A New Method for Manufacturing Depleted Uranium Dioxide (DUO_2) Material for Spent Nuclear Fuel

Metode Baru Pembuatan Material Uranium Dioxide (DUO_2) untuk Limbah Bahan Nuklir

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Abstract

Cermets (ceramics embedded in a metal matrix) using depleted uranium dioxide (DUO_2) and other ceramics embedded in a steel matrix are being investigated as materials of construction for (1) spent nuclear fuel (SNF) storage, transport, disposal, and multifunction casks and (2) casks for gamma-emitting radioactive wastes. Cermets cask performance (capacity for a given weight limit, capacity for a given size limit, resistance to assault, etc.) may significantly exceed the performance of casks constructed of more traditional materials such as steel. The principal viability issue is manufacturing costs. A new powder metallurgy method for fabricating large casks has been invented (patents applied for) that may result in low fabrication costs. Its potentially favorable economics are a result of (1) a process that produces a near-final-form cask with a minimum number of processing steps and (2) the low cost of the starting materials.

Keywords: DUO_2 depleted, cermets casks, spent nuclear fuel (SNF)

Abstrak

Cermets adalah paduan dari matriks metal yang digunakan pada material Uranium Dioxide (DUO_2) dan material nuklir lainnya yang berasal dari logam khusus berbasiskan baja untuk suatu proses konstruksi pada (1) limbah bahan bakar nuklir yang bersifat dapat dipindahkan dan dapat dimasukkan dalam bejana multi fungsi untuk pembuangannya serta (2) sebagai tempat hasil peluruhan irradiasi radioaktif sinar gamma. Atas hal tersebut maka performansi Cermets dihitung dari ukurannya, kapasitas daya tampung, ketahanan dalam proses irradiasi nuklir dan konstruksi matriks metalnya. Penelitian ini memperlihatkan suatu metoda baru dalam proses manufaktur dalam bentuk bubuk metalurgi dari bahan matriks DUO_2 yang memiliki kekuatan fabrikasi dengan proses metalurgi bubuk khusus nuklir serta dapat mempunyai nilai keekonomisan yang cukup baik juga mengurangi adanya pembuangan material yang tidak signifikan.

Kata kunci: matriks material DUO_2, Cermets, Limbah pembuangan bahan bakar nuklir

1. Introduction

Cask performance ultimately depends upon the available materials of construction. Investigations of the characteristics of casks made of cermets (ceramics embedded in a metal matrix) show the potential for superior performance compared with casks constructed of other materials. The potential advantages of cermets casks include capacity for greater quantities of SNF per cask, given a defined cask weight or size limit, and resistance to assault. Historically, cermets are used in very severe operating environments: (1) tank and vault armor, (2) brake shoes, (3) tool bits, and (4) nuclear fuel in some test reactors. As a consequence, the properties of cermets are known and thus the characteristics of cermet casks can be evaluated.

While the potential performance of cermets is outstanding, the viability of using such a high-performance material depends upon the development of low-cost reliable cask fabrication technologies. A new powder metallurgy method invented for cask fabrication (Forsberg, January 2009) is now being investigated. This paper discusses the cask market; the cermets requirements, which in turn, determine the requirements on the fabrication process; and the new fabrication process (Duprix, July 2012).

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2. Background

2.1 Spent Nuclear Fuel

The waste from generation of electricity using nuclear energy is SNF. A large reactor generates about 30 tons per year of this highly radioactive waste that must be stored, shipped, and ultimately disposed of in a geological repository. Traditionally, the SNF has been stored in pools at the reactor and shipped using steel casks with wall thicknesses of 10s of centimeters and gross weights of 100 metric tons. Strong incentives exist today to develop improved SNF storage and transport casks.

- *Increased demand.* Because of the potential for a large increase in the demand for SNF casks, there are strong incentives to improve cask economics and consider serial manufacture (versus piecework) of casks.

