A Preliminary Evaluation of Using Fill Materials to Stabilize Used Nuclear Fuel During Storage and Transportation

Fuel Cycle Research & Development

Prepared for
U.S. Department of Energy
Used Fuel Disposition Campaign
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SUMMARY

The objective of the research described in this report was to conduct a preliminary evaluation of potential fill materials that could be used to fill void spaces in and around used nuclear fuel contained in dry storage canisters in order to stabilize the geometry and mechanical structure of the used nuclear fuel during extended storage and subsequent transportation. The use of fill material to stabilize used nuclear fuel is not considered to be a primary option for safely transporting used nuclear fuel after extended storage. However, the evaluation of potential fill materials, such as those described in this report, might provide the U.S. Department of Energy Used Fuel Disposition Campaign with an option that would allow continued safe storage and transportation if other options such as showing that the fuel remains intact or canning of used nuclear fuel do not prove to be feasible.

As a first step in evaluating fill materials, previous work done in this area was summarized. This involved studies done by the Spent Fuel Stabilizer Materials Program, Allied-General Nuclear Services, the Canadian Nuclear Fuel Waste Management Program, the U.S. Department of Energy, Spain, Sweden, and the Department of Energy's Yucca Mountain Project. A wide variety of potential fill materials were evaluated in these studies, ranging from molten metal to particulates and beads to liquids and gases. The common element in the studies was that they were focused on the use of fill materials in waste packages for disposal, not in storage canisters or transportation casks. In addition, very few studies involved actual experiments that measured some physical property of the fill material to be used as a stabilizing material, and no studies were found that analyzed the performance of transportation casks containing fill material during the normal conditions of transport specified in 10 CFR 71.71 or under hypothetical accident conditions specified in 10 CFR 71.73. In addition, most studies did not address issues that would be associated with production-scale emplacement of fill material in canisters, as opposed to laboratory- or experimental-scale use of fill material. It is noteworthy that Sweden abandoned its plan to use fill materials to stabilize waste packages due to the complexity of emplacing the fill material.

As part of the evaluation of fill materials, conceptual descriptions of how canisters might be filled were developed with different concepts for liquids, particles, and foams. The requirements for fill materials were also developed. Elements of the requirements included criticality avoidance, heat transfer or thermodynamic properties, homogeneity and rheological properties, retrievability, material availability and cost, weight and radiation shielding, and operational considerations.

Potential fill materials were grouped into 5 categories and their properties, advantages, disadvantages, and requirements for future testing were discussed. The categories were molten materials, which included molten metals and paraffin; particulates and beads; resins; foams; and grout. Based on this analysis, further development of fill materials to stabilize used nuclear fuel during storage and transportation is not recommended unless options such as showing that the fuel remains intact or canning of used nuclear fuel do not prove to be feasible.

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ACRONYMS

GWd/MTU	Gigawatt-day per metric ton uranium
HAC	Hypothetical accident conditions
k_{eff}	Effective neutron multiplication factor
NCT	Normal conditions of transport
UFDC	Used Fuel Disposition Campaign

DEFINITIONS

Alumina	Al_2O_3
Bauxite	An aluminum ore composed of primarily aluminum hydroxide minerals as well as mixtures of silica, iron oxide, and other impurities.
Bentonite	Bentonite is a natural clay that swells with the absorption of water and has good ion exchange properties.
Bondate	Bondate is an organic-based chemical bonding agent for aggregates and fibers.
Boron carbide	B_4C
Dowtherm	Dowtherm is a heat transfer fluid.
Hematite	α-Fe_2O_3
Interprop	Interprop is a ceramic proppant composed of 35-65% mullite (aluminum silicate) and 35-65% corundum (aluminum oxide).
Magnetite	Fe_3O_4
Mullite	$Al_6Si_2O_{13}$
Olivine	$(Mg,Fe)_2SiO_4$
Phosphates	$Ca_5(PO_4)_3(OH,F,Cl)$
Proppant	A proppant is a material that will keep an induced hydraulic fracture open.
Quartz	SiO_2
Rutile	TiO_2
Silica	SiO_2
Spinel	$MgAl_2O_4$
Wood’s metal	A low melting fusible alloy that is a mixture of 50% bismuth, 25% lead, 12.5% tin, and 12.5% cadmium.
Zeolite	Hydrated aluminosilicates of the alkaline and alkaline-earth metals.
Zircon	$ZrSiO_4$
Zirconia	ZrO_2

USED FUEL DISPOSITION CAMPAIGN

A PRELIMINARY EVALUATION OF USING FILL MATERIALS TO STABILIZE USED NUCLEAR FUEL DURING STORAGE AND TRANSPORTATION

1. INTRODUCTION

Used nuclear fuel generated by commercial nuclear power plants in the United States may continue to be stored for an extended period of time, potentially much longer than originally intended. The U.S. Department of Energy Used Fuel Disposition Campaign (UFDC) is tasked with developing the technical bases for to support the continued safe and secure storage and subsequent transportation of used nuclear fuel while maintaining options for its final disposition.

However, most storage pools for used nuclear fuel at reactor sites are now filled to capacity. To provide space for continuing discharges of used nuclear fuel, plant operators began transferring the used nuclear fuel into dry storage systems. These systems are located on the reactor plant's site external to the original nuclear plant facilities. In the dry storage systems, used nuclear fuel is stored in a dry, inert environment in bolted direct-load metal storage casks or in sealed metal canisters. The metal canisters containing used nuclear fuel are stored within steel-reinforced concrete overpacks or storage modules.

The majority of the used nuclear fuel that is in storage is classified as "intact fuel". Intact fuel is the U.S. Nuclear Regulatory Commission classification of used nuclear fuel where the defects in cladding that could expose nuclear fuel material to an oxidizing environment or could allow release of fuel particles and radionuclides from inside the cladding are limited to hairline cracks and pinhole leaks. Fuel assemblies that are classified as "intact" can be stored and transported without having to be additionally enclosed within a "failed-fuel" can within a transportation cask or transportable dry storage canister. In some cases, plant operators have placed used nuclear fuel into failed fuel cans then into storage canisters because it was not feasible to verify that the fuel met the requirements for "intact" fuel.

During extended storage, structures, systems, and components that are important to safety (including fuel cladding and fuel assembly structures) may degrade. The stressors, degradation mechanisms, and data gaps associated with extended storage and subsequent transportation are discussed by UFDC (2012). UFDC (2012) also discuss the stressors, degradation mechanisms, and data gaps associated with extended storage and subsequent transportation of high burnup fuel (exceeding 45 gigawatt-days per metric tonne of uranium [GWd/MTU]). Much of the fuel currently being discharged from reactors exceeds the high-burnup threshold and there is limited information available on the properties of this used nuclear fuel (UFDC 2012).

The focus of the gap analysis by UFDC (2012) is on evaluating the likelihood that the used nuclear fuel remains undamaged (i.e., intact, retrievable, and transportable) after extended storage. The ability of the used nuclear fuel to remain intact is especially important for assuring that a nuclear criticality cannot occur in a storage system or a transportation cask. If fuel cladding degrades during long-term storage, the geometric configuration of a fuel assembly and its fuel component could not be assured under normal conditions of transport (NCT) or hypothetical accident conditions (HAC). A change in the geometric configuration of the fuel inside a transportation cask would change the nuclear reactivity of the cask's contents and could compromise the ability to ensure that a nuclear criticality could not occur in the fuel during transportation.

The UFDC is conducting research and development aimed at developing objective technical evidence that can be used to project and assess the condition of used nuclear fuel during and following extended storage and subsequent transportation. This evidence is expected to show that used nuclear fuel can sustain extended dry storage in an inert atmosphere without substantial change in its properties. However, it is possible that the research will identify unexpected degradation mechanisms or will determine the condition of cladding for high-burnup used nuclear fuel such that the integrity of fuel cladding cannot be sufficiently verified for NCT and HAC.

Thus, the UFDC could consider other options to ensure that used nuclear fuel can be transported following extended storage. The range of these options includes: requiring that all used nuclear fuel assemblies be placed into failed-fuel cans before being placed into a dry storage cask or canister system and use of a fill material to stabilize the contents of a metal canister prior to transportation. Ideally, the use of a fill material would render the question of whether used nuclear fuel was intact or damaged immaterial because the fill material would preserve the geometric configuration of the used nuclear fuel and/or provide for moderator exclusion and thereby prevent a nuclear criticality. The objective of this report is to evaluate potential fill materials that could be used for this purpose.

There are several reasons why the use of a fill material might be preferable to options such as demonstrating that the used nuclear fuel remains intact or canning of all used nuclear fuel. For example, it may not be possible to provide objective evidence with the requisite reasonable assurance, at a reasonable cost, that used nuclear fuel will remain intact after extended storage. Under this circumstance, the use of fill material or canning of individual fuel assemblies might be the only options available that would allow transportation of large amounts of used nuclear fuel to a geologic repository, a consolidated storage facility, or a reprocessing facility. However, canning of used nuclear fuel would require repackaging of fuel already in storage and could also substantially increase the number of shipments. If feasible, the use of fill materials could be desirable when compared to canning and repackaging of used nuclear fuel.

There are also disadvantages to the use of fill materials. For example, placing a fill material in a metal canister subsequently loaded into a transportation cask could increase the weight of the transportation cask to the point where it could not be handled or transported. In addition, verifying that the fill material was fully and uniformly distributed within the metal canister may not be feasible. A closely related issue is that it may be difficult to load the fill material into metal canisters that were not designed with this capability, and it could be difficult to subsequently retrieve the used nuclear fuel without having to resort to time consuming or costly measures. The fill material would also have to be chosen so that it did not have undesirable properties during the normal conditions of transport specified in 10 CFR 71.71 or hypothetical accident conditions specified in 10 CFR 71.73, and the transportation casks would have to be re-licensed by the U.S. Nuclear Regulatory Commission based on the presence of the fill material, or more likely, entirely new transportation casks would have to be licensed.

For the reasons stated above, the use of fill material to stabilize used nuclear fuel is not considered to be a primary option for safely transporting used nuclear fuel after extended storage. However, evaluation of potential fill materials could provide the UFDC with an option that would allow continued safe storage and transportation if other options such as showing that the fuel remains intact or canning do not prove to be feasible.

2. SUMMARY OF PREVIOUS WORK

This section summarizes previous work done to investigate the use of fill materials to stabilize used nuclear fuel in waste packages, storage containers, or transportation casks. The studies that are summarized were identified by literature searches and searches of project records from available U.S. and international sources. Other work involving fill materials that is not available in the literature or project records is not included in the descriptions that follow.

