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POWDER-METALLURGY EXPERIMENTS AND MANUFACTURING STUDIES ON DUO_2-STEEL CERMETS FOR SPENT NUCLEAR FUEL CASKS

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ABSTRACT

Two new methods are being investigated to manufacture cermet casks to be used for storage, transport, or disposal of spent-nuclear-fuel (SNF) casks. 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.

The addition of aluminium oxide (Al_2O_3) ceramic particles to the cermet can improve resistance to assault. The addition of depleted uranium dioxide (DUO_2) ceramic particulates can increase SNF cask capacity, improve cask performance in a repository, and potentially dispose of excess depleted uranium, a potential waste form. Other ceramic particulates provide other benefits.

Two powder-metallurgy fabrication methods are being developed. In each process, a mixture of iron and ceramic powders is ultimately converted into a high-integrity SNF cask. The recently patented Forge Cermet Cylinder (FCC) process 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 produces extruded cermet pieces that are then assembled into the cask body. The size of the individual pieces minimizes process equipment sizes and facility investments but adds steps to the fabrication process and results in a lower cermet content of the cask. The two fabrication processes are described and compared. The results of preliminary experimental work and fabrication studies are reported.

INTRODUCTION

Spent Nuclear Fuel (SNF) casks are required for storage, transport, and disposal. The casks may be designed for single or for multiple applications [1]. The functional requirements for an SNF cask include a handling package for the SNF, radiation shielding, cooling of 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 to design 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 [2] compared with casks constructed of conventional materials. The outstanding performance of cermets follows from their intrinsic characteristic: the encapsulation of *variable* quantities of different ceramic particulates into a strong continuous high-integrity high-thermal-conductivity ductile metal matrix. The mixture is optimized to meet specific requirements.

The use of cermet casks was not previously investigated because it was not evident that such casks could be fabricated economically. However, new fabrication techniques may prove economical; thus, there

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is presently a strong incentive to examine cermet casks. This paper describes investigations into two advanced methods of cermet cask manufacture that allow the construction of variable-composition cermet casks.

CERMET CASK BENEFITS AND 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. The cermet is clad in steel to provide a clean outer layer to allow easy decontamination of the cask during normal operations.

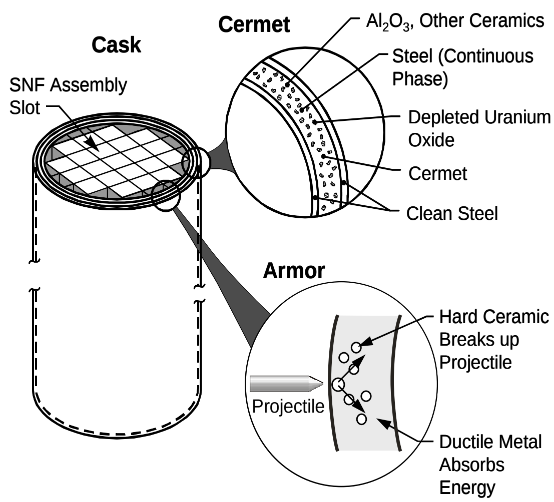

Fig. 1. Design of cermet cask

The cask functional requirements define the requirements for the cermet fabrication process.

Physical Protection

Recent security concerns [3] have resulted in increased interest in SNF casks that can withstand extreme events. Cermets are a traditional material used in tank armor and in various cutting tools. In traditional cermet armors, the ceramic is a hard material (such as Al_2O_3) near the outside surface that breaks up incoming projectiles and defocuses explosive charges. This 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. The hardest ceramics should be located near the outside of the cask.

Shielding

For aged SNF, cask capacity is limited by weight and size. 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 (grams per square centimeter) necessary to stop the gamma radiation. However, higher-density materials reduce the shield thickness and thus the outer dimensions of the cask. This favors the use of dense ceramics. DUO_2 is the leading candidate for this requirement.

If the cask weight can be reduced, the number of SNF assemblies per cask can be increased with the same gross (loaded) cask weight. There is a geometric effect in shielding. 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. Cask weight can be reduced by placing the high-density ceramics (such as DUO_2) in a cermet toward the inside of the cask body.

SNF Repository Disposal

Cermet casks can be used as multipurpose casks for storage, transport, and disposal of SNF in a repository. Alternatively, disposal-only cermet WPs can be manufactured. In these applications, the use of DUO_2 cermets offer two potential advantages.