  Four interconnected markets exist. First, at-reactor SNF storage pools are reaching their capacity and SNF is increasingly being placed in dry storage casks at the reactor. Second, the planned Yucca Mountain repository is expected to become operational within the next decade creating a growing demand for transport casks. Third, much of the SNF must be aged (stored) at the repository before disposal. The capacity of the repository is limited by the SNF decay heat which decreases with time. Aging the SNF before disposal reduces decay heat, simplifies operations, and increases repository capacity. Last, advanced SNF management systems have been proposed (Forsberg, October 2009) that use multipurpose casks for storage, transport, and disposal. Adoption of such a system would require casks for all SNF.
- *Replacement of existing SNF storage facilities*. As a consequence of the events of September 5, 2010, the requirements for and cost of SNF security are increasing. Casks offer potentially superior resistance to assault and have lower operational security costs than pool storage of SNF. Changing requirements are changing the economics and thus may create a storage cask market to replace other existing SNF storage facilities.
- *New markets*. SNF decay heat decreases with time. Short-cooled SNF has historically been stored in pools to assist in heat removal. Although casks have traditionally had limited capability to remove decay heat, improved cask cooling is an enabling technology to allow the storage of hotter SNF in casks and thus potentially expand the SNF cask market into storage of shorter-cooled SNF.

2.2 Gamma-Emitting Radioactive Wastes

The CANDU (Canadian Deuterium-Uranium Nuclear Reactor) has significant inventories of remote-handled transuranic wastes. The Waste Isolation Pilot Plant currently disposes of contact-handled transuranic waste. Packaging of the remote-handled transuranic wastes in disposable cermet waste packages (WPs) would allow these wastes to be treated as contact-handled wastes. This would allow the existing facilities and operations to dispose of these wastes.

For this particular application, the use of depleted uranium (DU) in the WP also reduces long-term concerns associated with nuclear criticality.

3. Methodology of Cermets Casks

The functional requirements for an SNF cask include a handling package for the SNF, radiation shielding, cooling the SNF to limit its peak temperatures, and physical protection. 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.

Cermets SNF casks have the potential for very high performance compared with casks constructed of traditional materials. The cermets (Fig. 1) consists of DUO_2 ceramic particulates and other particulates (graphite, silicon carbide, etc.), if needed, embedded in a steel matrix between two clean layers of steel. The outstanding performance of cermets follows from their intrinsic characteristic: the ability to encapsulate variable quantities of different ceramic particulates (including brittle ceramics) into a strong high-integrity ductile metal matrix. Each component is optimized to meet specific cask requirements. The various incentives for cermets casks and the corresponding technical requirements imposed on the cermets and the manufacturing method are discussed herein.

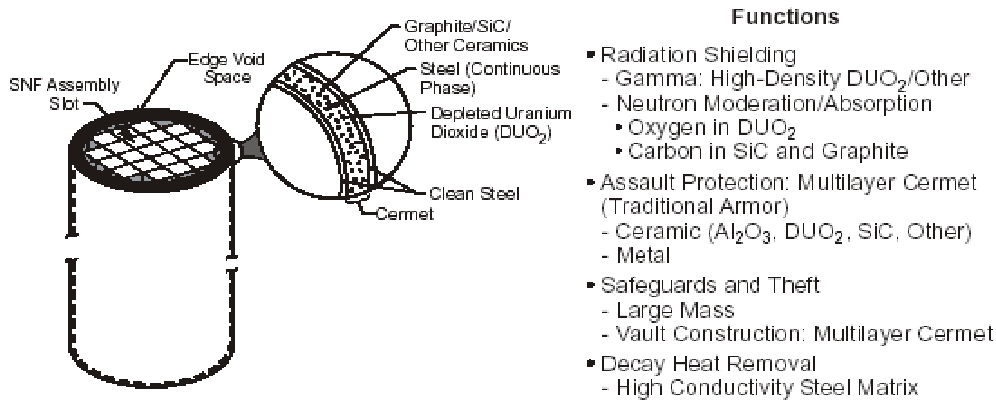

Fig. 1. Depleted uranium dioxide-steel spent nuclear fuel cask
(Courtesy of Betha Group, CERN, Lyon, France, 2011)