The majority of the studies have been literature studies that did not involve experimental work. The only studies that involved experimental work were studies conducted by the Spent Fuel Stabilizer Materials Program, the Canadian Nuclear Fuel Waste Management Program, and the Yucca Mountain Project. In addition, the majority of studies were focused on the use of fill materials in waste packages for disposal of used nuclear fuel. No experimental work was found that analyzed the performance of transportation casks containing fill material during the normal conditions of transport specified in 10 CFR 71.71 or during hypothetical accident conditions specified in 10 CFR 71.73.

2.1 Spent Fuel Stabilizer Materials Program

The Spent Fuel Stabilizer Materials Program was conducted for the National Waste Terminal Storage Program, a predecessor to the Office of Civilian Radioactive Waste Management in the U.S. Department of Energy, and had the objective of identifying, testing, and selecting stabilizer materials for use in used nuclear fuel waste packages for disposal. Stabilizers were materials that would fill the space in a waste package that was not filled with used nuclear fuel (Fish et al. 1982).

Wynhoff et al. (1982) identified 34 candidate stabilizer materials based on analysis of thermal gradients within the waste package, thermal stress analysis (thermal gradient stress analysis and differential thermal expansion stress analysis), nuclear criticality, radiation attenuation, and cost and material availability. Table 1 lists these candidate materials. Fish et al. (1982) conducted a series of experimental tests and evaluated the 34 materials against the following functions:

- Help resist lithostatic and hydrostatic pressures on the waste package after emplacement
- Maintain the used nuclear fuel geometry, prevent motion and mechanical abrasion or rod failure due to handling and accidents
- Promote heat transfer from the fuel assembly to minimize fuel temperature
- Chemical compatibility with the waste package
- Long-term chemical and radiation stability
- Use of an organic material was strongly discouraged because organic materials tend to decompose at elevated temperatures and in radiation environments creating a potential for harmful interaction with fill material after a waste package is breached.

Additional screening criteria used by Fish et al. (1982) included criteria for emplacement temperature limits, shrinkage and voids, material interactions, moisture release, and gas generation. The tests conducted by Fish et al. (1982) included temperature limit tests, fill process tests, prebreach disposal condition tests (including loss-on-ignition tests and tests to evaluate fuel cladding-stabilizer material interactions), and electrochemical tests. As a result of these tests and evaluations, 1% antimonial lead and zirconia were recommended to be used as the reference materials used in waste package designs calling for the use of stabilizers. Table 2 summarizes selected physical properties of these materials.

Table 1. Recommended Candidate Stabilizer Materials

Material	Material
Silica – amorphous	Sand
Silica – quartz	Graphite
Silica – quartz/bondate	Graphite/bondate
85% silica – quartz/15% bentonite	Air
Mullite	Helium
Mullite/bondate	Nitrogen
85% mullite/15% bentonite	1% antimonial lead
Zircon	Calcium lead
Zirconia	Commercial lead
Zirconia/bondate	Zinc alloy AG40A
85% zirconia/15% bentonite	Zinc alloy AC41A
Basalt	Zinc-copper-titanium alloy
Basalt/bondate	Commercial zinc
85% basalt/15% bentonite	Copper casting alloy 3A (high-lead tin bronze)
Granite	Copper casting alloy 8A (manganese bronze)
Shale	Copper casting alloy 13B (silicon brass)
Tuff	Commercial copper
Source: Wynhoff et al. (1982) Bondate is an organic-based chemical bonding agent for aggregates and fibers.	

Table 2. Physical Properties of Recommended Reference Stabilizer Materials

Material	Density (g/cm3)	Thermal Conductivity (W/m-K)
1% antimonial lead	11.27	33.47
Zirconia	5.68	1.45
Source: Wynhoff et al. (1982)		

2.2 Allied-General Nuclear Services

Anderson (1981) investigated the use of fill materials to be used to encapsulate used nuclear fuel within a canister during the dry storage. The purpose of the study was to determine if encapsulation of used nuclear fuel with a fill material was desirable, compare physical and economic characteristics of alternative fill materials, and to review appropriate means to seal the storage canisters if fill materials were used. Tables 3, 4, and 5 summarize the materials evaluated and selected physical properties.

Table 3. Selected Physical Properties of Granular Solid Fill Materials

Material		Solid Density (g/cm^3)	Solid-Gas Mixture Thermal Conductivity (W/m-K)	Solid Melting Temperature (°C)
Solid	Gas[a]			
Copper spheres	Air	8.97	0.68	1083
Aluminum spheres	Air	2.70	--	660
Graphite	Air	1.50	1.2	3700
Zinc spheres	Air	7.14	0.46	283
Steel spheres	Air	7.85	0.25	1426
Lead spheres	Air	11.3	--	327
Boron carbide	Air	2.52	--	2450
Uranium oxide powder	Helium	10.8	1.5	2750
Alumina	Air	4.00	0.67	2050
Sand	Air	1.52	0.26	--
Glass	Air	2.22	0.18	1200
Mortar	--	2.20	0.92	--
Rock or glass wool	Air	0.16	0.050	--

Source: Anderson (1981)
a. The gases listed fill the interstices of the solid fill material.

Table 4. Selected Physical Properties of Liquid Fill Materials

Material	Density (g/cm^3)	Boiling Temperature (°C)	Pressure at Boiling (psia)	Thermal Conductivity (W/m-K)
Water	0.956 0.786	100 260	14.7 680.8	0.67 0.61
Ethylene glycol and water	1.013 0.963	100 177	13.8 103.0	0.40 0.36
Dowtherm	0.860 0.739	258 380	14.7 119.0	0.10 0.084
Silicone	0.900 0.744	100 300	0.077 20.9	0.12 0.071

Source: Anderson (1981)
Dowtherm is a heat transfer fluid.

Table 5. Selected Physical Properties of Gaseous Fill Materials

Material	Density (g/cm^3)	Thermal Conductivity (W/m-K)
Helium	0.000164	0.18
Air	0.00120	0.034
Nitrogen	0.00120	0.033
Carbon dioxide	0.00184	0.025
Argon	0.00166	0.022
Source: Anderson (1981)		

Anderson (1981) noted several advantages of fill materials. For example, by selecting the proper fill material one might reduce corrosion of the fuel cladding, increase the thermal conductivity of the contained fuel assembly, and reduce criticality considerations by lowering the effective neutron multiplication factor (k_{eff}) value of the used nuclear fuel container. The main disadvantage to the use of fill materials that was noted was economic. A second disadvantage that was noted was feasibility. Another potential disadvantage of using fill materials noted by Anderson (1981) involves the increased difficulty of retrieving the used nuclear fuel if retrieval is necessary at a later date. If the used nuclear fuel has been stabilized in a solid matrix (for example, by melting a metal, pouring it in a canister containing used nuclear fuel, and allowing the package to solidify), the removal of the used nuclear fuel could be quite difficult (Anderson 1981). In addition, the fill material could be slightly contaminated resulting in the generation of radioactive waste or additional process steps to decontaminate the fill material (Anderson 1981).

For the dry storage of spent fuel, Anderson (1981) found that air would be the best fill material. The use of fill materials other than air for dry storage of used nuclear fuel could be justified only if a specific end result, e.g., containment or criticality control, was deemed very important.

2.3 Canadian Nuclear Fuel Waste Management Program

The Canadian Nuclear Fuel Waste Management Program investigated alternative fill materials to be placed inside two types of waste containers: a thin-walled particulate-packed container and a structurally supported particulate-packed container. The purpose of the fill material was to provide structural support for the container against the hydrostatic pressure that could exist in a flooded, 1000-m deep disposal vault.

Shelson (1983) established a set of initial criteria for selecting particulates for future study and experiments. These criteria included necessary properties and desirable properties. Necessary properties were further grouped into criteria related to mechanical strength and criteria related to stability. Table 6 lists these criteria.

Table 6. Necessary and Desirable Criteria for Fill Material

Necessary Criteria	Desirable Criteria
Mechanical Strength High strength to breakdown (>20 MPa) High bulk modulus (>200 MPa) High Young's modulus (>200 MPa) Stability Radiation stability Chemical stability Not Reactive with titanium or heavy metals No interference with welding of shells Thermal stability (>1500 °C) Low water absorptivity (low swelling) No change over container life (300-500 years)	High heat transfer coefficient Low thermal expansion coefficient Low dust content Impede radionuclide migration Attenuate radiation from fuel bundles Low specific gravity
Source: Shelson (1983)	

From initial studies (Shelson 1983), twelve candidate particulate materials were selected for study (Teper 1987). These materials were:

- Sand
- Fine glass beads (0.002-0.3 mm)
- Coarse glass beads (0.8-1.2 mm)
- Steel shot (0.6-1.0 mm)
- Aluminum oxide powder
- Crushed bauxite grains
- Sintered bauxite
- Interprop[a]
- Ceramic zirconia
- Rutile-Zircon-Garnet mixture
- Zircon
- Rutile

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[a] Interprop is a ceramic proppant composed of 35-65% mullite (aluminum silicate) and 35-65% corundum (aluminum oxide). A proppant is a material that will keep an induced hydraulic fracture open.

The criteria used by Teper (1987) to select the fill material to be used in the container included:

- Fill all voids without clogging
- Be small enough to flow between the fuel bundle elements (less than 1.2 mm diameter) but the grains should be heavy enough to avoid becoming airborne during vibratory compaction
- Have sufficient strength to withstand a pressure of 20 MPa
- Have adequate stiffness to prevent large plastic deformations of the container shell
- Have low dust content to minimize airborne particles
- Should not adhere to the container wall, to simplify welding of top lid
- Have small creep deformations over the 500-year container life
- Have sufficiently high bulk modulus under external pressure

The particulates underwent vibratory compaction tests, compression tests, and creep tests. The details of the tests and their results are discussed in Teper (1987). Based on the results of the tests, three fill materials were considered 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, and were therefore selected as the fill material for the packed particulate and structurally supported containers (Johnson et al. 1994). The use of glass beads as a fill material was abandoned because glass beads could not provide assurance that the container would not collapse due to anticipated hydraulic pressures in the vault and was replaced with a carbon steel inner vessel to provide mechanical strength to the used nuclear fuel container (NWMO 2005).

