

EVALUATION OF FILLER MATERIALS TO CONTROL POST-CLOSURE CRITICALITY OF DUAL-PURPOSE CANISTERS

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The US Department of Energy's Used Fuel Disposition Campaign is assessing the technical feasibility of direct disposal of dual-purpose canisters (DPCs) in a geologic repository. While canister designs attempt to eliminate water intrusion, analysis of these designs must include the assumption that water can intrude into and flood a canister. The identification of canister fill materials that could be used in existing and future DPCs to mitigate the potential for post-closure criticality in the disposal environment was one of the key research and development needs identified. An important observation is that a filler material should occupy most of the free DPC volume to preclude criticality over the repository time frame. Based on this analysis, the most promising fill materials for use in DPCs are low-melting-point metals, such as Pb/Sn, Sn/Ag/Cu, or Sn/Zn, and small solids, such as glass beads containing depleted uranium or UO₂ particulates.

I. INTRODUCTION

The US Department of Energy's Used Fuel Disposition Campaign (UFDC) is assessing the technical feasibility of direct disposal of dual-purpose canisters (DPCs) in a geologic repository. The DPCs are currently in use for dry storage of spent nuclear fuel at commercial sites and are suitable for storage and transportation in accordance with federal regulations. The ability to provide direct repository emplacement of the DPCs is conditional on the geologic disposal media, package spacing, ventilation system, and other attributes of the repository¹. The identification of canister fill materials that could be used in existing and future DPCs to mitigate the potential for post-closure criticality in the disposal environment and time frame, through moderator displacement and or neutron absorption, was one of the key research and development needs identified in a recent assessment of canister disposal alternatives².

II. TECHNICAL DISCUSSION

The UFDC is evaluating direct disposal of DPCs that are currently licensed for storage and transportation of used nuclear fuel (UNF) or spent nuclear fuel (SNF). There are many designs currently in use commercially, and there are strong incentives for disposing of this fuel without repackaging. The program is considering disposal in generic repositories, including crystalline rock, salt, clay, sedimentary rock, and hard-rock geologic environments.

Addition of filler to canisterized fuel has been considered extensively since the original concepts for disposal and dry storage of fuel were conceived. An earlier study performed by Maheras et al.³ evaluated the use of filler materials to meet 10 CFR Part 71 and 72 requirements to allow extended storage and subsequent transport of the UNF or SNF. This study also identified categories of materials potentially suitable for canister filling and evaluation criteria of material performance during storage and transportation.

For disposal, the geologic timescale following repository emplacement makes post-closure criticality a concern⁴. While canister designs attempt to eliminate water intrusion, analysis of these designs must include the assumption that water can intrude into and flood a canister during transport, and design features (e.g., flux traps, neutron absorbers) are provided to prevent nuclear criticality under flooded conditions. Due to degradation and mobility of these materials in certain environments, these features cannot currently be credited with maintaining subcritical configurations following disposal because of the time frames to consider (i.e., thousands of years). Conservative assumptions for fuel degradation and burnup could provide a basis for the addition of filler materials to the DPCs for post-closure criticality safety. A discussion of candidate filler materials to meet this objective is provided in section II.C.

In addition to criticality control, advantages of filler materials in emplaced DPCs could also include the following⁵:

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- Chemical buffering to reduce radionuclide migration following breach of containment barriers and water intrusion into the waste package (WP)
- Cathodic protection, by use of filler material with the highest electrochemical activity, in the event of water intrusion to minimize the effects of corrosion
- Mechanical support to maintain geometry, inhibit movement, and retain structural integrity of the fuel and WP
- Improved heat transfer to protect fuel cladding and engineered barriers in the disposal system. (This will diminish in importance as the fuel cools.)

II.A. Filler Material Requirements

Requirements for filler materials to be provided at repository emplacement are similar, but not identical, to those for use in storage and transportation. Table I compares potential evaluation metrics for filler materials when applied to storage and transportation with those for avoidance of post-closure criticality. This table is an adaptation of metrics proposed by Maheras et al.³ and highlights potential changes in the evaluation criteria for filler materials used for stabilization for extended storage and subsequent transport vs. those used at the repository site for repository post-closure criticality avoidance. Evaluation criteria for filler material performance include two general groups⁶: those required for criticality control (Table II) and other general attributes of the materials (Table III).