- *Repository performance*. The functional purpose of a WP is to delay the release of the radionuclides in the SNF until a large fraction of the radionuclides have decayed away. The degradation of SNF and release of radionuclides from the WP depend 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 DUO_2, iron, and other components can be adjusted to maximize WP performance [1] to slow the degradation of the UO_2 in the SNF. The UO_2 matrix in the SNF traps most of the radionuclides. Until the UO_2 is destroyed, the radionuclides are trapped. In this context, the DUO_2 plays a unique role. It has the same chemical behavior as the UO_2 in SNF and thus acts as a sacrificial material to preserve the SNF UO_2. As the only compound with the same chemical behavior, DUO_2 is the only compound that can protect SNF UO_2 against all types of chemical degradation mechanisms until it is consumed.

- *Nuclear criticality*. SNF contains many fissile isotopes such as ^{235}U, ^{239}Pu, and ^{241}Pu. The plutonium isotopes decay to long-lived uranium isotopes (^{235}U and ^{233}U). Under some conditions, there is the potential for uranium migration in groundwater and formation of uranium deposits where nuclear criticality may occur—the same mechanism that caused natural uranium reactors to exist several billion years ago. The use of DUO_2 reduces the potential for long-term

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repository criticality by isotopically diluting the fissile uranium isotopes to lower enrichment levels.

Waste Management

Depleted uranium dioxide (DUO_2) is the by-product of the enrichment of natural uranium to produce enriched uranium for light-water reactors. Approximately a million tons of DUO_2 is in storage worldwide with no identified large-scale uses. This application could use most of this inventory and avoid treating this material as a waste.

CERMET CASK FABRICATION

Traditional Processes

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 cermet matrix with an external steel surface. 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 powder-metallurgy methods for cermet cask fabrication are being investigated: the Forge Cermet Cylinder (FCC) process and the Cermet Extrusion Section (CES) process. Both potentially have lower costs than traditional cermet processes and avoid welding of cermets.

FCC Cask Fabrication

The FCC cask fabrication process is a new process, with the patent issued late last year [4]. 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, and material compositions), the cermet-forming processes are the same as those associated with the traditional techniques. The FCC process (Fig. 2) 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. 3. 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 may be 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. Many 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 preformed 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 filling is completed, 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 (Fig. 2).

  – <u>Traditional forging.</u> The hot heated perform can be hammered to consolidate the particulate mixture into a cermet and produce

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the final cask form. In one method, 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.