The Canadian Nuclear Fuel Waste Management Program also investigated a metal matrix container, where cast metal surrounded the fuel bundles and forms a layer between the outer bundles and the shell of the container. Johnson et al. (1994) lists the following requirements for a candidate casting metal or alloy:

- The cast matrix should be free of major defects such as shrinkage voids
- During casting, the molten metal should neither chemically react with the corrosion-resistant shell nor otherwise reduce the thickness of the corrosion barrier.
- Interactions with the used nuclear fuel cladding should be minimal to ensure that the fuel elements are not damaged.
- Following solidification of the cast matrix, chemical stability between the matrix and the container shell should persist.
- The casting process should be conducted at as low a temperature as possible in order to reduce the preheating requirements of the container and its contents, decreasing the possibility of promoting thermal stress defects in the used nuclear fuel cladding material, and shorten the solidification period, during which chemical interactions between the matrix and the used nuclear fuel cladding material and/or the container shell are more likely.

Lead, zinc, and aluminum, and lead-antimony, aluminum-silicon, and aluminum-copper alloys were studied as candidate casting materials, and lead or zinc were recommended as the preferred casting materials. Table 7 summarizes some selected physical properties of lead and zinc. Subsequent research and development activities focused on lead. Four half-scale models, denoted MM1, MM2, MM3, and MM4, were cast and structural performance tests conducted. Testing and analysis showed that a metal matrix container was a viable option.

Table 7. Selected Material Properties for Lead and Zinc

Material	Density (g/cm^3)	Thermal Conductivity (W/m-K)	Melting Point (°C)
Lead	11.35	33.0	327.5
Zinc	7.10	112.2	419.58

2.4 U.S. Department of Energy

The U.S. Department of Energy has studied the use of depleted uranium oxide particulates as a fill material in used nuclear fuel waste packages (Forsberg 2000), and the use of depleted uranium silicate glass beads as a fill material in used nuclear fuel waste packages, storage containers, and transportation casks (Forsberg et al. 1995, 1996; Pope et al. 1996a, 1996b). In terms of the long-term performance of a geologic repository, the use of either depleted uranium oxide particulates or depleted uranium silicate glass beads has two advantages. First, it will retard the release of radionuclides from the waste package by creating a chemically reducing environment that slows the degradation of the uranium oxide contained in the used nuclear fuel, and by reducing ground water flow through the waste package (Forsberg 2000). In addition, the use of depleted uranium as a fill material minimizes the potential for a long-term criticality by isotopic dilution of U-233 and U-235 (Forsberg 2000).

In terms of storage and transportation, the use of depleted uranium silicate glass beads could have several benefits (Forsberg et al. 1995):

- The amount of gamma shielding material in the walls of the storage casks and transportation casks may be reduced.
- The neutron shielding materials in the walls of the storage casks and transportation casks may be reduced.
- The need to include burnup credit for criticality control may be eliminated.

Pope et al. (1996a, 1996b) acknowledges that there significant uncertainties associated with using depleted uranium silicate glass beads as a fill material, and that additional studies are necessary. The studies recommended by Pope et al. (1996a, 1996b) included:

- Developing and demonstrating the ability to produce depleted uranium silicate glass.
- Performing leaching tests on the depleted uranium silicate glass.
- Defining a preferred method for loading the depleted uranium silicate glass into a storage or transportation cask after they have been loaded with used nuclear fuel assemblies.

- Performing design alternative studies and defining costs and benefits of the various alternatives, including assessments of storage canister, transportation cask, storage cask, and waste package alternatives.
- Assessing trade-offs for and defining systems and interfaces for applying the concept of using depleted uranium silicate glass as a fill material to the waste management system.

2.5 Belgium

Belgium incorporated sand as a fill material in their used nuclear fuel canister design (Bennett and Gens 2008, ONDRAF/NIRAS 2001). The sand is a dry, halide-free rolled sand which fills the voids in the canister after being vibrated (ONDRAF/NIRAS 2001). As noted in ONDRAF/NIRAS (2001), the sand has a number of functions:

- The walls of the canister can be made thinner as the sand provides resistance to crushing
- The sand stabilizes the used nuclear fuel assembly in a centered position and so reduces criticality risks by mechanical convergence
- The sand limits the moderator density should water penetrate the canister
- The sand limits the void space which is a general requirement for waste intended for deep disposal.

After the canister has been filled with sand it is purged with a dry inert gas to minimize the risks of corrosive agents such as nitric acid being produced by radiologically induced reaction with humid air (ONDRAF/NIRAS 2001). The use of glass frit to fill the annulus between high level radioactive waste canisters and their overpacks is also being evaluated (ONDRAF/NIRAS 2001).

2.6 Spain

Puig et al. (2008a, 2008b, 2009) evaluated alternative fill materials that could be placed inside a used nuclear fuel canister that would be disposed of in a geological repository. The primary purpose of the fill material was to avoid the possibility of a criticality event once the canister was breached by corrosion and was flooded by ground water. Five groups of requirements for these fill materials were developed. These included requirements for criticality, general requirements to fulfill, general requirements to avoid, performance improvement requirements, and other interesting requirements. These requirements are listed in Table 8. Eight materials were evaluated:

- Cast iron or steel
- Borosilicate glass
- Spinel
- Depleted uranium
- Dehydrated zeolites
- Hematite
- Phosphates
- Olivine

Based on the evaluations of the materials against the requirements, four materials were found to be promising for use as a fill material: cast iron or steel, borosilicate glass, spinel, and depleted uranium.

Table 8. Requirements for Fill Materials (Spain)

Criticality Requirements Fill 60% of the canister inner free volume Significant neutron absorption capability Minimize neutron moderation Radiation resistance Thermal stability Chemical stability
General Requirements to Fulfill Thermodynamic equilibrium with conditions and materials in repository Homogeneous batches Good rheological properties to ensure proper filling Ability to be placed in canister without damaging fuel assemblies Does not affect fabrication, encapsulation, or other processes (i.e., welding of canister lid) Possible to disassemble canister Allow retrievability if needed
General Requirements to Avoid Limited availability of material Potential to increase corrosion of the canister, fuel cladding, or fuel itself. Increase the potential for radionuclide transport through bentonite barrier or chemically alter the barrier’s properties Retain significant amounts of air that could lead to formation of nitric acid through radiolysis and contribute to stress corrosion cracking
Performance Improvement Requirements High mechanical strength to contribute to canister structural integrity Sorption capability for key radionuclides
Other Interesting Properties Well-documented long-term durability Low material density to reduce additional weight of canister Low overall cost of material (raw materials, processing, and fabrication) Good intrinsic radiation shielding properties Material that allows a relatively simple process, including the necessary facilities and equipment
Source: Puig et al. (2008a)

2.7 Sweden

Oversby and Werme (1995) evaluated alternative fill materials that could be placed inside a copper and steel used nuclear fuel canister that would be placed inside a geological repository.

As with the fill materials analyzed by Puig et al. (2008a, 2008b, 2009), the primary purpose of the fill material was to avoid the possibility of a criticality event once the canister was breached by corrosion and was flooded by ground water. Design requirements were developed for the canister fill material and divided into three classes: essential requirements, desirable features, and undesirable features. These requirements are listed in Table 9. Eleven materials were evaluated:

- Glass beads
- Lead shot
- Copper spheres
- Sand
- Olivine
- Hematite
- Magnetite
- Crushed rock
- Bentonite
- Other clays
- Concrete

Based on the evaluations of the materials against the design requirements, three materials were found to be candidates for further evaluation as fill materials: glass beads, copper spheres, and magnetite. Because of the complexity of the filling process, canister designs without fill material were evaluated (Werme and Eriksson 1995) and current canister designs do not include a fill material (SKB 2010).

Sweden has also investigated a steel canister with lead fill, a copper canister with lead fill, and a titanium canister with concrete fill (SKB 1992). The titanium canister with concrete fill was used for very deep hole disposal, not for disposal in a geologic repository. Emplacing the lead in a steel or copper canister involved pre-heating the canister in an induction furnace to 380 °C for 6 hours, adding molten lead which was then allowed to solidify slowly from the bottom up to avoid voids, and cooling the canister for 12 hours to 60 °C. The entire time to pre-heat, fill, and cool a canister was estimated to be 24 hours (SKB 1992).

Table 9. Requirements for Fill Materials (Sweden)

Essential Requirements The filling material must be capable of being placed into the canister in a manner that does not damage the fuel and that results in a residual void volume of less than 40% of the void volume in the absence of the filling material. The filling material must have a solubility of less than 100 milligrams per liter at 50 °C in pure water and in waters of the expected repository environment. The filling material shall not compact by more than 10% of its original volume under its own weight or as the result of shipping, handling, or emplacing the canister in storage or disposal sites.
Desirable Requirements Material is in thermodynamic equilibrium with the disposal system, thus ensuring chemical compatibility. Material has homogeneous properties within a batch and between batches, which makes quality control and performance modeling more secure. Material possesses well documented long-term durability, which ensures that predictions concerning the condition of the material through time will be reliable. Material has good rheological properties for emplacement into the canister, which ensures that the operations in the encapsulation facility will not be unduly difficult. Material contains a burnable poison to absorb neutrons, which will enhance the criticality control of the filling material even if the void volume exceeds 40%. Material has the potential to sorb radionuclides from aqueous solutions, thus lowering the release of radioactive materials from the waste package. Material has the potential to suppress generation of hydrogen, which helps protect the bentonite buffer material from disruption due to passage of gas bubbles through the bentonite. Material has low cost. Material has low density, so performs its space-filling function with minimal addition of weight to the canister system.
Undesirable Requirements Limited availability of the material. Potential for the material to enhance corrosion of the canister, the fuel cladding, or the fuel. Material generates gas when it alters. Material contains water, which diminishes the effectiveness of the material to prevent moderation of the neutron energies. Material has a high affinity for absorbing air on its surface, which is undesirable because the nitrogen in air can be converted to nitric acid in the presence of water and radiation.
Source: Oversby and Werme (1995)

2.8 Yucca Mountain Project

Wallin et al. (1994) evaluated alternative fill materials that could be placed inside a waste package which in turn would be emplaced inside a geologic repository located at Yucca Mountain, Nevada. The objectives of adding the fill materials included (Wallin et al. 1994):

- Criticality control: moderator displacement by means of minimization of waste package internal void space, to minimize the amount of water which could enter the waste package in the event of repository flooding and a breach of the waste package containment barriers
- Chemical buffering for radionuclides in the event of water intrusion into the waste package upon breach of the containment barriers
- Cathodic protection by virtue of having highest electrochemical activity, in the event of water intrusion into the waste package upon breach of the containment barriers
- Function as mechanical packing to inhibit movement (collapse) of other materials internal to the waste package (fuel rods, fuel pellets, and/or basket materials)
- Improve thermal conductance, which would improve heat transfer and decrease fuel rod temperatures

Seven materials were evaluated:

- Tin (emplaced molten)
- Lead (emplaced molten)
- Zinc (emplaced molten)
- Zinc alloy (emplaced molten)
- Magnetite
- Iron shot
- Borosilicate glass beads

Iron shot was chosen as the first fill material to be experimentally investigated. Characteristics of iron shot that led to this choice included: 1) relative ease of placement (near-spherical shot “flows” readily), 2) commercial availability in a variety of graded sizes, 3) cost (inexpensive), 4) iron is a plentiful natural resource, 5) iron is a reactive anodic material providing protection to the fuel cladding and to Stainless Steel 316 components, and 6) iron would inhibit radionuclide release (Wallin et al. 1994).