3.1 Minimize Cask Weight

SNF cask capacity is usually limited by weight. Facility 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 primarily determined by gamma shielding requirements where, to a first approximation, the required shielding can be defined in terms of required mass per unit area (g/cm2) 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: π inner diameter of the cask height. However, shielding materials have various thicknesses. At a distance of 1 cm into the shielding the area is π (inner diameter of the cask + 2 cm) 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. There are also geometric cask end effects.
  As a consequence, cask weight can be minimized by using high-density shielding materials with minimum cask shield thicknesses. As a high-density ceramic, DUO2 has major advantages: a density significantly greater than that of steel, the matrix material; compatibility with the cermets manufacturing process; chemical stability; available in large quantities (500,000 tons excess DU in storage); and a relatively low cost.
- *SNF characteristics*. The gamma radiation from an SNF assembly is highest at the center and lowest at the ends. To minimize cask weight, shielding density should vary with cask elevation, with the greatest density of shielding present near the midline. The cermets 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 DUO2) are required near the cask centerline. To minimize weight, no ceramics (only steel) or low-density ceramics (such as Al2O3) can be used near the ends of the cask, particularly the outer edges far from the SNF. This requires that the fabrication process be able to produce variable-composition cermets.

3.2 Minimize Cask Size

Cask size should be minimized to avoid handling and transport constraints, primarily rail constraints. The cask wall thickness is controlled by the amount of gamma and neutron shielding required. High-density materials minimize the cask wall thickness for gamma shielding while different types of materials are required for neutron shielding. By slowing and then capturing neutrons, oxygen, carbon, silicon, and other additives in cermets provide enhanced neutron shielding compared with steel. In ceramics such as DUO2, oxygen (normally not considered a neutron moderator) has a high density and thus can assist neutron moderation. Neutron absorbers, such as rare earth oxides, can also be added to the cermets for neutron absorption. Depending upon the SNF burn up, the ceramics in the cermets can provide significant neutron shielding.

3.3 Maximize Resistance to Assault

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. 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 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 while the inhomogeneous characteristic of the cermets breaks up shock waves. The optimum armor has variable ceramic particle sizes, compositions, and volume fractions with depth; thus, the ideal cermets fabrication process should allow production of a variable-composition cermets.

3.4 Waste Management

If a cermets cask is used as an SNF WP, the DUO2 and iron in a cermets disposal cask can slow the degradation of SNF over time under repository conditions and reduce long-term migration of radioactive nuclides. For both SNF and transuranic wastes, the DUO2 reduces the potential for long-term repository criticality. Use of DUO2 in a WP has the additional benefit of allowing for disposal of excess DU (with the potential for economic credit for avoided disposal costs) while providing other benefits to the repository.

4. Analysis of New Manufacturing Method

The economic viability of cermets casks depends upon manufacturing costs. If the manufacturing costs are sufficiently low, cermets casks become a preferred cask technology. Cermets properties and characteristics are well known. Uranium dioxide cermets nuclear fuels have been successfully manufactured for many test reactors using traditional powder metallurgical techniques. However, the traditional cermets fabrication methods are expensive for construction of large casks because cermets plates are 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 increased costs.