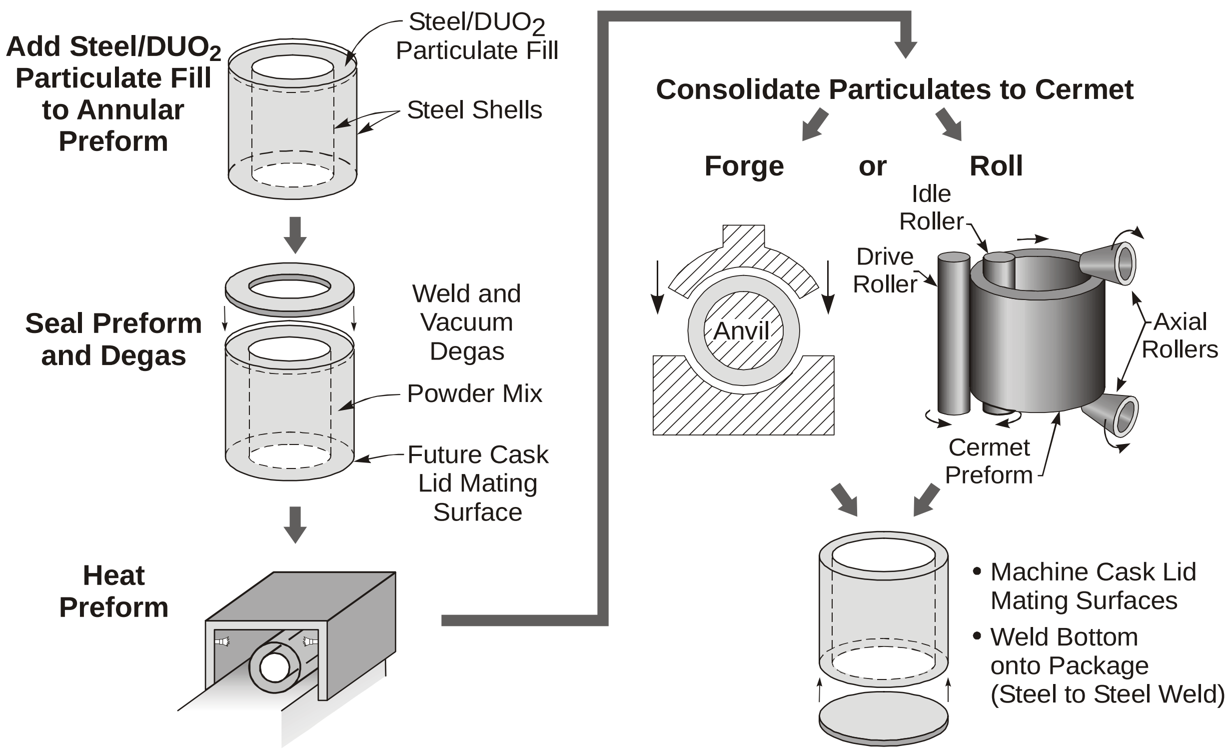

Fig. 2. FCC process for the manufacture of DUO_2–steel SNF casks.

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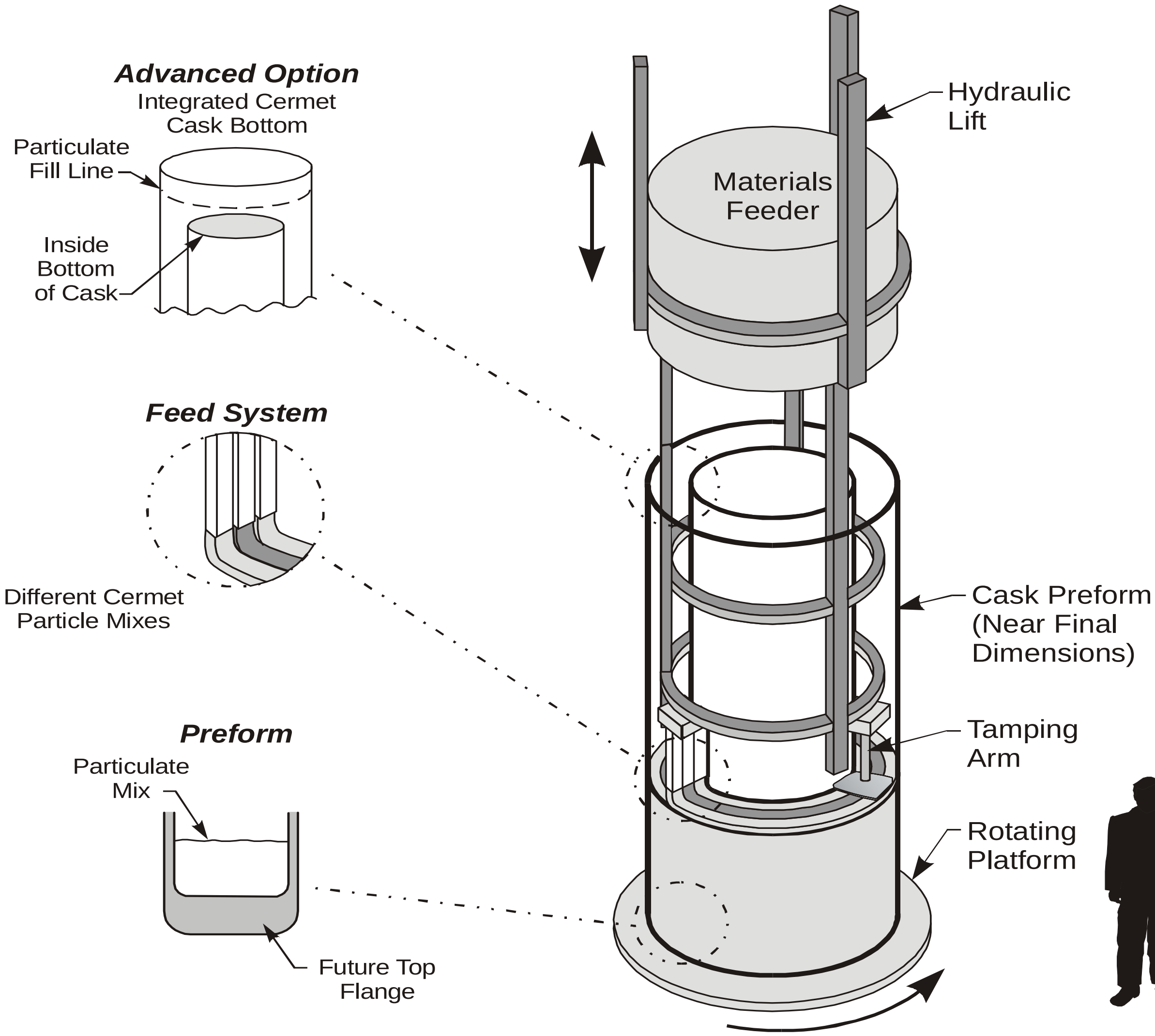

Fig. 3. Loading of the cermet preform with variable cermet particulate compositions and steel powder.