Cogar (1996a and 1996b) contain the plans and technical guidelines used to conduct experiments conducted on steel shot, which was chosen over iron shot for the experiments because it was more readily available. These experiments involved:

- Fabricating two dummy fuel assemblies, a 15×15 B&W Mark-B pressurized water reactor assembly and a 17×17 B&W Mark-BW pressurized water reactor assembly.
- Fabricating a simulated spent nuclear fuel basket test fixture from Lexan. The dimensions of the test fixture were 8.81 × 8.81 × 180 inches. The test fixture had two vibrators attached.
- Using two grades of shot: SAE Shot Size S230 and SAE Shot Size S330. The S230 shot had a nominal diameter of 0.7 mm and the S330 shot had a nominal diameter of 1.0 mm.
- As-poured versus vibrated fill tests.

Cogar (1996c) conducted bulk density tests, fill placement tests, eight fill tests, angle of repose tests, and thermal conductivity tests. The eight fill tests conducted involved combinations of shot size (S230 and S330), assembly (15×15 and 17×17), and as poured versus vibrated conditions. Cogar (1996c) contains the detailed results of experiments. Table 10 summarizes these results.

Table 10. Summary of Experiments Involving Steel Shot

Material	Density (g/cm^3)	Thermal Conductivity (W/m-K)
SAE Shot Size S230	4.490-4.538 (as-poured) 4.568-4.653 (vibrated)	0.379-0.658
SAE Shot Size S330	4.353-4.397 (as-poured) 4.441-4.483 (vibrated)	0.325-0.591
Source: Cogar (1996c)		

Arthur (2000), Montierth (2000), and Radulescu (2000) also evaluated the use of aluminum shot containing gadolinium phosphate as a fill material in waste packages containing Shippingport light water breeder reactor thorium-uranium oxide seed assemblies. The results show that the Shippingport used nuclear fuel would not form critical configurations for any credible degradation scenarios when 1 weight percent gadolinium is added to the aluminum shot-gadolinium phosphate fill material. Similar analyses were performed for Enrico Fermi fast reactor used nuclear fuel using iron shot containing gadolinium phosphate as a fill material (Mobasheran 1999, Moscalu et al. 2000).

3. FILLING OF CANISTERS

The introduction of a fill material into a dry storage canister containing used nuclear fuel assemblies would be a significant departure from established industry practice for dry storage and planned subsequent transportation of used nuclear fuel. Consequently, any initiative to use such an approach would have to surmount a high hurdle of justification including consideration of alternatives such as repackaging the used nuclear fuel into another canister. Such justification could include:

- Use of a fill material was determined to be the best alternative for remediating a known defect in a canister or canister contents in order to provide reasonable assurance of continued protection of public safety and to ensure continued compliance with regulatory requirements.
- Use of a fill material was determined to be the best alternative for eliminating uncertainties regarding the integrity of fuel cladding, fuel structures, or canister internal structures or safety-related components to provide reasonable confidence in storage or transportation safety performance and assurance of compliance with regulatory requirements.

It is unlikely that fill materials could be introduced into dry storage canisters in an operating nuclear power plant’s used nuclear fuel storage pool. The reasons include issues regarding the compatibility of fill materials with the chemistry of the fuel pool water and the added operational complexity of adding fill materials. As a consequence, any activity to introduce fill materials to dry storage canisters would need to be conducted in a facility that would have the necessary health protection systems for workers and the public and systems to prevent releases of radioactive materials to the environment. It is beyond the scope of this report to provide a concept for such a facility. However, Carlsen and Brady Raap (2012) discusses various dry transfer systems for used nuclear fuel that could be applicable for use in introducing fill materials into dry storage canisters.

The objectives for introducing fill material into a canister could be several including:

1. To structurally stabilize (hold in place) the canister’s contents and geometry by filling in all of the available free space in and around the nuclear fuel assemblies and in and around the structures of the fuel assembly basket. This would protect the used nuclear fuel cladding from damage and preserve the geometric orientation of nuclear fuel and other materials and structures in order to provide assurance that a nuclear criticality could not occur.
2. To provide a medium that would exclude the potential for a significant amount of water moderator to intrude in and around the fuel assemblies thereby assuring a nuclear criticality could not occur.
3. To provide a medium that contains neutron absorber materials to enhance assurance that a nuclear criticality could not occur

4. To provide a barrier that impedes the release of radioactive material from used nuclear fuel assemblies to the environment.
5. To provide radiation shielding to reduce the radiation dose rate external to the canister.

Possible approaches for introducing a fill material that fills the free space in a dry storage canister containing used nuclear fuel include:

- Adding fill material to a canister containing used nuclear fuel before the closure lid is first installed.
- Using canisters that have access ports that are designed to be removed at a future date to provide openings for adding fill material.
- Unsealing and re-opening the ports that were originally used to drain, vent, dry, and backfill the canister with inert gas to provide openings through which fill material could be added.
- Unsealing and removing the canister lid to add fill material.
- Cutting access ports through the canister lid to provide openings for adding fill material.
- Cutting access ports through the side of a canister to provide openings for adding fill material.

The time when fill material might be added to a canister could be as early as when the canister is first loaded with used nuclear fuel or it could be 100 to 300 years in the future when the canister is being prepared for shipment following extended storage. Fill material might also be added to a canister at any time available information indicates that the integrity of fuel cladding, fuel structures, or canister internal structures or safety-related components has (or may have) degraded in a manner that compromises storage or transportation safety performance. This would include canisters with detected defects or when research results or other information suggest there are likely safety related defects in a particular canister design or design feature or a category of used nuclear fuel contained in a canister.

Fill material might also be added to a canister immediately prior to transportation whenever the integrity of fuel cladding, fuel structures, or canister internal structures or safety-related components cannot be verified sufficiently to provide reasonable assurance of transportation safety and compliance with regulatory requirements for transportation. This would include canisters containing used nuclear fuel following extended storage and canisters containing high burnup used nuclear fuel.

Possible fill materials can be grouped into 3 categories:

1. Liquids, including molten metals, waxes, resins, and grout, that would flow into and fill a canister before undergoing physical change to become a solid.
2. Particulates, including sand, borosilicate glass beads, and metal shot, that would be introduced to canisters to fill available spaces through cascading gravity flow (Wallin 1996).

3. Foams that would be introduced into selected locations in a canister and then would expand and infuse through available internal openings and gaps to fill open spaces.

These categories determine the process that would need to be used to introduce fill materials into a canister.

Processes for filling canisters would be determined by the type of fill material that was used and the approach taken to transfer the material into the canister. Table 11 summarizes the conceptual filling processes that could be employed for each of the different kinds of fill materials and for the different approaches to filling a canister that are described above. Figure 1 provides a conceptual illustration of the process for filling a canister that has its lid removed. The processes described assume that the canister is filled in a facility designed and dedicated for that purpose. The concepts described are unproven. It would be necessary to design and conduct a program that would include tests that demonstrated the feasibility of a fill material concept before any decision was made to use a fill material to stabilize the used nuclear fuel contents of a storage canister.

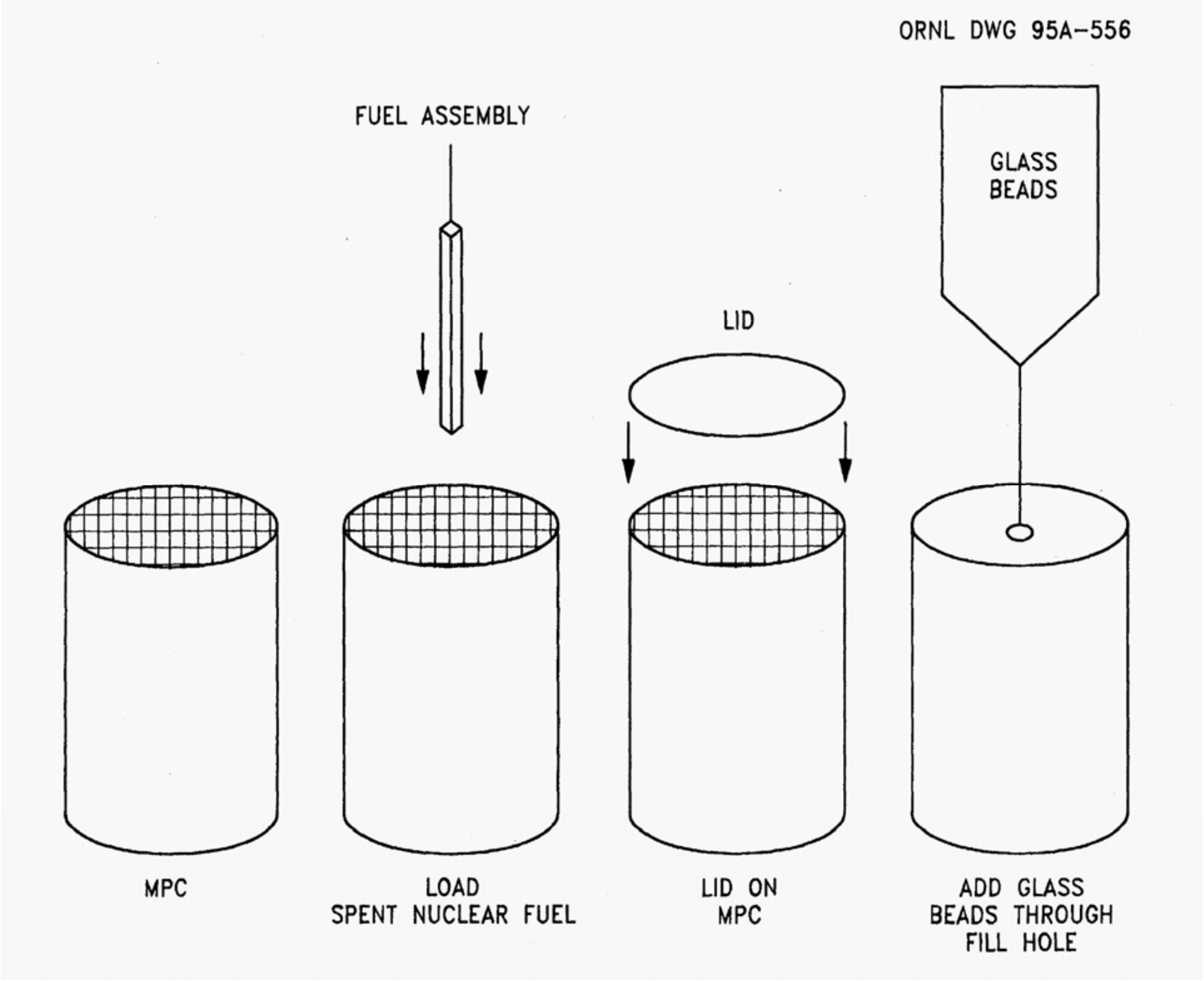

Figure 1. Used Nuclear Fuel Canister Loading Sequence

(Source: Forsberg et al. 1995)

