Use of Depleted Uranium Silicate Glass To Minimize Release of Radionuclides from Spent Nuclear Fuel Waste Packages

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January 20, 1996

Prepared For Presentation At

Paper # 14a06
Session # 99
Spent Nuclear Fuel Conditioning for Disposal
Sponsored by: Group 14 - Nuclear Engineering Division
Spring Annual Meeting
American Institute of Chemical Engineers
New Orleans, Louisiana
February 25 - 29, 1996

Unpublished

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ABSTRACT

A Depleted Uranium Silicate COntainer Backfill System (DUSCOBS) is proposed that would use small, isotopically-depleted uranium silicate glass beads as a backfill material inside repository waste packages containing spent nuclear fuel (SNF). The uranium silicate glass beads would fill the void space inside the package including the coolant channels inside SNF assemblies.

Based on preliminary analysis, the following benefits have been identified. DUSCOBS improves repository waste package performance by three mechanisms. First, it reduces the radionuclide releases from SNF when water enters the waste package by creating a local uranium silicate saturated groundwater environment that suppresses (a) the dissolution and/or transformation of uranium dioxide fuel pellets and, hence, (b) the release of radionuclides incorporated into the SNF pellets. Second, the potential for long-term nuclear criticality is reduced by isotopic exchange of enriched uranium in SNF with the depleted uranium (DU) in the glass. Third, the backfill reduces radiation interactions between SNF and the local environment (package and local geology) and thus reduces generation of hydrogen, acids, and other chemicals that degrade the waste package system. Finally, DUSCOBS provides a potential method to dispose of significant quantities of excess DU from uranium enrichment plants at potential economic savings. DUSCOBS is a new concept. Consequently, the concept has not been optimized or demonstrated in laboratory experiments.

1. INTRODUCTION

Spent nuclear fuel (SNF) is highly radioactive. The radionuclides in SNF decay in time to nonradioactive isotopes. The basic approach for safe disposal of SNF is to isolate the SNF from the environment until the decay has resulted in very low levels of radiation. The decay time is measured in thousands of years.

The approach adopted by the United States for disposal of SNF is to bury the SNF hundreds of meters underground in special waste packages in a geological repository. The primary potential failure mode for a geological repository is dissolution of radionuclides with SNF into groundwater and movement of groundwater to the open environment. This can be prevented by two mechanisms: (1) mechanical barriers to slow groundwater movement and (2) chemical barriers to slow the dissolution of SNF in the repository. In traditional chemical engineering one objective is to create conditions for rapid chemical reactions to minimize plant size. For a SNF geological repository, the chemical engineering objective is to create chemical conditions to delay SNF dissolution until most of the radioactive materials have decayed to non-radioactive materials.

A new approach (Forsberg et. al., 1995) to improve the performance and potentially lower the costs of SNF disposal has been identified - the Depleted Uranium Silicate COntainer Backfill System (DUSCOBS). This involves backfilling SNF waste packages with small glass beads with a high concentrations of depleted uranium (DU) in the glass. Depleted uranium is uranium low in the fissile isotope ^{235}U. A mechanical description of the DUSCOBS is provided herein and the projected depleted uranium silicate (DUS) glass characteristics are summarized. The operation of DUSCOBS is described in terms of minimizing radionuclide release to the environment.

The United States [U.S. Department of Energy (DOE), 1994] is considering use of a multipurpose canister (MPC) for the storage, transport, and disposal of light-water reactor (LWR) SNF. DUSCOBS is analyzed in terms of this specific system. DUSCOBS is applicable to alternative MPC designs and other SNF repository systems. DUSCOBS is also applicable to alternative types of SNF and other fissile materials.

The MPC (Fig. 1) is a thin-walled, stainless steel container that could be produced in different sizes. A representative MPC would contain 21 pressurized-water reactor (PWR) SNF assemblies. The MPC could be loaded with SNF at the reactor, at an away from reactor (AFR) storage facility, or at the repository. Once loaded, it is intended that the SNF would never be handled again as individual fuel assemblies. The MPC would become an inner container for all future SNF operations. For SNF storage at the reactor site or an AFR storage facility, the MPC would be placed inside a shielded storage module. A transportation cask would be used to transport the MPC from a reactor or an AFR storage facility to the repository. The MPC would then be placed within a disposal overpack. The MPC and the disposal overpack, which is known as the waste package, would then be placed in a geological repository. The use of the MPC (1) minimizes SNF handling, (2) reduces the potential for radiation contamination, and (3) avoids the need to build multiple baskets to hold SNF during transport, storage and disposal.

