cask diameter. As a consequence, cask weight can be minimized by using high-density shielding materials with minimum cask shield thicknesses. As a high-density ceramic, DUO_2 has major advantages: a density significantly greater than that of steel, the matrix material; compatibility with the cermet manufacturing process; chemical stability; availability in large quantities (500,000 tons excess DU in storage); and a relatively low cost.
- *SNF characteristics.* To minimize cask weight, shielding density should vary with cask elevation, with the greatest density of shielding where the radiation levels are highest. The cermet allows variable density with cask height by the choice of ceramics and the volume fraction of the cask that is a ceramic. High-density ceramics (such as DUO_2) are used where the gamma-radiation levels are high. To minimize weight, only steel ($\rho = 7.86 \text{ g/cm}^3$) and low-density ceramics, such as Al_2O_3 ($\rho = 3.97 \text{ g/cm}^3$), can be used where the radiation levels are lower, particularly at the outer cask edges far from the SNF.

The shielding capabilities [3] of a DUO_2 cermet cask were evaluated relative to the licensed Holtec HI-STAR 100 SNF transport cask [4]. *This initial assessment assumed a single composition of the cermet—not an optimized variable composition.* HI-STAR is a steel cask that uses multiple layers of steel for gamma shielding and an external neutron shield of Holtite-A. The cask consists of a 6.35-cm-thick (2.5-in.-thick) layer of SA#203E steel, followed by a 15.24-cm-thick (6-in.-thick) multi-layer section of SA#516 grade 70 steel for gamma shielding. The Holtite-A is a moderator and neutron absorber developed and licensed by Holtec International.

Shielding properties were investigated using the SAS1 module from the SCALE 4.4a code [5]. To produce the radiation source terms, ORIGEN-ARP Express from the SCALE 4.4a code was used. The most radioactive SNF listed in the Final Safety Analysis Report (FSAR) of Holtec International’s HI-STAR 100 cask system [4] was chosen as the source term: B&W 15 x 15 pressurized-water reactor fuel assemblies with an enrichment of 3.4% and a burnup of 40,000 MWd/ton.

The cask model was simplified to a cylinder with concentric layers. The inside of the cask is a multipurpose canister (MPC) with 24 SNF assemblies, the basket structure, and helium backfill. The MPC model consisted of (1) a smeared homogeneous material representing the SNF and grid structure, with the radiation source spread evenly throughout; (2) the outer steel shell of the MPC; and (3) the thin annulus backfilled with helium between the MPC and shielding. The source terms derived from ORIGEN-ARP are higher than those used for the Holtec FSAR. The cylinder was modeled with a height of 365.76 cm, which is the height of the active fuel region listed in the FSAR for the HI-STAR 100. The radiation doses reported herein are the doses at the outer wall of the cask at midplane of the the active fuel region.

The calculations (see Table 1) did not include the Holtite layer of material but only compared the steel versus cermet cask body. The cermet calculations used 1.27-cm-thick (0.5-in.-thick) layers of SS#316 on either side of the layer of cermet. The cermet was composed of 50% iron and 50% DUO_2 by volume with the with a ^{235}U enrichment of 0.30%. The external surface dose of the 21.59-cm-thick (8.5-in.-thick) steel Holtec cask consisted of a gamma dose of 507 mrem/h and a neutron dose of 119 mrem/h. To achieve equivalent gamma shielding requires a 15.14-cm-thick (5.96-in.-thick) cermet cask, which is only 70% of the thickness of the steel cask. The corresponding cermet cask radiation levels are 373 mrem/hr gammas and 130 mrem/hr neutrons. The total cask weight (sides, bottom, etc.) is 86% of the weight of the equivalent steel cask. The cermet cask wall consists of 12.6-cm (4.96-in.) of cermet sandwiched between two 1.27 cm (.5 in) layers of SS#316.