II.B. Canister Filling Requirements

Filling operations are assumed to require access to sealed canisters received at the repository. For existing canisters, access can be obtained through the vent and drain lines, removal of the canister lid to expose the fuel, or a new penetration into the top or sidewall of the canister. The access selected may depend on the physical and chemical form of the selected filler material and the DPC design variant.

Placement of the filler material must ensure⁷

- no degradation of canister structural integrity,
- no damage to the fuel,
- a minimum of 60% void space fill, which is comparable to 90+% fill considering interparticle voids,
- filling can be completed without requiring repackaging,
- measurement of completeness of void fill is allowed for,
- reseal of DPC is allowed for, and
- the inert atmosphere in the DPC is reestablished to provide corrosion protection and heat transportation while the canister is intact.

II.C. Candidate Filler Materials

Extensive investigation has been conducted into the identification and evaluation of canister filler materials to support storage, transportation, and disposition of SNF^{3, 6, 8-10}. Potential materials that can satisfy the criteria described above are assumed to be liquids or molten materials that solidify upon chemical reaction or cooling, or solids introduced directly. Although liquids and gases have been evaluated previously, they are not considered to be viable candidates because of postulated WP degradation in the disposal environment.

It may be desirable that the melting point of molten filler material be high enough that the residual heat from the fuel would not keep the fill material in the molten state. However, filler materials that remain molten for some period of time while the fuel continues to cool in the repository may be acceptable if it can be shown that they are not displaced with water in the case of a canister breach.

Examples of filler materials introduced as liquids which then solidify include both metals (tin, lead, zinc, and alloys) and non-metals (e.g., plastics, resins, foams, and grout). Materials introduced as solids include minerals, metal, glasses, and clays. The suitability of a particular material is not only dependent on its physical and chemical properties but also on the properties of the fuel, DPC geometry, and the disposal environment. The choice of filler material may vary if the principal performance requirement changes from criticality mitigation to water displacement or radionuclide sequestration.

Lead and tin are some of the most common low-melting-point metals. By combining lead and tin in the proper ratio, a lower-melting-point eutectic can be formed. The Sn₆₃ Pb₃₇ alloy, a solder used in electronics, has a melting point of 183°C. A number of lead-free solders are also available. These include Sn_{95.6} Ag_{3.5} Cu_{0.9} with a melting point of 217°C and Sn₉₁ Zn₉ with a melting point of 199°C. The melting point of zinc is 419.5°C, and the zinc-aluminum eutectic alloy is 382°C. Table IV provides a summary of possible liquid fill materials and their melting points.

A description of the Canadian nuclear fuel waste management program was provided by Forsberg¹¹ and indicated that during a 15-year development program of its repository concept for CANadian Deuterium Uranium (CANDU) reactor SNF, the use of a glass-bead or silica-sand fill was explored. Similarities and differences between the Canadian and US designs are described. In both concepts, the fuel is placed in the empty WPs. The void space between the fuel pins and the outer void space between SNF assemblies and the inner WP wall would be filled with small particles. The WPs are then sealed and placed into the repository. The CANDU and light-water-reactor (LWR) SNF assemblies are both Zircaloy-clad UO₂ fuel with significantly different geometries. The Canadian

Table I. Comparison of DPC Filler Metrics Relevant to Transportation and Post-Closure Criticality Avoidance