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The FCC process minimizes the number of processing steps and maximizes cermet performance. A variable-composition cermet cask can be constructed. However, large equipment is required—particularly for the one-piece forging step. Thus, a key economic issue for this fabrication process is the total demand for casks. The fabrication economics are dependent on cask production rates.

CES Cask Fabrication

The CES process produces extruded cermet segments that are then assembled into the cask body. Figure 4 shows the various pieces required to fabricate a cask. A radial cermet design is envisioned to prevent radiation line-of-sight problems along seams in the cask. Studies are currently evaluating the use of 12 to 48 extruded segments to make a complete cask.

The FCC and CES processes have many similarities. The cermet is made by filling long steel boxes with the appropriate powder mixtures, welding tops onto the boxes, and then heating and evacuating the gases from the boxes. The filled boxes are then hot extruded to consolidate the compacted powder mixture into a cermet. The extrusion process produces the desired final cross section. The extruded cermet sections will be press fitted between two steel cylinders to produce the final desired shape. Major advances in extruding technology have occurred in the last decade that (1) allow the original box to be filled with a variable-composition powder that results in a variable-composition cermet and (2) enable high-temperature extrusions. Final processing would include a heat-treatment step to bond the cermet sections and preform cylinders together.

The small size and weight of the individual pieces minimizes process equipment sizes and facility investments but adds steps to the fabrication process. Each of the cermet sections requires a full steel exterior for the extrusion process. Consequently, the greater the number of cermet sections used to manufacture the cask, the more steel that is located in interior sections of the cask. This reduces the fraction of the cask made of a cermet.

LABORATORY EXPERIMENTS AND PATH FORWARD

The fabrication processes have many common steps. Experiments have been initiated to develop appropriate processing parameters to produce cermet casks. Once the processing parameters are known (including time to conduct each step), a preconceptual design of the manufacturing facility can be developed along with reasonable estimates of cask fabrication costs.

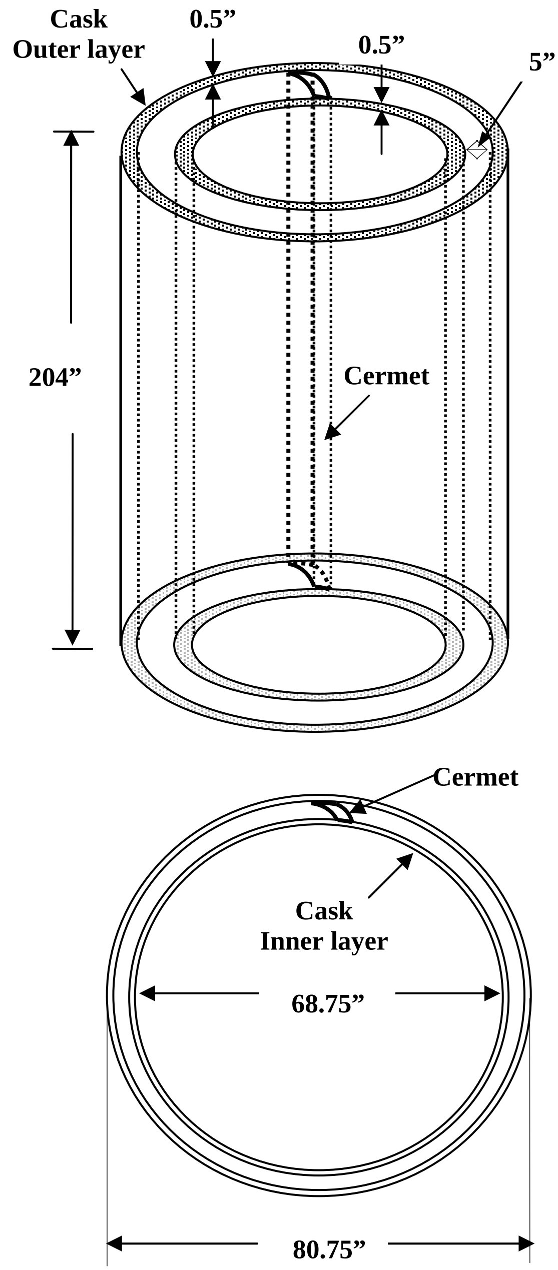

Fig 4. CES cask with cermet segments between inner and outer cask shells.