**Table 1. Neutrons and Neutron Radiation Doses from Steel and Cermet Casks
(Without Holtite Neutron Shielding Layer)**

	HI-STAR		Cermet cask		Cermet cask with B ₄ C	
Shielding thickness (cm):	21.59		15.64		20.44	
Total gamma dose (mrem/hr):	506.89		372.99		37.38	
Total neutron dose (mrem/hr):	119.44		130.10		73.76	
Upper energy limit	<u>Spectrum</u> (neutrons/cm ² /sec)	<u>Dose</u> (mrem/hr)	<u>Spectrum</u> (neutrons/cm ² /sec)	<u>Dose</u> (mrem/hr)	<u>Spectrum</u> (neutrons/cm ² /sec)	<u>Dose</u> (mrem/hr)
20 MeV	4.22E-01	0.063016	2.66E+00	0.397177	1.14E+00	0.170412
6.34 MeV	3.60E+00	0.520222	2.19E+01	3.172172	8.99E+00	1.300881
3 MeV	9.97E+00	1.266495	4.28E+01	5.442163	1.89E+01	2.404985
1.85 MeV	1.07E+01	1.376094	3.20E+01	4.095805	1.45E+01	1.85903
1.4 MeV	4.08E+01	5.292981	6.83E+01	8.857854	3.27E+01	4.248527
900 keV	2.48E+02	25.54376	3.71E+02	38.14683	2.14E+02	21.95179
400 keV	1.25E+03	64.13895	1.09E+03	55.80227	6.56E+02	33.59104
100 keV	1.53E+03	18.80468	9.10E+02	11.21499	5.55E+02	6.842077
17 keV	2.98E+02	1.14256	3.42E+02	1.312068	1.99E+02	0.764139
3 keV	7.82E+01	0.291203	2.22E+02	0.827792	1.09E+02	0.405621
550 eV	6.61E+01	0.265378	1.14E+02	0.456373	4.00E+01	0.160788
100 eV	5.24E+01	0.224815	4.19E+01	0.179963	1.06E+01	0.04536
30 eV	4.46E+01	0.199335	1.96E+01	0.087587	3.07E+00	0.013714
10 eV	3.57E+01	0.162895	6.26E+00	0.028576	6.02E-01	0.002749
3.05 eV	1.28E+01	0.058389	3.75E+00	0.017111	1.11E-01	0.000504
1.77 eV	5.95E+00	0.026902	2.33E+00	0.010518	3.18E-02	0.000144
1.3 eV	2.40E+00	0.010785	1.08E+00	0.004857	9.72E-03	4.36E-05
1.13 eV	1.84E+00	0.008591	9.36E-01	0.004367	5.82E-03	2.72E-05
1 eV	2.75E+00	0.012189	1.67E+00	0.007403	6.04E-03	2.68E-05
0.8 eV	4.79E+00	0.020734	4.38E+00	0.018944	4.85E-03	2.1E-05
0.4 eV	9.27E-01	0.00389	1.00E+00	0.004199	6.11E-04	2.56E-06
0.325 eV	9.03E-01	0.003701	1.16E+00	0.004747	3.31E-04	1.36E-06
0.225 eV	6.25E-01	0.002401	9.75E-01	0.003743	9.64E-05	3.7E-07
0.1 eV	1.79E-01	0.000657	3.13E-01	0.001151	1.24E-05	4.55E-08
0.05 eV	4.58E-02	0.000168	8.44E-02	0.00031	1.63E-06	5.99E-09
0.03 eV	1.52E-02	5.58E-05	2.66E-02	9.78E-05	2.44E-07	8.97E-10
0.01 eV	1.45E-03	5.32E-06	8.96E-04	3.29E-06	2.15E-09	7.91E-12

The option exists to add a neutron absorber to the cermet and replace the Holtite-A neutron shielding layer on the outside of the cask. This may be attractive for applications (such as in a repository) where an organic neutron shielding material is unacceptable. The Holtec cask with the Holtite neutron shielding has at the surface an external neutron dose of 1.08 mrem/h and an external gamma dose of 107 mrem/hr. In these calculations 1.0% by volume B₄C was added to replace some of the cermet in the shielding. With a 20.44-cm (8.05-in.) thick cermet cask wall, the radiation doses outside the cask were 37.38 mrem/h gamma and 73.76 mrem/h neutron. The cermet cask wall consisted of 18-cm (7.09-in.) of cermet with 1.27-cm (0.50-in.) inner and outer steel liner. When the neutron absorber is included in the cask, the cermet cask weight is 93% of that of the body weight of the steel cask.