Table 11. Conceptual Approaches for Introducing Fill Material into Dry Storage Canisters Containing Used Nuclear Fuel

Fill Material Type	Approach for Introducing Fill Materials				
	Unseal and remove lid from loaded canister	Unseal and reopen canister drain and vent ports	Canister with ports for adding fill material	Cut openings into canister lid	Cut openings into canister side wall
Liquid	The canister lid's seal weld is cut and lid is removed. Liquid fill is introduced into the canister via a down-tube (vent tube) into the bottom of the open canister and is allowed to flow up to fill the canister before solidifying. The canister's lid is reinstalled and re-welded. An additional external canister may be used if the canister is defective.	The canister's drain and vent ports are unsealed and opened. Liquid fill is introduced into the canister via the vent tube into the bottom of the open canister and is allowed to flow up to fill the canister before solidifying. The canister's drain and vent ports are closed and resealed. An additional external canister may be used if the canister is defective.	Liquid fill is introduced via the inlet port into the canister and is allowed to flow up to fill the canister before solidifying. The canister's ports are re-sealed. An additional external canister may be used if the canister is defective.	Liquid fill is introduced via the cut opening into the canister and is allowed to flow up to fill the canister before solidifying. The openings are re-sealed. An additional external canister may be used if the canister is defective.	Liquid fill is introduced via the cut opening into the canister and is allowed to flow up to fill the canister before solidifying. The openings are re-sealed. An additional external canister may be used if the canister is defective.

Table 11. (contd)

Fill Material Type	Approach for Introducing Fill Materials				
	Unseal and remove lid from loaded canister	Unseal and reopen canister drain and vent ports	Canister with ports for adding fill material	Cut openings into canister lid	Cut openings into canister side wall
Particles	The canister lid's seal weld is cut and lid is removed. Particulate fill is introduced into the top of the canister and is allowed to fill the canister by cascading gravity flow. Vibration may be used to speed up the flow of particulates and to enhance compaction of the particle bed. The canister's lid is reinstalled and re-welded. An additional external canister may be used if the canister is defective.	This approach will not work with particles. There are only two relatively small diameter ports in a canister lid and, even with vibration, the particles will not flow to fill the remaining sections of the canister once a section directly under the ports is filled in.	The canister fill ports are removed. Particulate fill is introduced into the top of the canister through the ports and is allowed to fill the canister by cascading gravity flow. Vibration may be used to speed up the flow of particulates and to enhance compaction of the particle bed. The canister's lid is reinstalled and re-welded. An additional external canister may be used if the canister is defective.	Particulate fill is introduced into the top of the canister through the cut openings and is allowed to fill the canister by cascading gravity flow. Vibration may be used to speed up the flow of particulates and to enhance compaction of the particle bed. The openings are closed and sealed. An additional external canister may be used if the canister is defective.	This approach will not work with particles. Particles will not flow up channels and openings to fill spaces and therefore will not move from the sides of a canister toward the center.

Table 11. (contd)

Fill Material Type	Approach for Introducing Fill Materials				
	Unseal and remove lid from loaded canister	Unseal and reopen canister drain and vent ports	Canister with ports for adding fill material	Cut openings into canister lid	Cut openings into canister side wall
Foam	The canister lid's seal weld is cut and lid is removed. Liquid that will expand to become foam is introduced via a down-tube into the bottom of the canister and is allowed to fill the canister and all available spaces by expanding upward as a medium-viscosity high-density foam. The canister's lid is reinstalled and re-welded. An additional external canister may be used if the canister is defective.	The canister drain and vent ports are unsealed and opened. Liquid that will expand to become foam is introduced via the vent tube into the bottom of the canister and is allowed to fill the canister and all available spaces by expanding upward as a medium-viscosity high-density foam. The canister's drain and vent ports are closed and sealed. An additional external canister may be used if the canister is defective.	The canister fill ports are unsealed and opened. Liquid that will expand to become foam is introduced via a down-tube into the bottom of the canister and is allowed to fill the canister and all available spaces by expanding upward as a medium-viscosity high-density foam. The canister's fill ports are closed and sealed. An additional external canister may be used if the canister is defective.	Liquid that will expand to become foam is introduced via a down-tube extending from the cut opening in the canister lid into the bottom of the canister. The foam is allowed to fill the canister and all available spaces by expanding upward as a medium-viscosity high-density foam. The openings in the canister lid are closed and sealed. An additional external canister may be used if the canister is defective.	Liquid that will expand to become foam is introduced through a side hole that is cut into the canister wall and flows down into the inside wall into the bottom of the canister. The foam then fills the canister and all available spaces by expanding upward as a medium-viscosity high-density foam. The openings in the canister lid are closed and sealed. An additional external canister may be used if the canister is defective.

4. REQUIREMENTS FOR FILL MATERIALS

The previous work discussed in Chapter 2 used various criteria for choosing a fill material. Many of these criteria were specific to used nuclear fuel in waste packages for disposal and thus were related to post-closure performance of a geologic repository. Although many of the criteria could be applicable, they were not selected with consideration of storage or transportation of used nuclear fuel.

This section discusses potential criteria that could be considered when selecting a fill material for a used fuel canister. In contrast to many of the criteria presented in Section 2, these criteria are based on storage and transportation and do not consider post-closure performance. Each requirement is presented in Table 12 along with a summary of the critical elements of that requirement.

Table 12. Potential Requirements for Fill Materials

Evaluation Criteria for Candidate Canister Fill Material	Elements
Criticality Avoidance	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
Heat Transfer or Thermodynamic Properties	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
Homogeneity and Rheological Properties	Homogeneous batches Good rheological properties to ensure proper filling Ability to be placed in the canister without damaging fuel assemblies
Retrievability	Allows for safe retrieval of used nuclear fuel from a canister without need to resort to time consuming or costly measures and without further compromise of the integrity of used nuclear fuel assemblies
Material Availability and Cost	Low cost Material available in required purity
Weight and Radiation Shielding	Fill material doesn’t add significantly to the weight of the container/cask system Good radiation shielding properties
Operational Considerations	Easy to emplace Fill material does not adversely react to normal conditions of transport or hypothetical accident conditions

4.1 Criticality Avoidance

One of the most important criterion for fill material to meet is criticality avoidance, and one potential outcome of the use of certain fill materials could be the ability to eliminate the need to consider burnup credit in the design of the storage container or transportation cask. The standard for criticality is maintaining the effective neutron multiplication factor (k_{eff}) at 0.95 or below. There are several ways to ensure a k_{eff} of 0.95, such as use of a fill material with significant neutron adsorption capability, by moderator exclusion, or by dilution of fissile radionuclides. The material should also be chosen so that it does not provide neutron moderation to ensure that a subcritical k_{eff} is maintained. In addition, based on analyses cited in Oversby and Werme (1995), the candidate fill material should occupy at least 60% of the original void space in the canister and the material should not compact under its own weight or as the result of shipping or handling by more than 10%.

The need for criticality avoidance as a criterion for fill material may be reduced based on the draft burnup credit guidance contained in NRC (2012), which extends the current major actinide burnup credit (NRC 2002) to include minor actinides and fission products. As discussed in Marshall and Wagner (2012), identification of credible fuel configurations may also reduce the need for a criticality avoidance criterion for fill material.

4.2 Heat Transfer and Thermodynamic Properties

The fill material should also not have a detrimental impact on heat transfer. The temperature of the cladding should be demonstrated to not exceed 400 °C which is regarded as protective of used nuclear fuel cladding. Depending on the fill material and how compacted it is, the radiative and convective heat transfer mechanisms may be virtually eliminated. It is possible that this could be offset by the increase in the thermal conductivity of the fill material. However, each material would need to be evaluated for thermal properties and their effect on the thermal performance of the used nuclear fuel in the canister.

The fill material should also be chemically compatible with the fuel cladding, fuel, neutron poisons, fuel baskets, and other structural materials within canister. Also, the fill material should not undergo adverse interactions with the residual moisture within the canister such as rusting of steel shot, catalysis or radiolytic decomposition of water, or galvanic interactions with cladding or neutron shielding materials. Also, the fill material should be stable within the canister and not degrade due to heat, radiation, or by chemical reaction. The fill material should also not degrade and produce hydrogen or other gases.

Because the duration of long-term storage is also uncertain, the fill material should be relatively unaffected by age or the importance of the fill material properties demonstrated to be less important as the fuel ages. Aging of the fill material would not be important if the fill material was emplaced a short time before transportation and removed soon thereafter.

4.3 Homogeneity and Rheological Properties

Good rheological properties would allow easier flow or flow readily into the canister without agitation and would allow greater assurance of meeting the fill criteria. Another consideration is that the fill material should have homogeneous properties throughout the canister fill. The fill material should also have the ability of being placed in the canister without damaging the fuel assemblies.

4.4 Retrievability

The fill material should allow for the safe retrieval of used nuclear fuel from a canister without the need the resort to timely or costly measures and without further compromise of the integrity of the used nuclear fuel assemblies.

4.5 Material Availability and Cost

The cost and availability of the fill material should be considered when choosing a fill material. This would include the costs of the raw materials with the required purity, processing the materials, and the cost of emplacing the materials in the canister. The ease at which the fill material could be provided to multiple locations such as nuclear power plant sites should also be considered.