Evaluation Criteria for Candidate Canister Fill Material	Elements	Relevant to Storage and Transportation	Relevant to Avoidance of Post-Closure Criticality
Criticality Avoidance	<ul style="list-style-type: none"> • Provide moderator exclusion • Neutron absorption capability • Minimize neutron moderation • Provide dilution of fissile radionuclides • Capacity to fill over 60% of the inner free volume of the canister • Fill material does not compact by more than 10% of its original volume under its own weight or as the result of shipping or handling 	X X X X X	X Secondary Secondary Secondary X X
Heat Transfer or Thermodynamic Properties	<ul style="list-style-type: none"> • Promote heat transfer from the fuel • Thermal stability • Chemical stability • Radiation stability • Chemically compatible with fuel cladding, fuel, neutron poisons, fuel baskets, and other structural materials within canister 	X X X X X	Secondary Secondary Secondary Secondary Secondary
Homogeneity and Rheological Properties	<ul style="list-style-type: none"> • Homogeneous batches • Good rheological properties to ensure proper filling • Ability to be placed in the canister without damaging fuel assemblies 	X X X	X X X
Retrievability	<ul style="list-style-type: none"> • Allows for safe retrieval of UNF from a canister without need to resort to time-consuming or costly measures and without further compromise of the integrity of UNF assemblies 	X	NA
Material Availability and Cost	<ul style="list-style-type: none"> • Low cost • Material available in required purity 	X X	X X
Weight and Radiation Shielding	<ul style="list-style-type: none"> • Fill material does not add significantly to the weight of the container/cask system • Good radiation shielding properties 	X X	NA NA
Operational Considerations	<ul style="list-style-type: none"> • Easy to emplace • Fill material does not adversely react to normal conditions of transport or hypothetical accident conditions 	X X	X NA

“The “X” indicates the associated element in the criteria list is highly important. Differences between the last two columns are highlighted in red.

Table II. Criticality Control Criteria

Attribute	Basis
Low water solubility	Prevents dissolution and migration of filler following package degradation and water inflow
Low compaction ratio	Ensures that the void volume remains constant after filling
Low neutron moderation	Low content of hydrogen or other light elements allows neutrons to escape the package
High neutron absorbance	Presence of neutron absorbers (B, Cd) can reduce the Δk_{eff} of the system
Water exclusion	Filler material may physically block the introduction of water to a degraded package
Isotope dilution	Depleted uranium can reduce the bulk ^{235}U concentration from failed fuel

Table III. General Performance Criteria

Attribute	Basis
Thermal, chemical, mechanical, radiological stability	Ensures that the filler properties are consistent and compatible with application
Good rheological properties	Provides for continuous, replicable filling of the canister
Good heat transfer	Reduces thermal loading on fuel cladding
Chemical compatibility with, fuel, WP, and disposal environment	Reduces corrosion and delays/eliminates the migration of radionuclides to the environment
Material availability	Provides domestic supplies of sufficient quantity and purity, with minimal environmental impacts
Material cost	Ensures incremental cost of material is insignificant with cost of filling and disposal of the WP

Table IV. Melting Points of Potential Liquid Fill Materials

Candidate Liquid Fill Material	Eutectic	Melting Point (°C)
Pb		327.6
Sn		231.9
Zn		419.5
Pb-Sn (60/40) (common solder)		188
Pb-Sn (37/63)	Yes	183
Sn-Ag-Cu (95.6/3.5/0.9)	Yes	217
Sn-Zn (91/9)	Yes	199
Zn-Al (95/5)	Yes	382
Paraffin		46–68

WP fill requirements were that the material fill all empty spaces within the WP and that it be chemically inert, structurally strong, and inexpensive. The fill was also required to support the WP wall against 10 MPa of external hydrostatic pressure.

The fuel assemblies for CANDU reactors and LWRs have a number of differences, but the fuel, materials of construction, and other features are similar. A key difference is that the internal clearances between pins in the CANDU fuel assembly are, on average, smaller than those of pressurized water reactor (PWR) assemblies. In the CANDU fuel assembly, the minimum clearance between rods is 1.3 mm, while in a standard PWR fuel assembly, the clearance is 3.4 mm. As a result, it is expected that filling the void spaces inside the CANDU fuel assemblies would be more difficult than filling the void spaces within PWR fuel assemblies. In addition, only 41% of the cross section of the PWR fuel assembly is composed of pins, while pins compose 61% of the CANDU fuel assembly cross section¹¹.

Fill tests were conducted to define the particle size, vibration frequencies, and accelerations that provided the most rapid and reliable filling. These tests indicated that the maximum practical packing density was about 70%. Vibratory filling was identified as the preferred option for this type of WP as it provided higher fill densities and shorter fill times¹¹.