Based on this and other analysis, several conclusions are drawn.

- *Shielding effectiveness.* The DUO₂ cermets are much better shielding materials than steel per unit thickness and better shielding materials per unit of mass. Analysis of neutron shielding as a function of neutron energy (Table 1) indicates some generation of fission neutrons by the DUO₂, indicating an incentive to use DUO₂ with a minimum ²³⁵U content. Future studies will examine the use of higher-volume-fraction DUO₂ toward the inner surface and Al₂O₃ toward the outer surface.
- *DUO₂ cermet.* Increasing the DUO₂ volume fraction improves gamma shielding and will lower cask weight; however, it reduces the strength of the cermet. With the option to produce a variable composition cermet, the strength can be tailored as a function of depth in the wall to maximize total system performance. Cermets with up to ~70% volume of ceramic are possible. The gamma shielding should be as close to the SNF as possible to minimize total shielding weight.
- *Al₂O₃ cermet.* If the cermet is to replace the neutron shielding of the exterior Hottite layer, the addition of Al₂O₃ (oxygen as moderator) with a neutron absorber toward the outer cask surface may further minimize shielding weight. As long as the gamma dose is the controlling dose, a high volume fraction DUO₂ cermet (with its high electron density that maximizes gamma shielding) is preferred. If the neutron dose dominates at further distances from the SNF, lower density cermets with better neutron moderating and absorption characteristics (higher nuclear cross sections) are preferred. The same general configuration is expected to maximize resistance to assault.

2.3 SNF Repository Disposal

Cermet casks can be used as multipurpose casks for storage, transport, and disposal of SNF in a repository. If required, a corrosion-resistant overpack can be added at the repository to improve cask disposal performance. Alternatively, disposal-only cermet WPs can be manufactured. In these applications, cermets offer two potential advantages.

- *Repository performance.* The functional purpose of a WP is to delay the release of the SNF until a large fraction of the radionuclides have decayed away. The degradation of SNF and release of radionuclides from the WP depends upon the local chemical environment [redox conditions (Eh) and acid-base conditions (pH)]. These, in turn, are determined by the chemistry of the WP. In a cermet WP, the ratio of DUO₂, iron, and other components can be adjusted to maximize WP performance [1]. In this context, any depleted uranium in the cask is preferred to be in the form of DUO₂ to avoid chemical incompatibility concerns relative to SNF.
- *Nuclear criticality.* SNF contains many fissile isotopes such as ²³⁵U, ²³⁹Pu, and ²⁴¹Pu. The plutonium isotopes decay to long-lived uranium isotopes (²³⁵U and ²³³U). Advanced reactors, such as high-temperature reactors, may have fresh fuel enrichments approaching 20% ²³⁵U and resultant SNF enrichment levels of 2 to 3% ²³⁵U. These enrichment levels create the long-term potential for uranium migration in groundwater and formation of deposits where nuclear criticality may occur—the same mechanism that caused natural uranium reactors to exist several billion years ago. The use of DUO₂ reduces the potential for long-term repository criticality by isotopically diluting the fissile uranium isotopes to lower enrichment levels. If there is good uranium isotopic mixing, nuclear criticality becomes impossible.

3. Decay Heat Removal

Economics favors the use of large casks with their lower cost per fuel assembly. Cermets are an enabling technology to increase cask capacity. However, a second constraint exists: allowable peak SNF temperatures. If excess temperatures occur (>350°C), the SNF degrades. Improved cask cooling methods (Fig. 2) are being investigated [6] to ensure that SNF temperature constraints do not limit the benefits from using higher-capacity cermet casks. There are three resistances to heat transfer: from SNF to cask body, within the cermet cask body (the smallest resistance to heat transfer), and from cask body to the atmosphere. Methods to improve heat transfer and to lower peak temperatures are being investigated for use inside and outside the cermet cask body.