4.6 Weight and Radiation Shielding

First, the additional mass that the fill material provides should not result in the canister and cask weight exceeding or approaching weight restrictions for handling or transportation. However, it is possible that certain fill materials would result in a reduction in the need for shielding and as a result a reduction in the overall mass attributed to shielding. This would need to be verified with modeling. Second, the project fill material mass would need to be evaluated against the overall mass of the cask/canister system and its mass limits and any potential modifications to the canister design (wall thickness) to accommodate the additional mass. Third, the fill material may require a reduction in the number of assemblies that a canister would hold to accommodate the mass and volume occupied by the fill material and its desired properties.

4.7 Operational Considerations

The fill material should be easy to emplace in the canister and the fill material should also not interfere with the sealing of the canister, such as welding of the canister lid. The fill material should not adversely react to normal conditions of transport or hypothetical accident conditions.

5. POTENTIAL FILL MATERIALS

This section discusses potential fill materials. Potential fill materials were grouped into several categories such as molten materials, particulates and beads, resins, foams, and grout.

5.1 Molten Materials

Two types of molten materials were evaluated, molten metals and paraffin. As discussed in Chapter 2, molten metals have been evaluated as potential fill materials for waste packages in several studies but no studies were found that had evaluated paraffin as a fill material.

5.1.1 Molten Metals

The first instance identified of a molten material being proposed for use to stabilize used nuclear fuel during transportation was a patent granted in 1974 to Wurm and Heylen (1974). In this patent, fuel rods would be placed inside a can and the filling alloy would fill the space between the fuel rods and the can. These cans containing the alloy-encased fuel rods would then be placed inside a transportation cask and shipped to their destination, typically a reprocessing plant. The filling alloy performs several functions: 1) the filling alloy stiffens the structure of the fuel element so that the fuel rods cannot break during transportation, 2) if the rods were to break, no radioactive gas would escape, and 3) the filling alloy would conduct heat from the rods to the can. It is not known if used nuclear fuel has ever been transported using the filling alloy method outlined in Wurm and Heylen (1974).

The use of molten metals as a fill material has also been investigated by the Spent Fuel Stabilizer Materials Program and the Canadian Nuclear Fuel Waste Management Program. The Spent Fuel Stabilizer Materials Program recommended 1% antimonial lead as a reference material, while the Canadian Nuclear Fuel Waste Management Program focused research and development activities on lead.

The Yucca Mountain Project also evaluated the use of a tin, lead, zinc, and zinc alloy as fill materials inside a waste package as an alternative to an inert gas (Wallin et al. 1994). Tin was rejected by Wallin et al. (1994) as not being sufficiently plentiful. Lead was rejected because it is toxic, very heavy, and can cause embrittlement of other metal components. Unalloyed zinc was rejected because it was determined that the zinc will interact with the Zircaloy fuel cladding material. Zinc alloys such as AG40B could possibly be acceptable from that standpoint, as they would have a lower tendency to interact with the cladding.

In this evaluation, 5 representative molten metals were considered:

- Tin
- Lead
- Zinc
- Zinc alloy (AG40A and AG40B)
- Wood’s metal

These materials are representative of materials with relatively low melting points, less than approximately 420 °C. This temperature was chosen because it would limit the potential for gross rupture of the cladding and preserve the geometric configuration of the used nuclear fuel (NRC 2003). In addition, this temperature is well below the melting point of aluminum (660.37 °C), which is often contained in structural components of metal canisters and in neutron poisons. Table 13 lists representative properties of these materials.

Table 13. Representative Properties of Candidate Molten Metals

Material	Melting Point (°C)	Density (g/cm^3)	Thermal Conductivity (W/m-K)
Tin	232	7.29	63.2
Lead	327.5	11.35	33.0
Zinc	419.58	7.10	112.2
Zinc alloy (AG40A or AG40B)	381-387	6.60	113
Wood’s metal	70.0	9.58	18.0
Source: MatWeb (2012)			

One of the primary requirements for a fill material is weight. Based on current designs for used nuclear fuel storage systems, the free volume in a storage canister is in the range of about 4000 to 7000 liters. Assuming that a storage canister was completely filled with molten metal, the weight of a canister would be increased by 58,200 to 102,000 lbs. for zinc alloy, the lowest density material, and by 100,000 to 175,000 lbs. for lead, the highest density material. Current canisters weigh in the range of about 80,000 to 100,000 lbs., so adding a molten metal fill material would approximately double the weight of an existing canister. The addition of a molten metal fill material to a canister would result in the canister not meeting the current requirements of the canister’s 10 CFR 72 storage certificate of compliance and the 10 CFR 71 transportation certificate of compliance under which it would be shipped. The additional weight and changed contents would make it necessary to reanalyze the performance of the canister, and recertify the modified canister for continued storage. The changes would also be significant in regard to the design of the transportation cask and would require reanalysis, a probable redesign, and recertification. Therefore, for current canister designs, adding a molten metal fill material appears not to be feasible based on weight and other considerations. Future canisters and their associated transportation casks would need to be designed, possibly with lower capacities and thicker walls, to allow for the increase in weight due to the fill material. Unless the weight of the transportation cask could be reduced as a result of longer cooling times for the used nuclear fuel and possibly because of increased self-shielding by the canister, the decrease in the capacity of the canisters would be as much as 50 percent, which would double the number of canisters that would eventually have to be shipped.

5.1.2 Paraffin

No studies were found where paraffin wax had been investigated as a fill material for used nuclear fuel canisters. Paraffin wax is a mixture of pure alkanes with a chemical formula of C_nH_{2n+2}. It has a melting point between about 46 and 68 °C, has a density of about 0.9 g/cm^3, and burns readily if a fire retardant is not incorporated. Paraffin also has a relatively low thermal conductivity, 0.25 W/m-K. Because of its low melting point, paraffin wax could be melted for easy pouring and then hardened to insure complete covering of the used nuclear fuel in a canister. However, there are issues with the use of paraffin as a fill material. For example, paraffin is a hydrocarbon and, if neutron absorber materials are not incorporated, it is an effective neutron moderator. Thus, to make this material a viable fill material, a neutron absorber such as boron would need to be added to the paraffin before pouring. In addition, because of its flammability, a flame retardant would need to be added to the paraffin so that the paraffin material would not burn if released during a transportation accident. Also, because paraffin is a hydrocarbon, it would be subject to radiolytic decomposition that would progress over time. Consequently, except for used nuclear fuel that had been stored for long periods of time such that the source of ionizing radiation was significantly diminished, paraffin could not be used if it was to remain in a canister for a prolonged period of time. Future tests would need to be conducted to determine if paraffin would generate hydrogen or other gases during transportation, especially for used nuclear fuel that had relatively short cooling times. Interactions of paraffin with used nuclear fuel cladding and canister material would also need to be evaluated.

Another issue that would need to be resolved regarding the use of paraffin is its relatively low melting point and at what time in the future the decay heat from used nuclear fuel would be low enough such that the material would remain a solid during transportation. Alternatively, it would be necessary to determine whether a paraffin-containing transportation cask could be shipped when the paraffin was in a liquid state.

A key benefit of using paraffin would be its weight. The density of most paraffin waxes is slightly less than that of water: approximately 0.9 g/cm^3. Based on current designs for used nuclear fuel storage systems, the free volume in a storage canister is in the range of about 4000 to 7000 liters. Assuming that a storage canister was completely filled with paraffin, the weight of a canister would be increased by 7,900 to 13,900 lb. This is much less than other candidate fill materials. Nonetheless, this increase in the weight of the canister would result in the canister not meeting the requirements of its current 10 CFR 71 transportation certificate of compliance. It is assumed that because paraffin would undergo radiolytic decomposition, it could not be used to stabilize the contents of a canister that would continue to be used for storing used nuclear fuel. The additional weight and changed contents would make it necessary to reanalyze the performance of, and recertify, the modified canister and transportation cask. Therefore, for current canister designs, it is uncertain whether adding paraffin would be feasible based on weight considerations. Assuming that the weight of the transportation cask would be reduced as a result of longer cooling times for the used nuclear fuel that is stabilized by paraffin fill material, it is likely that future canisters and their associated transportation casks would have capacities comparable to present day systems. This would be the case even though the canister would weigh more because of a paraffin fill material. Thus, unlike canisters that would be filled with molten metal, there would not be an increase in the number of canisters that would need to be shipped.

5.2 Particulates and Beads

As discussed in Section 2, the use of particulates and beads has been extensively studied as a fill material for waste packages. It has also been studied on an extremely limited basis as a fill material for storage containers, and transportation casks. Table 14 lists particulates and beads that have been previously studied.

In experiments conducted to determine potential interactions between particulates and Zircaloy-4, Fish et al. (1982) found that mullite, graphite, basalt, zircon, zirconia, amorphous silica, and quartz formed a brittle interaction layer at the cladding-particulate interface. Fish et al. (1982) postulated that the interaction layers consisted of zirconium oxide. The interaction layers were thought to form due to the extraction of oxygen from the silicon oxide contained in these materials and the formation of zirconium oxide. Graphite also formed an interaction layer with Zircaloy-4 cladding which was likely zirconium carbide. The formation of these interaction layers has the potential to weaken the cladding. Other materials such as interprop and sand/bondate also contain silica and would also likely form the interaction layers observed by Fish et al. (1982).

Fish et al. (1982) also conducted loss-on-ignition tests of candidate particulates. Basalt and bentonite were found to have greater than 1 percent moisture release, which could contribute to corrosion and internal pressurization of a canister.

The density of solid lead is 11.35 g/cm^3. Assuming a packing fraction of 65% for lead spheres, the effective density of lead spheres would be about 7.4 g/cm^3. As with the molten metals discussed in Section 5.1.1, this would substantially increase the weight of the canister by 65,300 to 114,000 lb. and would result in an extremely heavy canister. Lead spheres could also potentially compact under their own weight and form voids within the canister.

Depending on how the canisters containing the used nuclear fuel were filled, generation of dust is likely to be an issue because this dust could contaminate the welds used to seal the canister. During packing experiments conducted for the Canadian Nuclear Fuel Waste Management Program, Teper (1987) found that aluminum oxide powder, sand, zircon, rutile, ceramic ziconia, and rutile-zircon-garnet generated excessive dust. In addition, ceramic zirconia had a tendency to form voids.