The review of possible filler materials for transportation and storage by Maheras et al.³ indicated that the Canadian program considered three fill materials as viable: glass

beads, Interprop[®], and sintered bauxite. Coarse glass beads generated the least amount of dust during compaction and produced the highest bulk modulus of elasticity in the compacted state. The glass beads were selected as the candidate fill material; however, they were later abandoned because glass beads could not provide the necessary assurance that the WP would not collapse. If the structural requirement is removed, then glass beads may still be viable for the purpose of moderator exclusion.

Limited testing has been performed using filler materials on surrogate fuel and canister assemblies. An extensive test program will be required to ensure that all performance requirements for materials, fuel, and WPs are satisfied. In addition, the performance of the filled canister must be evaluated for each disposal environment under consideration to determine the post-closure impacts to the repository.

II.D. Criticality Analysis of Filled DPC

After disposal, features, events, and processes (FEPs) that can affect repository performance are evaluated, and criticality is one such FEP. Criticality potential is measured by the effective neutron multiplication factor (k_{eff}) of a system (also referred to as reactivity in this report). System reactivity for commercial LWR fuel typically increases in the presence of a moderator (e.g., water), which allows neutrons to slow down to energies where they can easily be absorbed by fissile isotopes and cause additional fission reactions. When $k_{eff} = 1.0$, the system is considered critical.

Note that DPC post-closure criticality is plausible because of potential groundwater infiltration of the DPC and the associated material and structural changes that can occur as a result of that infiltration over the repository time frame of thousands to hundreds of thousands of years. Therefore, an engineering option to mitigate the potential for DPC post-closure criticality is to fill the canister cavity with engineering filler materials that can prevent flooding of the DPC (moderator displacement) over the repository time frame. Filler material can also be selected to provide neutron absorption in addition to its moderator displacement functionality.

Previous studies¹⁰ suggest that more than 50% of the void (free volume) must be filled to prevent sufficient water moderation to support criticality. This section investigates 1) the moderator displacement and 2) neutron absorption aspects of the filler materials.

II.D.1 Codes and Methods

The criticality analysis is performed using burnup credit. Taking credit for the reduction in reactivity because of fuel burnup is commonly referred to as burnup credit. Burnup credit criticality safety analysis for UNF in storage systems requires the determination of isotopic number densities for fuel assemblies by simulating assembly-specific irradiation histories, commonly known as a depletion calculation. A depletion calculation is followed by a canister criticality evaluation, which uses the isotopic number densities of the fuel from the depletion step to determine the neutron multiplication factor. Both of these calculations—depletion and criticality—require different tools and methods.

The SCALE¹² code system is used for the criticality analyses presented in this paper. The TRITON two-dimensional (2D) depletion sequence is used to perform depletion calculations that generate cross-section libraries for generic assembly/reactor-specific classes and a range of fuel operating conditions. This information is subsequently used by ORIGEN for processing of problem-dependent cross sections. The TRITON 2D depletion calculation sequence employs CENTRM for multi-group cross-section processing, NEWT for 2D discrete-ordinates transport calculations, and ORIGEN for depletion and decay calculations. The resultant nuclide concentrations are passed to the criticality analysis codes. The SCALE CSAS6 criticality analysis sequence is used to perform criticality calculations for a loaded fuel cask using the KENO-VI Monte Carlo code with the continuous-energy ENDF/B-VII.0 cross-section library to determine the effective neutron multiplication factor, k_{eff} . Note that a prereleased version of SCALE 6.2, which is under development, is used for decay and continuous-energy criticality calculations.

The isotope set, credited in the criticality calculations, is selected based on the burnup credit isotopes recommended by NUREG/CR-7108 and -7109¹³⁻¹⁴.

Additionally, bounding burnup-dependent axial profiles are used for the criticality analyses¹⁵.