- SNF to cask body.** Decay heat is transferred from the SNF to the cask body by two mechanisms: conduction through the inert gas and basket structure and circulation of inert gases from the SNF to the inner cask wall. The use of xenon-helium or argon-helium gas mixtures are being investigated to replace the traditional helium gas mixture and improve heat transfer. In terms of conductive heat transfer, helium has a very high thermal conductivity compared with that of any other inert gas. However, in terms of convective heat transfer, the use of xenon (the highest-density gas) best maximizes natural circulation of the fill gas from the SNF to the cask body. Natural convection currents are driven by differences in the density of gases in the hot (high-temperature) fuel channel versus the cooler walls of the cask body.

Measurements of the physical properties of gas mixtures [7] containing helium and more dense gases indicate the potential for superior performance of gas mixtures to transfer heat in an SNF cask compared with pure gases. The addition of helium to another inert gas greatly improves the thermal conductivity of the gas mixture while maintaining the high average molecular weight that maximizes convective gas heat-transport flow. If xenon and helium are mixed so that the average molecular weight is equal to that of argon (~40), the natural circulation currents driven by density differences with this gas mixture are approximately equal to those of argon. However, the thermal conductivity of such a mixture is over three times that of argon. In effect, the gas mixture has some of the desirable properties of heavy gases (convective heat transfer) and some of the desirable properties of helium (conductive heat transfer).

- Cask body to atmosphere.** On the outside cask wall, liquid-cooled (water with antifreeze and corrosion inhibitors), natural-circulation bolt-on cooling fins can replace solid fins to improve transfer of heat from the cask body to the air [6]. Compared with equivalent solid fins, liquid-cooled fins reduce the temperature drop from cask surface to atmosphere by up to a factor of 3 for the same amount of decay heat. Liquid natural circulation creates almost isothermal conditions on the inside of the fins, which enhances heat transfer. Such fins are similar to those used commonly in electrical transformers.

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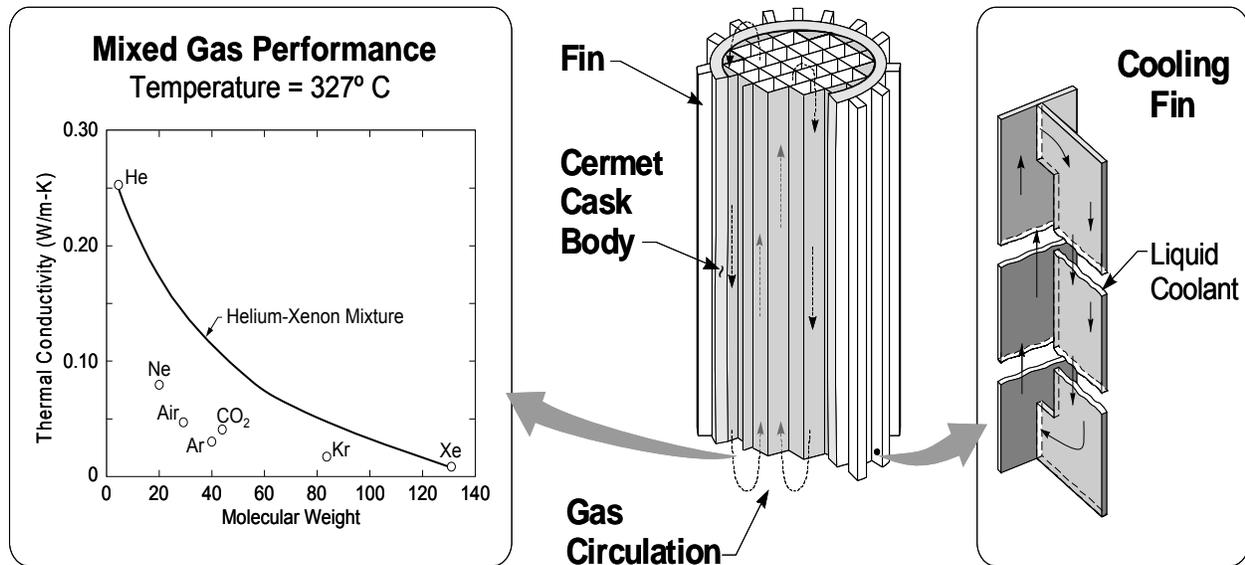


Fig. 2. Methods to enhance cooling of SNF in casks.