Depleted uranium oxide particulates or depleted uranium silicate beads have been evaluated by Puig et al. (2008a), Forsberg (2000), Forsberg et al. (1995, 1996), and Pope et al. (1996a, 1996b). Depleted uranium particulates as a fill material for waste packages could have several desirable qualities such as criticality control, radiation shielding, and slowing the release of radionuclides from the waste package. The density of depleted uranium oxide is 10.96 g/cm^3. Assuming a packing fraction of 65% for depleted uranium oxide particulates, the effective density of depleted uranium oxide particulates would be about 7.1 g/cm^3. As with the molten metals discussed in Section 5.1.1 and lead spheres, this would substantially increase the weight of the canister by 62,600 to 110,000 lb. and would result in an extremely heavy canister.

Depleted uranium silicate beads have a density of about 4.1 g/cm^3. Assuming a packing fraction of 65% for depleted uranium silicate glass beads, the effective density of depleted uranium silicate beads would be about 2.7 g/cm^3, which would increase the weight of a canister by 23,800 to 41,700 lb. This increase in weight would mean that filling existing canisters might not be feasible. However, if used nuclear fuel were cooled long enough, the added weight of the fill material might be offset by a reduction in the weight of the transportation cask. Nonetheless, because of the changed contents and added weight, it would be necessary to provide a new analysis of the performance of the canister for storage and transportation. Based only on weight, it is possible that filling future smaller (less capacity) canisters with the depleted-uranium silicate beads would be feasible, with a corresponding increase in the number of used nuclear fuel canisters. Filling canisters with depleted uranium silicate beads might also eliminate the need for burnup credit for these new canister-transportation cask systems. However, the same benefit could be realized by using boron-containing glass beads, i.e., borosilicate glass beads, which would have a poured density of about 1.9 g/cm^3 and consequently would not have as much of a weight penalty as would the depleted uranium silicate beads and would also not have the radiation protection issues associated with the use of depleted uranium.

Particulate materials such as magnetite, hematite, olivine, phosphates, and zeolites have been studied as waste package fill materials by Puig et al. (2008a, 2008b, 2009) and Oversby and Werme (1995). The properties of interest were oriented towards post-closure performance of a geological repository, such as the ability to sorb radionuclides and the ability to maintain reducing conditions in the near field around a waste package. In addition, based on the results of packing experiments involving aluminum oxide powder, sand, zircon, rutile, ceramic ziconia, and rutile-zircon-garnet, there is the potential that these materials could generate excessive dust.

Metal shot, such as aluminum, steel, and copper shot, and borosilicate glass beads have been suggested as a potential fill material in several of the studies discussed in Chapter 2. The density of emplaced shot would range from about 1.8 g/cm^3 for aluminum shot to about 5.8 g/cm^3 for copper shot, and would be about 1.9 g/cm^3 for glass beads. As with other materials of relatively high densities, this would increase the weight of existing canisters containing the used nuclear fuel and would result in the canisters not meeting the requirements of their 10 CFR 72 storage certificate of compliance and their 10 CFR 71 transportation certificate of compliance. Therefore, for current canister designs and certifications, adding metal shot or borosilicate glass beads would not be feasible solely based on weight considerations. Future canisters and their associated transportation casks would need to be designed, possibly with lower capacities, to allow for the increase in weight due to the metal shot or borosilicate glass beads. If the capacities of these canisters were less than that of current canisters the increase in the number of canisters would not be as large as for other fill materials with higher densities.

An additional issue associated with materials such as particulates and beads is the potential to compact during the normal conditions of transport. For example, during transportation of a prototype container from Toronto, Ontario, Canada to the Whiteshell Laboratories located in

Pinawa, Manitoba, Canada, the glass-bead particulate within the container appeared to have settled, causing a 14 mm gap between the top head of the container and the top of the particulate (Crosthwaite 1994).

and Beads Evaluated in Previous Studies

Purpose	Materials Studied	References
Waste package fill material, used nuclear fuel storage container fill material, transportation cask fill material	Sand Copper spheres Aluminum shot and spheres Zinc spheres Lead spheres Steel shot and spheres Iron shot Magnetite Rutile Amorphous silica Quartz Mullite Zircon Zirconia Basalt Graphite Sand/bondate Bentonite Glass beads and spheres Boron carbide powder Uranium oxide powder Alumina and alumina powder Depleted uranium oxide Depleted uranium silicate glass	Anderson (1981) Pope et al. (1996a, 1996b) Forsberg et al. (1995, 1996) Forsberg (2000) Wallin et al. (1994) Cogar (1996) Montierth (2000) Arthur (2000) Fish et al. (1982)
Waste package fill material	Steel shot Glass beads Spinel Depleted uranium oxide spheres Zeolites Hematite Phosphates Olivine	Puig et al. (2008a, 2008b, 2009)

Table 14. (contd)

Country	Purpose	Materials Studied	References
Canada	Waste package fill material	Sand Fine glass beads (0.002-0.3 mm) Coarse glass beads (0.8-1.2 mm) Steel shot (0.6-1.0 mm) Aluminum oxide powder Crushed bauxite grains Sintered bauxite Interprop Ceramic zirconia Rutile-Zircon-Garnet mixture Zircon Rutile	Teper (1987) Forsberg (1997)
Belgium	Waste package fill material	Sand	Bennett and Gens (2008) ONDRAF/NIRAS (2001)
Sweden	Waste package fill material	Glass beads Lead shot Copper spheres Sand Olivine Hematite Magnetite Crushed rock	Oversby and Werme (1995)
Interprop is a ceramic proppant composed of 35-65% mullite (aluminum silicate) and 35-65% corundum (aluminum oxide). A proppant is a material that will keep an induced hydraulic fracture open. Bondate is an organic-based chemical bonding agent for aggregates and fibers.			

5.3 Resins

No studies were found where liquid resins had been investigated as a fill material for used nuclear fuel canisters. Resins are potentially good candidates for a fill material due to their ability to be poured into a canister as a liquid and then solidify to provide for total coverage of the used nuclear fuel. The fact that resins are organic and are thus moderators of neutrons could be compensated for by adding neutron absorbing materials to the resin. There are other concerns, however, that must be addressed to allow resins to be a viable fill material. These include thermal conductivity, softening point, radiation stability, density, viscosity, and ignition point.

There are many types of resins, each with varying properties, so several are researched in the present paper. These resins include FF grade wood rosin, polyurethane resin, polystyrene resin, epoxy resin, unsaturated polystyrene resin, acrylic resin and silicone resin. The material properties for several resin types are summarized in Table 15.

The densities of these resins are all relatively the same, ranging from 1.00 to 2.00 g/cm^3. This equates to the addition of 8,800 to 30,800 lb. to the weight of the canister, depending on the void volume within the canister. These values are adjustable based on what curing agent is used. These curing agents can also greatly affect the other properties of the resin. Due to the high degree of fill expected when using these resins, existing canisters could not be filled with resin and shipped unless an analysis to demonstrate performance was done and approval was given by the U.S. Nuclear Regulatory Commission, and future canisters and their associated transportation casks would need to be designed with somewhat lower capacities unless the designs assumed longer cooling times before transportation and the resulting reduced weight of transportation casks offset the increased weight of the canisters.

The thermal conductivities of the resins are also all expected to be relatively the same, ranging from 0.10 to 1.00 W/m-K. This is relatively low, but with longer-cooled or low-heat used nuclear fuel it is not likely to be a concern. If necessary, it may be possible to add a material to the resin to help conduct the heat to the canister structure, such as a metal. There is very little data on thermal conductivities for resins, and so future tests would need to be conducted to establish this physical property.

Another similar characteristic shared by most resins are their resistance to radiation damage. The major damage to resins (and most polymers) from radiation is induced cross-linking or chain scission. Since the resins are cured, this damage would be reduced significantly. As stated in ATL (2001), most resins can withstand a radiation dose of 10^6 Gy. However, a radiation dose 10^7 to 10^8 Gy can produce damage. If the resin were poured inside the canister just before shipment of used nuclear fuel that had been in extended storage for 100 to 300 years, this potentially would probably not be an issue. Future tests would need to be conducted to verify this.

Pour viscosity is another property where most resins share a similar value. Most resins have a viscosity on the order of 1 Pa-s, about 1000 times more viscous than water and on the same order of viscosity as honey. Although this is relatively viscous, it should pose no real impedance to filling a canister other than allowing for an appropriate amount of time to fill the canister before the curing process can take place. However, future tests would have to be conducted to verify that resins could be poured into a canister without creating significant void spaces.

The major issues with resins lie in their chemical stability, both in terms of softening and ignition points. Although cured resins soften at higher temperatures than the uncured resins that are poured into the canister to begin with, these softening points can still be well below the 400 °C temperature that is regarded as protective of used nuclear fuel cladding. In this capacity, polyurethane, epoxy and silicone resins perform best with softening points of approximately 150 °C. Further research is needed to find curing agents that would be able to increase the softening point if 150 °C is not sufficiently high.

Although resins melt at low temperatures, their ignition points can exceed 400 °C. For example, if the right curing agents are used, polystyrene, polyester and silicone resins will not ignite until temperatures of 430, 500 and 760 °C, respectively. If the resin could be exposed to the atmosphere following a fire accident, it might be necessary to include ignition retardants in the resin formulation or to conduct tests to verify that the ignition point of the resin used as a fill material is not reached, especially during the hypothetical accident conditions specified in 10 CFR 71.73. Future tests would also be needed to determine if resins could generate hydrogen or other gases when they decompose.

Table 15. Material Properties of Resins

Resin	Density (g/cm^3)	Softening Point (°C)	Thermal Conductivity (W/m-K)	Viscosity (mPa-s)	Ignition Temperature (°C)
FF Wood Rosin	1.089	100–120	--	4000	--
Polyurethane	1.490	144	0.65	6000	N/A
Polystyrene	1.040	105	--	--	430
Epoxy	1.335	80–162	0.2	5000	390
Unsaturated Polyester	1.900	70–100	--	2000	500
Acrylic	1.160	108	--	1500	340
Silicone	1.000	7–138[a]	--	200	760

a. Flashpoint.
Note: The material properties are representative of the type of resin and the properties of specific resins may vary.

5.4 Foams

No studies were found where foams were investigated as a fill material for used nuclear fuel canisters. As with resins, there are many types of foams. Table 16 summarizes the material properties of several foams.

Foams could potentially insure an easy filling process with complete coverage and support of the used nuclear fuel as well as having a low density which would not increase the weight of the canister significantly. However, the ability to inject foam inside a used nuclear fuel canister without significant void spaces would need to be verified with future experiments.