II.D.2 Criticality Scenarios

An important assumption for criticality analysis is that water enters a WP at some point over the repository time frame. While the different geologic settings and material degradation mechanisms might yield a large number of potential configurations, two simplified and conservative configurations are used in this report to assess both moderator displacement and neutron absorption aspects of the DPC filler materials:

1. total loss of neutron absorber panels from the internal basket (resulting from unspecified degradation and material transport processes) and
2. loss of the internal basket structure (including the neutron absorber) resulting in elimination of assembly-to-assembly spacing.

In this paper, the aforementioned two configurations (degradation scenarios) are analyzed for a representative DPC flooded with freshwater and filled with different levels (volumes) of filler materials. The actual extent of basket material degradation that must be accounted for should be revisited in the future when more thorough corrosion data will be available under identified repository conditions and time frames.

A canister system representative of one of the Holtec International's multi-purpose canisters (MPC) is selected for the filler material criticality study. Specifically, the MPC-32 was selected. The MPC-32 (a type of DPC) criticality model is modified to represent the above two scenarios. Two representative filler materials are considered that include aluminum and boron carbide (B_4C). While aluminum only provides water displacement, B_4C provides both water displacement and neutron absorption. Only the aluminum results will be presented as these are the most conservative. Packing densities of 58% and 68% are considered. Packing density is modeled as a volumetric mixture (e.g., 58% aluminum is modeled as a mixture of 58% by volume aluminum powder and 42% by volume water to form an aluminum slurry). The criticality analysis is performed for a uniform canister loading. Note that the criticality calculation presented in this paper is a representative calculation to show the reactivity reduction trend with increasing volume fraction of filler material. Figure 1 depicts the cross section of the canister containing filler material with the complete loss of neutron absorber as modeled in KENO VI. It is assumed that the filler material for the loss-of-neutron-absorption scenario uniformly fills all the basket cells. The corrosion products from the basket materials are not credited in the criticality analysis. Modeling was also performed for the canister with complete loss-of-basket structure (Figure 2).

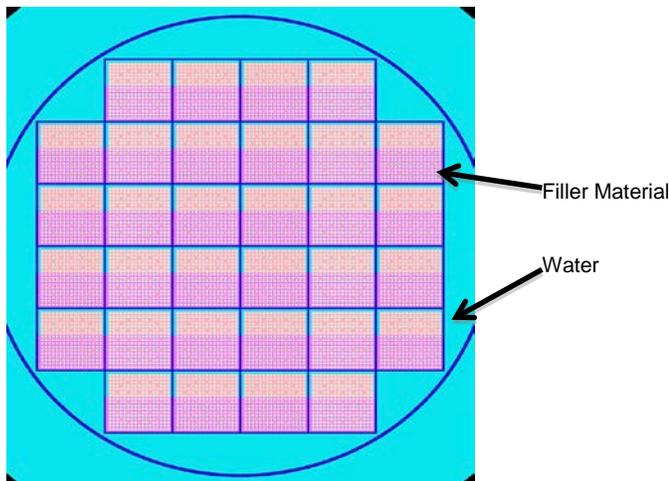


Figure 1. Graphical depiction of the center plane of the MPC-32 KENO model with complete loss of neutron absorber used for filler materials study.

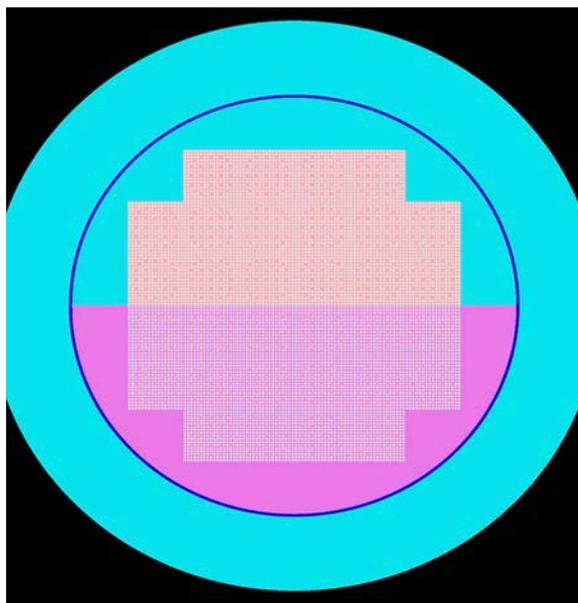


Figure 2. KENO depiction of the MPC-32 degraded basket scenario with filler material.