4. Manufacturing

The economic viability of cermet casks depends upon manufacturing costs. Traditional cermet fabrication methods are expensive for construction of large casks because cermet plates must first be produced and then fabricated into casks. The traditional powder metallurgical technique involves (1) mixing the metal powders and ceramic particulates, (2) enclosing the mixture in some type of close-fitting metal box, (3) heating the mixture while removing the gases between the particulates by vacuum, and (4) compressing the box and mixture at high temperatures to create a monolithic matrix of metal plate that encapsulates ceramic particulates. The plates must then be fabricated into casks; however, cermets are very difficult to form and weld. The multistep process thus results in high costs.

Two new methods for cermet cask fabrication are being investigated: a method involving casting a cermet and a new (patent pending) powder metallurgy method [8, 9]. Each has potential major advantages over traditional technologies. The new powder metallurgy method (Fig. 3) is described herein. The potential favorable economics are a result of (1) a process that produces a near-final-form cask, which minimizes the number of processing steps, and (2) the low cost of the starting materials. Although the fabrication technique is new, on a microscopic scale (temperatures, pressures, material compositions), the cermet-forming processes are the same as those associated with the traditional techniques. The new process consists of the following steps.

- *Preform fabrication.* A preform slightly larger than the final annular cask body is constructed of steel and serves as the inner and outer layer of clean steel in the final cask. The preform consists of the inside, outside, and top surfaces of the cask body but excludes the cask bottom.
- *Preform filling.* The preform is filled with a particulate mixture of DUO_2 , other ceramics, and steel powder. A schematic of the filling process is shown in Fig. 4. The upside-down cask preform is placed on a table that can be rotated. The particulate distribution heads of the fill machine are lowered to the bottom of the preform. As the table rotates, the fill machine (1) feeds particulate mixes (steel and ceramic particulates) to the preform in a continuous layer that maybe several centimeters deep, (2) compacts each layer as it is placed in the preform, and (3) is withdrawn as the preform is filled with a continuous spiral particulate layer from the bottom of the preform to the top. Hundreds of rotations are required to fill the preform. The use of multiple particulate feed nozzles makes it possible to vary the composition of individual layers from the inside of the cask preform to the outside. The composition of the particulate mix can also be varied in the vertical direction. The compaction is to increase the green density of the powder bed and to prevent movement of the particulate fill during subsequent handling operations. The gas composition within the preform is maintained under chemically inert conditions to avoid oxidation of the steel powder.
- *Welding, heating, and gas evacuation.* After the preform is filled, an annular ring is welded to the preform to create a loaded, sealed annular preform. The preform is then evacuated while being heated, which removes gases in the void spaces in the particulate mixture and those gases sorbed on the particulates.
- *Forging.* The preform is heated and compressed to (1) eliminate void spaces and (2) weld the metal particles together to form a continuous, strong steel matrix containing various ceramic particulates. The compression is performed at high temperatures to (1) minimize the forces necessary to eliminate voids in the particulate mixture and (2) rapidly weld the steel particulates into a solid matrix by solid-state diffusion. The forging temperature is significantly below the melting point of the metal. If this were a molten system, the high-density ceramics would sink to the bottom and the low-density ceramics would float on the surface of the molten metal. However, the powder metallurgy technique allows the variable-composition cermet to be fabricated. Two standard industrial processes to consolidate the preform and particulate mixture currently exist.
 - Traditional forging. The hot heated perform mix can be hammered to consolidate the particulate mixture into a cermet and produce the final cask form. In one method (shown in Fig. 3), a cylindrical anvil the size of the interior of the final cask is placed inside the preform. The forge then strikes the exterior to consolidate the particulate mixture.
 - Ring-rolling forging. The hot loaded preform can be placed in a ring-rolling machine and rolled to its final form.
- *Finishing.* The cask bottom is welded onto the cylindrical cask body. After completion of this step, a vertical boring mill is used to obtain the final dimensions and to drill holes in the top of the cask for the lid bolts. All welding and machining operations are performed on the preform, not on the internal cermet. This avoids the very difficult operations of welding or machining cermets.