Cermet manufacture requires powder mixes with constituents that have very different densities. Appropriate handling and precompaction steps are required to avoid segregation of these constituents during processing operations. Tests help define the thickness of a powder mixture in a single layer before it is necessary to compress the mixture and add the next layer of the unconsolidated cermet powder mixture. Initial segregation tests were conducted by filling a graduated cylinder with a blended mixture of commercial HfO_2 (a UO_2 surrogate) and steel powders to about

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7 cm. The arrangement was then put on a vibration table for 2 h. Samples were then recovered from different levels and the steel content determined. The results are shown in Fig. 5. As indicated, the mixture was close to the as-blended concentration (44.3 wt% steel) with the exception of the bottom section, which was lower in steel concentration. The segregation of the HfO_2 to the bottom can be attributed to its high density (9.8 g/cm^2) and small average particle size (~3 µm). The steel powder, on the other hand, had a density of 7.8 g/cm^2 and a mean particle size of ~100 µm.

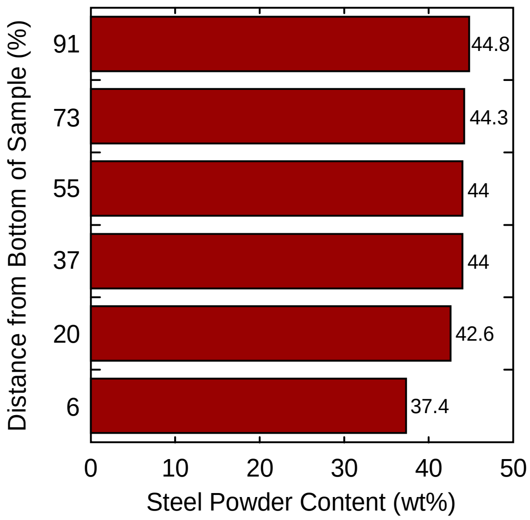

Fig. 5. Results of segregation test of steel and HfO_2 powder blends.

Powder mixtures are compacted before the final high-temperature step that produces the cermet. Compaction reduces the required consolidation in the final step. In any powder-fabrication method, the compaction characteristics must be determined to define reasonable compaction pressures that balance the level of densification versus the compaction pressure. The results of the compaction testing of the blended powders are shown in Fig. 6. The apparent density of the powder blend is ~35% of theoretical density, which is typical for powders. With the application of pressure, the densities are increased as particle rearrangement occurs. The steel powder used contained 0.75 wt% zinc stearate as a lubricant. Warm compaction (above the softening point of the lubricant) showed that the density could be improved by a few percent. The compacted powder density is 60 to 70% of the theoretical density.

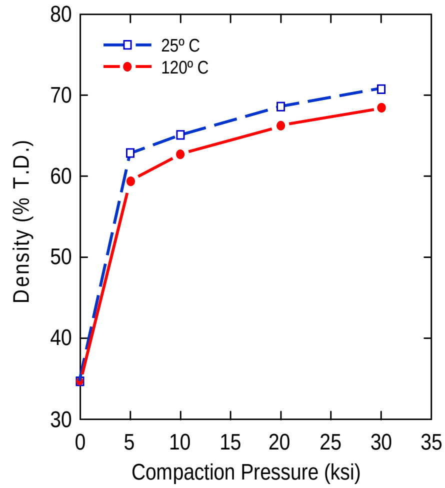

Fig. 6. Results of compaction study of steel and HfO_2 powder blends.

These studies will be completed this year. The development of a detailed manufacturing flowsheet will then provide the basis for reliable manufacturing cost estimates as a function of various parameters.

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. Cermets intrinsically have the potential for the best cask properties because they combine the characteristics of ceramics and metals. Two potentially low-cost alternative cermet cask fabrication methods are being examined. Each has specific advantages and disadvantages. Manufacturing studies are under way to understand the requirements for each manufacturing step. These studies will in turn be used to develop costing models of the fabrication process under a wide range of conditions.

The two processes have different advantages and disadvantages. The CES process may be the preferred option if the annual cask production rate is low. The equipment required is relatively small in size (lower investment costs) and the process is expected to have lower R&D costs. The FCC process has advantages at higher manufacturing rates because it requires fewer manufacturing steps.

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