II.D.3 Criticality Results

The volume fractions used to present the criticality results depend on the degradation scenario. The volume fraction is calculated by dividing the volume of the filler material in a basket cell by the free volume of that basket cell for the complete loss-of-neutron-absorber case. The free volume of a cell is calculated by subtracting the volume of the assembly from the total volume of the cell. Note that assembly volume only includes the volume of the active fuel region (fuel rods, guide tubes, and instrument tubes) without the spacer grids. Guide tubes and instrument tubes are modeled as filled with water. The filler material volume in a

cell is calculated by subtracting the part of the assembly volume covered by the filler material from the filler material volume. On the other hand, the volume fraction is calculated by dividing the filler material volume by the free canister volume for the fully degraded basket scenario. Figure 3(a) and 3(b) present the reactivity reduction, presented in terms of negative Δk_{eff} , as a function of fractional aluminum volume in the MPC-32 with complete loss of neutron absorber and degraded basket, respectively. Note that for simplicity, biases and uncertainties of the criticality analysis presented in this paper are ignored. Figure 3(a) indicates stepwise reactivity reduction with increasing filler volume for the loss-of-neutron-absorber scenario, whereas Figure 3(b) shows that the reactivity reduction up to a certain threshold volume fraction is insignificant and significant reactivity reduction beyond the threshold for the degraded basket scenario. However, for both the cases a significant volume fraction (>70%) is required to be filled by powdered aluminum to obtain a large reactivity reduction ($\Delta k_{eff} > -0.1$) that may be necessary to maintain subcriticality over the repository performance period. Note that criticality studies with as-loaded DPC configurations may provide more realistic volume fraction required to be filled by a filler material.

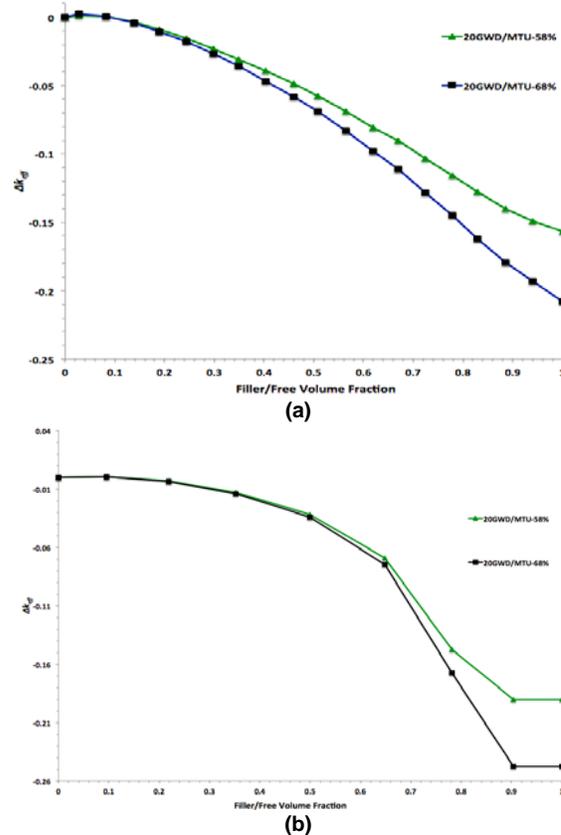


Figure 3. (a) Reactivity as a function of aluminum volume fraction for complete loss of neutron absorber; (b) reactivity as a function of aluminum volume fraction for degraded basket configuration.

II.E. Fill Methods

There are two fundamentally different approaches to filling the DPC. The first approach would utilize the vent plug and drain port as the means for introducing the filler material. Such an approach would minimize direct exposure to the fuel as the shielding would remain in place. However, some form of containment would still probably be required as there could be a potential for release of airborne particulates upon opening the DPC. For molten materials the ability to heat the DPC to above the melting point of the fill material would be required to prevent premature solidification.