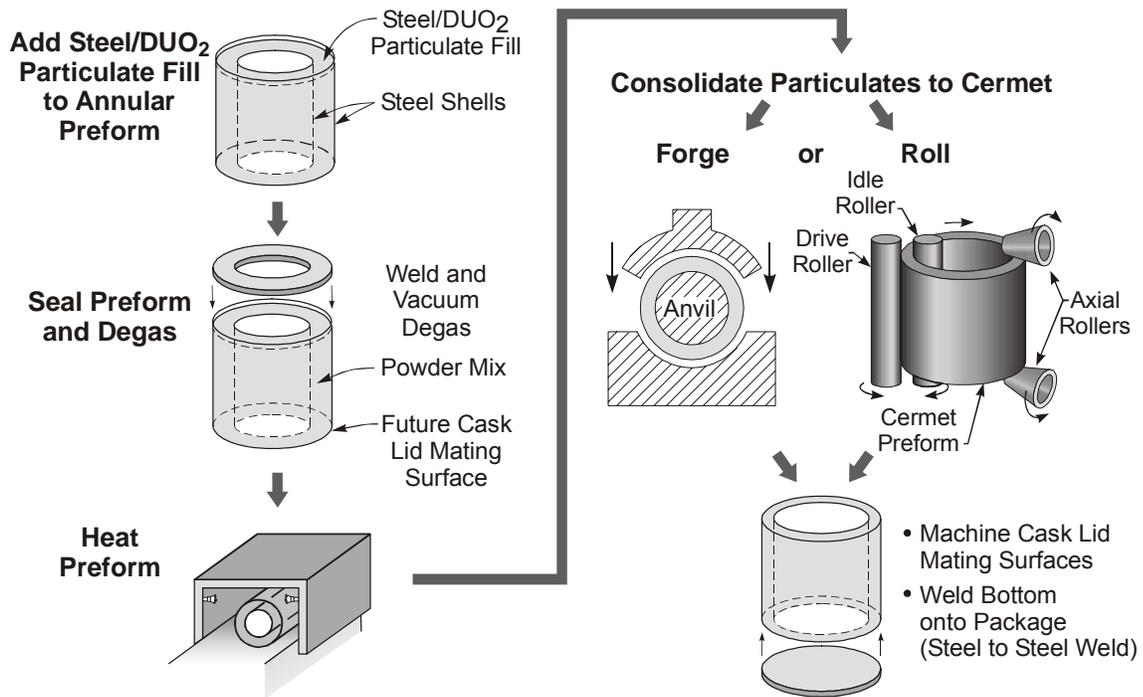


Fig. 3. New method for the manufacture of DUO₂-steel SNF casks.