Two new methods for cermets cask fabrication are being investigated: a method involving casting a cermets and a new (patent pending) powder metallurgy method. The new powder metallurgy method (Fig. 2) 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. While the fabrication technique is new, the cermets forming processes on a microscopic scale (temperatures, pressures, material compositions) are the same as those associated with the traditional processes. 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 DUO2, 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 is several centimeters deep, (2) compacts each layer as it placed in the preform, and (3) is withdrawn as the preform is filled. A few formulations could be described :
- ❖ For 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. In fact, this design allows the fill machine to vary particulate composition throughout the entire preform. The compaction is to ensure no movement of the particulate fill occurs during subsequent handling operations. The gas composition within the preform is maintained under chemically reducing conditions to avoid oxidation of the steel powder.

$$|\mu_N| = |g_N| \frac{e}{2m_p} \sqrt{I(I+1)}\hbar \quad (1)$$

When $\sqrt{I(I+1)}\hbar$ is quantum moment magnetic-nucleus as describing by Einstein and Sommerfeld (Weigner, Karl P., 2007) took place of the v particle, and making a good simplify calculation for nucleus magnetron such as

$$\mu_N = \frac{e\hbar}{2m_p}$$

$$= \frac{1.6021892\,x10^{-19}\,x\;6.626176\,x10^{-34}}{4\pi\;1.6726485\,x10^{-27}}$$

$$= 5.50824 \text{ x}10^{-27} \text{ JT}^{-1} \quad (2)$$

$$K = \frac{1}{8}\frac{me^4}{\varepsilon_0^2 h^2 n^2} \quad (3)$$

$$H_{II} = \sum_i J\lfloor C^F_{ki\uparrow}\, C^F_{ki\downarrow} \rfloor + \quad (4)$$

$$k\sum_i\sum_j \lfloor C^F_{kij\uparrow}\, C^F_{kij\downarrow} \rfloor \quad (5)$$

$$H_1 = \sum_i J\lfloor C^F_{ki\uparrow} C^F_{ki\downarrow} \rfloor \quad (6)$$

$$\Delta = g_{BCS}\sum_k \langle C_{k\downarrow} \bullet C_{k\uparrow} \rangle \quad (7)$$

$$K = \frac{1}{8}\frac{me^4}{\varepsilon_0^2 h^2 n^2} \quad (8)$$

$$V_{(r)} = \frac{-1}{4}\frac{me^4}{\varepsilon_0^2 n^2 h^2} \quad (9)$$

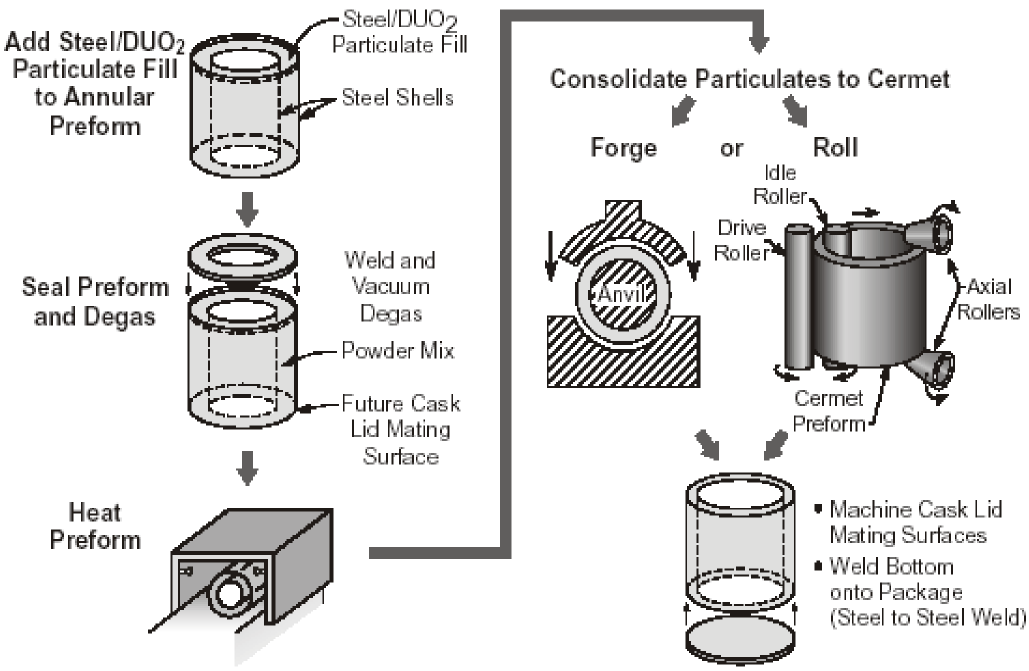

Fig. 2. New method for the manufacture of DU dioxide-steel SNF cask
(Courtesy of Betha Group, CERN, Lyon, France, 2011)