Foams have several downfalls, but like paraffin, these downfalls may be compensated for by the addition of other materials or further research. Most foams are organic in nature, and so are excellent neutron moderators. This can be compensated for by adding a neutron absorbing material to the foam before injection, or by using inorganic foams instead. Foams may also provide moderator exclusion.

Organic foams can also burn readily at relatively low temperatures (approximately 400 °C). However, foams have been used in the design of Type B radioactive materials containers. For example, the TRUPACT-II container (Docket Number 71-9218) and TRUPACT-III container (Docket Number 71-9305) both contain polyurethane foam. Nonetheless, tests would be needed to determine if foams could ignite or decompose inside a canister, especially during hypothetical accident conditions. Future tests would also be needed to determine if foams could generate hydrogen or other gases when they decompose. In addition to chemical and radiation stability, foams must have desirable properties in the thermal stability, rheology, density, and strength criteria. Based on the ignition temperatures in Table 16, organic foams my not be the best choice for a fill material due to low ignition temperatures, and metal or ceramic foams may have desirable properties in the areas of structural integrity, increased thermal conductivity, lack of neutron moderation, and their high temperature performance.

The density of foams can vary widely based on what material they are, as well as whether they have open- or closed-cell structures. An open-cell foam is one where the gas is not trapped within the foam structure, much like that of a common sponge, while a closed-cell foam is a solid material with gas bubbles are trapped inside. Thus, the open-cell foams are less dense, but the closed-cell foams have a higher strength.

Although the addition of neutron absorbers to the foam before pouring would prevent criticality, the radiation from the used nuclear fuel may also cause material degradation. In Huang et al. (2007), the radiation dose at which degradation begins is 10^6 Gy. This is similar to the degradation threshold for resins. If foam were injected into the canister just before shipment of the used nuclear fuel that had been in extended storage for 100 to 300 years, this potentially would not be an issue. Future tests would be needed to verify this.

Table 16. Material Properties of Foams

Foam	Density (g/cm^3)	Thermal Conductivity (W/m-K)	Ignition Temperature (°C)
Polyurethane	0.013–0.160	0.03	400
Polystyrene	0.032–0.050	0.03	350
Aluminum	0.216–0.675	5.80	660[a]
Steel	0.040–0.950	0.80	1535[a]
Silicon Carbide	0.257–0.803	5.28	2700[b]

a. Melting point
b. Sublimation point
Note: The material properties are representative of the type of foam and the properties of specific foams may vary.

5.5 Grout

Two studies (Anderson 1981 and Oversby and Werme 1995) discussed in Chapter 2 evaluated the use of cement grout (i.e., concrete or mortar) as a fill material for used nuclear fuel canisters. Sweden also evaluated concrete as a fill material in a titanium canister used for very deep hole disposal (SKB 1992). Grout is commonly used to solidify liquid low-level radioactive and to stabilize low-level radioactive waste prior to disposal. Grout has also been used to solidify liquid high-level radioactive waste in Italy (Alonzo et al. 2001) and to stabilize empty high-level radioactive waste tanks at the Savannah River Site. Grout has also been used to stabilize used nuclear fuel sludge at the Hanford Site.

A primary issue associated with using grout as a fill material would be its weight. Grout has a density of about 2.0 g/cm^3 which would increase the weight of a canister with used nuclear fuel contents by 17,600 to 30,900 lb. A significant consequence of the combined increased weight and addition of grout to the canister’s contents would be that existing canisters would not comply with the requirements of their current 10 CFR 72 storage certificate of compliance and their 10 CFR 71 transportation certificate of compliance. Therefore, for current canister designs, adding grout would not be possible unless the safety analyses for storage and transportation were revised to demonstrate that the canisters with grout fill material would satisfy the requirements of U.S. Nuclear Regulatory Commission regulations and U.S. Nuclear Regulatory Commission approval was obtained. It might be feasible to ship the heavier canisters if the used nuclear fuel had cooled sufficiently to allow an offsetting reduction in the shielding needed and a new design for the transportation cask with reduced weight was developed and certified. Future canisters and their associated transportation casks would need to be designed with somewhat lower capacities unless the designs assumed longer cooling times before transportation and the resulting reduced weight of transportation casks offset the increased weight of the canisters.

Another issue associated with using grout as a fill material would be its ability to flow between the fuel rods and around other structural materials in the canister. Future tests would be needed to verify that this was feasible. In addition, cement grout contains water, which is a neutron moderator, so a neutron absorbing material might have to be added to the grout to ensure subcriticality. Also because grout contains water, future tests would also be necessary to evaluate radiolysis and interactions of grout with fuel cladding, fuel, neutron poisons, fuel baskets, other structural materials within canister, the canister itself.

6. CONCLUSIONS AND RECOMMENDATIONS

Based on the weight of the potential fill materials discussed in Chapter 5, adding fill materials to existing canisters would result in the canisters not meeting the current requirements of their 10 CFR 72 storage certificate of compliance and their 10 CFR 71 transportation certificate of compliance. Depending on the cooling time assumed for the used nuclear fuel, future canisters and their associated transportation casks might need to be designed with lower capacities and thicker walls to allow for the increase in weight due to the addition of the fill material. Foam fill materials might be an exception to this.

Most studies that have evaluated fill materials and their properties have been literature reviews; few have been studies that conducted experiments. Also, from the perspective of the Used Fuel Disposition Campaign, a significant gap in the existing studies is that none have evaluated the performance of the fill materials during the normal conditions of transport or during hypothetical transportation accident conditions. Studies that addressed this gap would need to include ones that assessed the ability of the fill material to maintain its own geometric configuration (e.g., not slump) and maintain the geometric configuration of the used nuclear fuel under normal conditions of transport and under hypothetical accident conditions. Such studies would provide the information that would be needed to determine whether credit could be taken for the fill material being able to exclude water moderator or provide neutron absorbers such that the fissile material package requirements in 10 CFR 71.55 could be shown to be satisfied.

Consequently, the use of fill materials to stabilize used nuclear fuel in canisters would require a comprehensive experimental program. Especially important would be:

- Experiments that would evaluate the interactions among the fill material, fuel cladding, fuel, fuel baskets, neutron poisons, and other structural materials including the canister itself.
- Experiments that would determine if a fill material could be efficiently, effectively, and reliably emplaced inside a canister containing used nuclear fuel, filling the free volume without leaving an excessive number of, or large voids
- Experiments that would evaluate the efficacy of heat conduction from fuel rods in fuel assemblies through the fill material to the heat removal features of the canister and determine the resulting temperatures of fuel cladding.

Molten materials, particulates, and beads have been extensively studied as fill materials for waste packages and their ability to function in this capacity is reasonably well known. Nonetheless, the scope of the research and development effort would be greatest if molten metal fill was used, for which canisters that contain used nuclear fuel would have to be preheated and cooled under carefully controlled conditions. The research and development would necessarily determine the process and procedure, and alternatives, for retrieving used nuclear fuel from canisters where molten metal fill had been used. Other issues such as the compatibility of the molten metal fill material and fuel cladding and safety related components of a canister would need to be determined. Techniques would also need to be developed and demonstrated for filling a canister with molten metal, for determining that the fill was successful and that voids did not remain within the cast metal matrix, and for recovering from an unsuccessful fill.

Paraffin is an alternative molten material that might be used to fill canisters if the decay heat of the used nuclear fuel was not too great. Although paraffin is a neutron moderator and is flammable, a neutron absorber material might be dissolved in it as might a fire retardant material. As with molten metal fill materials, it would likely be necessary to heat canisters to ensure that the molten paraffin infiltrated into all of the available spaces in the canister. Unlike molten metal, high temperatures would probably not be necessary. It would be necessary to demonstrate that the paraffin would not be molten during normal transportation, could maintain the geometric configuration of the used nuclear fuel during normal and hypothetical accident conditions of transportation, and that it would not leak out following a transportation accident if temperatures were great enough to re-melt the material. Paraffin would be subject to radiolysis and therefore could not be used for extended storage during periods when the radiation source of the used nuclear fuel remained high.

If the fill material was a particulate or bead, it is likely that the canister and its used nuclear fuel contents would have to be vibrated during the filling process to ensure that the particles filled the available void spaces and with the desired packing density. It would be necessary to conduct research to develop techniques and tests to demonstrate, with high confidence, that particulate fill material would successfully infiltrate into all of the available open spaces within a canister and in and around the fuel assemblies leaving few if any voids. Because the condition of the fuel cladding would be suspect or unverified (otherwise, it would not be necessary to introduce fill material into a canister), tests would be necessary to determine if vibrating the canister could further damage the fuel cladding.

Because resins contain organic compounds, it may not be possible to formulate one that does not decompose or produce hydrogen or other gases when subjected to the heat and radiation environment in a dry used nuclear fuel canister, or when subjected to temperatures that might occur during hypothetical accident conditions.

Foams, especially inorganic foams, show some promise for use as fill materials. Nonetheless, it would be necessary to conduct extensive tests and demonstrations to show that a foam would reliably flow into and fill, at the required density, all of the void spaces and in and around the fuel rods in a canister that contained used nuclear fuel. Also, as with resins it would also be necessary to demonstrate that the foam did not decompose or produce hydrogen or other gases as a consequence of being exposed to heat and radiation or to temperatures that would exist following hypothetical transportation accident conditions.

Grout has also been extensively studied for stabilizing low-level radioactive waste and other waste and its ability to function in this capacity is also reasonably well known.

In addition to the research and development that would be required for the fill material that would be used, the process for emplacing the fill material into canisters containing used nuclear fuel would have to be demonstrated for its reliability, safety, and efficiency. The process would need to be located in a dedicated facility, possibly a facility that can be disassembled and moved for use at multiple sites, or a dedicated area within an existing facility. Whether the process was to be installed in a new facility or an existing facility, it would be necessary to design, license, and construct/install the facility at every site where canisters were to be filled with a fill material, or develop a mobile facility. Conceptual designs for a facility that could be moved among sites have been proposed in the past. Such a design might be adopted, with modifications for use at a single site where fill material was to be placed into canisters containing used nuclear fuel. The design of such a facility could require substantial research and development.

Before fill materials could be used to stabilize used nuclear fuel contained in storage and transportation canisters a substantial development, design, and licensing effort would need to be undertaken. In addition, the results of previous work show that use of fill materials to stabilize used nuclear fuel inside storage and transportation canisters would present significant technical challenges. Therefore, further research on the use of fill materials to stabilize used nuclear fuel during storage and transportation is not recommended unless options such as showing that the fuel remains intact or canning of used nuclear fuel do not prove to be feasible.

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