The second approach would require the opening of the DPC. This operation would need to be conducted in a shielded facility, such as a hot cell. Forsberg et al.¹⁶ proposed that glass beads could be loaded into a fuel canister while it is open or added through a small hole in the lid once the canister is closed. Figure 4 shows this filling concept on a MPC after the canister is closed. In the Canadian WP fill station concept, the WP containing the SNF is placed on a vibratory table located in a hot cell facility and the particulate fill is added while the entire WP is vibrated. The particulate loading station hopper, metering device, and other components are located outside the hot cell. A filler hose that passes through the hot cell roof connects the hopper to the WP¹¹.

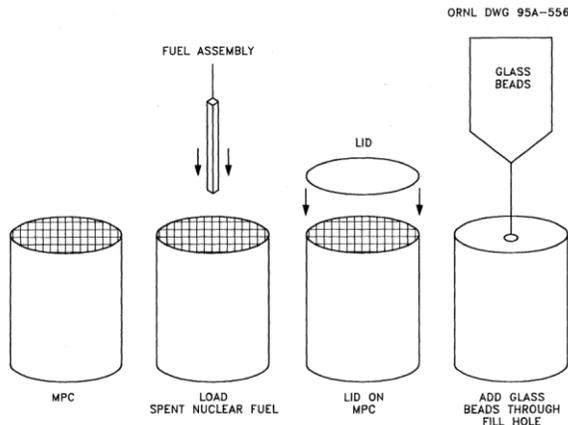


Figure 4. MPC canister filling concept¹⁶.

Under 10 CFR Part 72, all cask designs have some design features that would allow them to be opened to recover the fuel if necessary. Several options are available for a welded cask. These are described in reports prepared by AREVA¹⁷ and CB&I¹⁸ for the Department of Energy. The methods included

- plasma torch cutting,
- laser cutting,
- grinding,
- water jet cutting, and
- lathing (skiving) and end mill cutting.

It was assessed that all could effectively open a fuel cask, but several of the methods did not meet all of the following criteria¹⁸:

- not causing damage to the SNF,
- not creating a foreign material concern,
- not causing damage to the transfer cask,
- opening the cask within a reasonable time,
- performing the operation remotely or semi-remotely,
- allowing initial access to the vent and drain ports for water filling,
- maintaining the ability for post-unloading recovery of the cask, and
- accommodating cask geometry changes (e.g., warping).

The preferred method from both assessments was lathing. This operation has been demonstrated by AREVA on the NUHOMS[®]-type canister¹⁷.

III. RESULTS AND IMPLICATIONS

An important observation of the filler materials criticality study is that a filler material—regardless of whether it is a neutron absorber—should occupy most of the free DPC volume to provide criticality control over the repository time frame. Additionally, the eventual corrosion product(s) of a filler material and its criticality implication as a filler material selection criterion must be considered.

Based on this analysis, the most promising fill materials for use in DPCs for the avoidance of post-closure criticality are low-melting-point metals, such as Pb/Sn, Sn/Ag/Cu, or Sn/Zn, and small solids, such as glass beads. In the case of low-melting-point metals, provisions will be needed to heat the entire DPC and contents to temperatures of 225–250°C to ensure that the liquid flows to all parts of the container without solidifying on contact with surfaces. For solid feed, some provision for vibration of the entire DPC may be required to ensure adequate settling and complete filling of the container. Among the challenges are ensuring that the fill material is uniformly distributed and monitoring the distribution of the fill materials.

For current DPC designs, two potential filling methods are possible. The first is through the vent plug, but ensuring complete distribution of the fill material may be problematic. The second approach requires the removal of the lid from the DPC. Depending on the approach chosen, a separate hot cell facility may be required for receipt, filling, closure and post-closure testing of the DPC prior to emplacement in the repository, but in any case, some type of containment will be required to control a potential release from the DPC when it is opened or during the filling operation.

If pre-emplacement addition of filler materials becomes a programmatic requirement, canister designs should be developed that would include modifications to reduce the time, cost, and complexity of filling operations.

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