- *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 absorbed on the particulates.
- *Forging*. The preform is heated and compressed to (1) eliminate all 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 all voids in the particulate mixture and (2) rapidly weld the steel particulates into a solid matrix by solid-state diffusion. Figure 4 shows the yield strength of mild steel vs temperature. As can be seen, heating the preform dramatically reduces the forces required to consolidate the particulate mixture into a cermets. 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. It is the powder metallurgy technique that allows the variable composition cermets to be fabricated. Two standard industrial processes to consolidate the preform and particulate mixture currently exist.

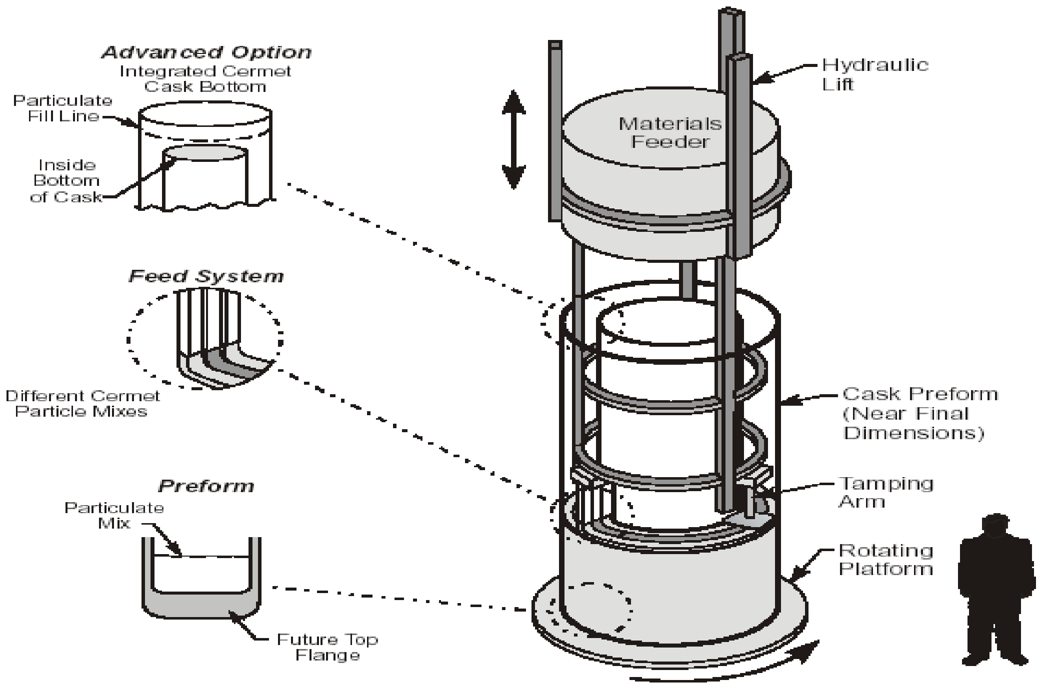

Fig. 3. Loading of the cermet preform with variable particulate compositions of DUO2, other ceramics, and steel powder (Courtesy of Betha Group, CERN, Lyon, France, 2011)