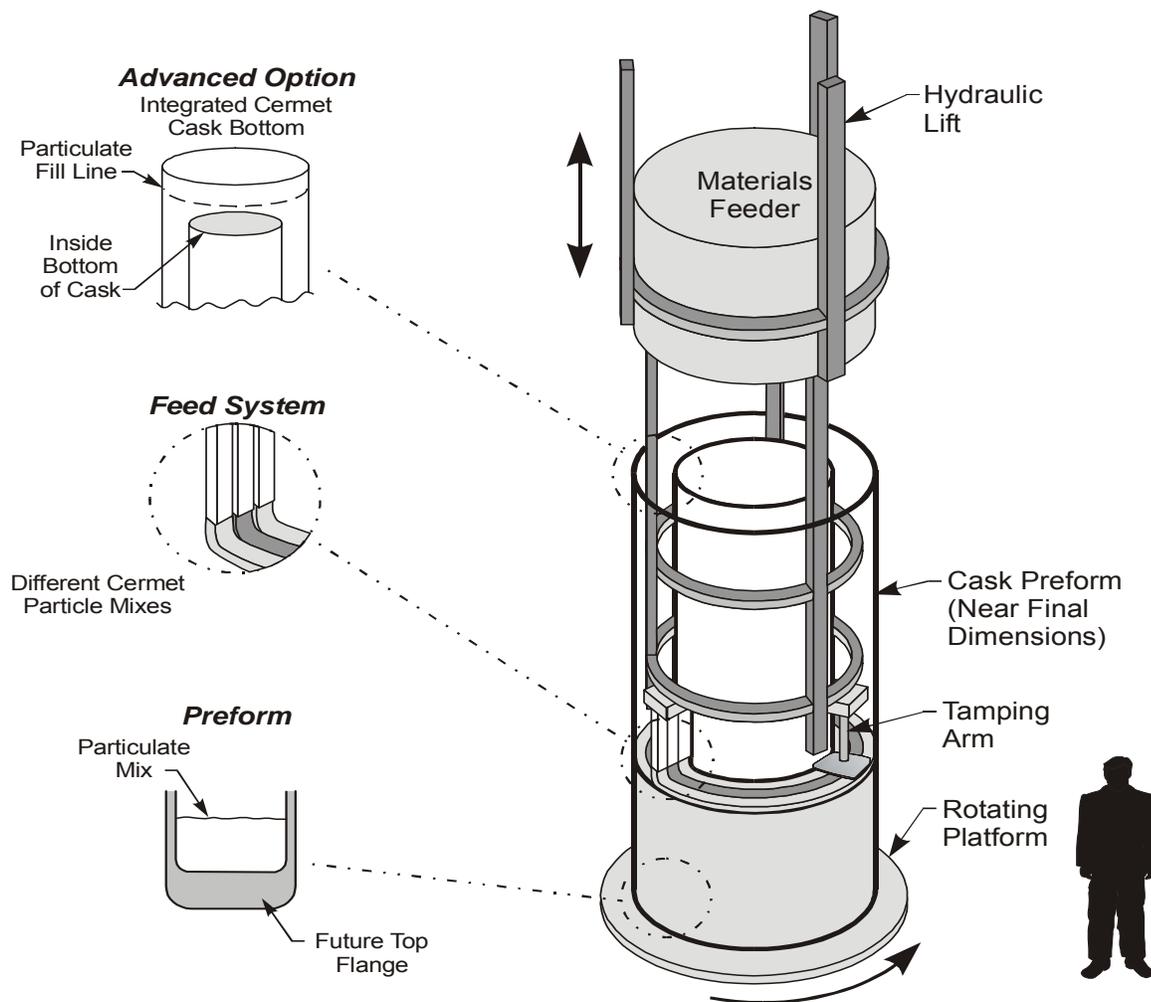


Fig. 4. Loading of the cermet preform with variable cermet particulate compositions and steel powder.

Many fabrication variants exist. The preform can include the bottom of the cask (Fig. 4). Although this technique allows the fabrication of a preform that incorporates the cask bottom, more-sophisticated forging operations are required to produce an integral bottom and side cermet cask.

Powder metallurgy production techniques have the potential for low costs. Millions of tons of iron and steel powders are produced for the fabrication of many products; thus, the costs of raw materials are low. The current cost for steel powder purchased in large quantities is about \$600/ton. Manufacturability and costing studies are under way.

5. Conclusions

Changing requirements for casks (security, higher-burnup SNF, longer SNF storage times, and disposal) have created strong incentives for development of new materials for casks. Cermet intrinsically have the potential for the best cask properties because they combine the characteristics of ceramics and metals. New cermet fabrication methods have the potential for low costs. Research is under way to develop (1) a design methodology to define the optimum cermet compositions and (2) a low-cost manufacturing methods for cermet casks.

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