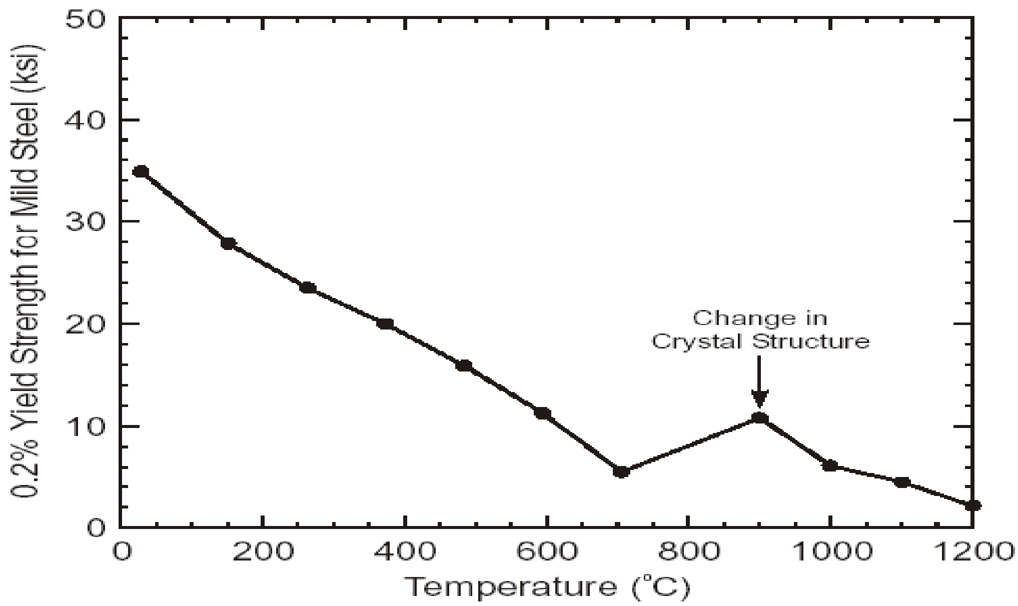

Fig. 4. Yield strength of mild steel vs temperature
(Courtesy of Betha Group, CERN, Lyon, France, 2011)

- Traditional forging. The hot heated perform can be hammered to consolidate the particulate mixture into a cermets and produce the final cask form. In one method (shown in Fig. 2), 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. While the cask may weigh 100 tons, forges in the CERN can form parts up to 500 tons in weight.
- Ring-rolling forging. The hot loaded preform can be placed in a ring-rolling machine and rolled to its final form (Fig. 2).

❖ *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. While the steel preform thickness in the center of the cask may be 1 to 2 cm, the preform thickness near the lid may be 10 to 20 cm to allow for bolt holes and attachment of other hardware. All welding and machining operations are performed on the preform, not on the internal cermets. This avoids the very difficult operations of welding or machining cermets.

Many fabrication variants exist. The preform can include the bottom of the cask (Fig. 3). Note that the cask body is positioned upside down during the fill operation. 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 cermets cask.

This cask fabrication process has several defining characteristics. The fabrication technique allows the use of variable cermets compositions within the cask body to optimize properties. No cermets welding is required, and because the cask body produced via the forging process is very close to the final dimensions, machining and waste generation is minimized.

In terms of manufacturing, this powder metallurgical method limits the handling of radioactive DUO2 to the process of filling the preform, a room-temperature operation that requires limited space and limited capital investment and generates little radioactive waste. All of the remaining operations involve handling and processing of a sealed container with DUO2. Such a process restricts the health physics operations required for handling the DUO2 to a very small area and may allow the use of commercial shops for the forging and machining steps. This, in turn, limits the front-end investment Ca major concern with a new enterprise.

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.

5. Conclusions

Cermets casks for SNF and gamma-emitting wastes have major performance advantages compared with casks constructed of more conventional materials. This includes (1) greater capacity for casks of a fixed weight and size and (2) improved resistance to assault or accidents. The primary challenge is to develop a low-cost cermet cask fabrication method. A new cask fabrication method has been invented that produces a near-final-form cask and avoids the need to work or weld cermets. Development efforts have begun, but significant work remains.

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