17 X 17 SQUARE ARRAY
PIN DIA 0.8 cm
PIN SPACING 1.25 cm

(a)

**MPC
NUCLEAR FUEL**

FUEL ASSEMBLY

(b)

**LOAD SPENT
NUCLEAR FUEL**

LID

(c)

**LID ON MPC
NUCLEAR FUEL**

FUEL ASSEMBLY

MULTIPURPOSE
CANISTER
SYSTEM

GLASS
BEADS

(d)

DISPOSAL
OVERPACK

**ADD GLASS
BEADS THROUGH
FILL HOLE**

Waste Package System
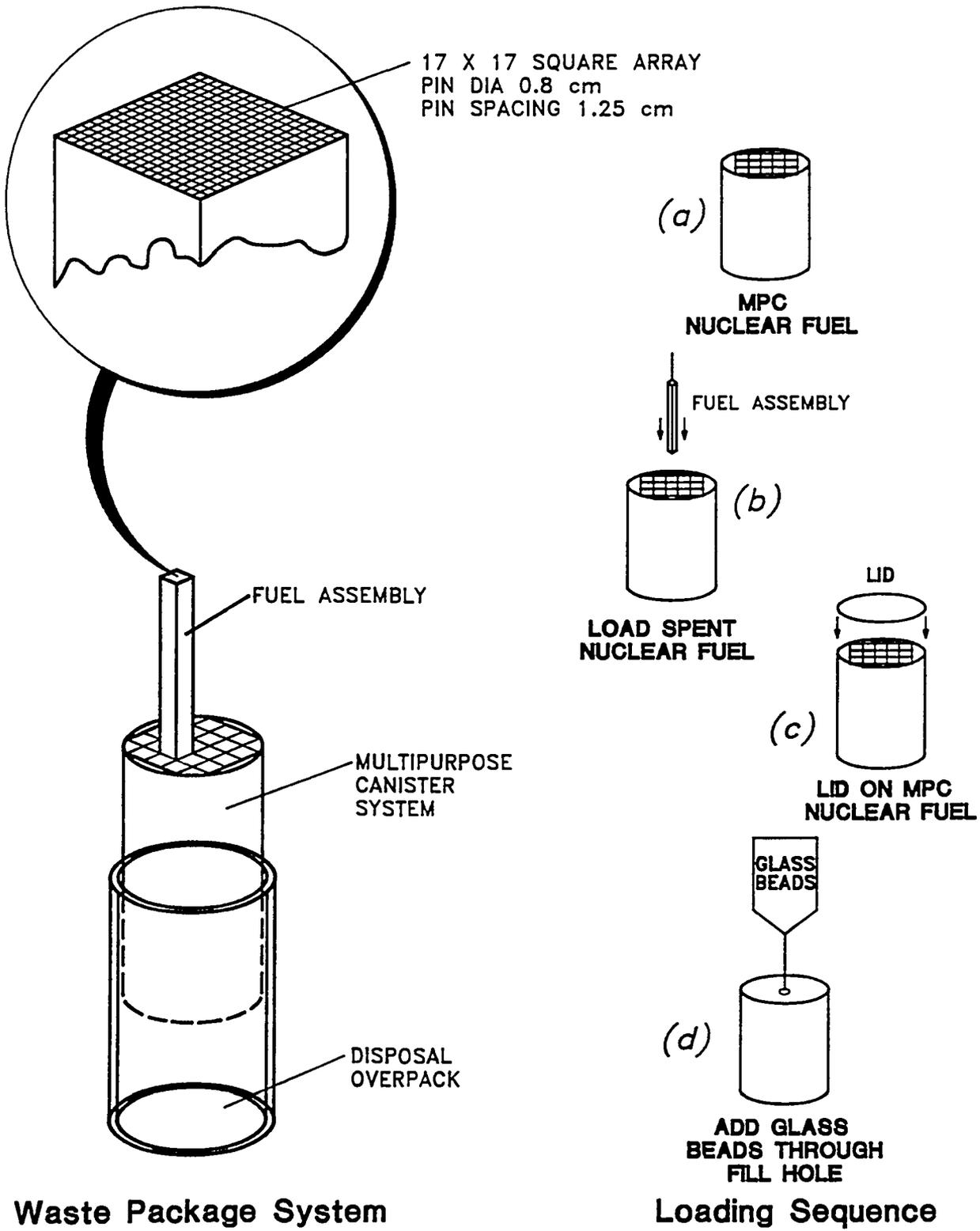
Loading Sequence

FIGURE 1: Waste package system and DUSCOBS and loading sequence.

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2. MECHANICAL DESCRIPTION OF THE DUSCOBS CONCEPT

In the implementation of DUSCOBS, the MPC would first be loaded with SNF (Fig. 1). After the SNF is loaded, the MPC would be backfilled with DUS glass beads. Depending upon the loading location, the glass beads would be loaded while the canister is open or added latter through a small fill hole in the MPC lid.[1] The beads would be sufficiently small—similar in size to fine sand—to fill the coolant channels between fuel rods. After the package is filled, the MPC would be sealed. One method of filling the MPC is illustrated in Fig. 1.

If desired, there are multiple options to hold the glass beads in place so the beads can not run out if the package is breached in an accident. For example, the package can be vacuum dried and heated to weld the glass beads together. With appropriate glass compositions, this can be achieved at relatively low temperatures compared to those required to melt glass.

For a large MPC designed for 21 PWR SNF assemblies, most of the space in the MPC is void space that can be filed with glass beads. The MPC has an internal volume of 7.9 m³ with the fuel assembly baskets having a solid volume of 1.1 m³. The 21 PWR SNF assemblies have a solid volume of only 1.6 m³. The fuel assemblies are mostly empty coolant channels. Thus, 5.2 m³ of the 7.9 m³ of internal volume in the 21 PWR assembly-loaded MPCs can be filled with beads. The fraction of the canister that is void space depends upon the MPC size and fuel type, but in all LWR SNF waste package designs, most of the canister volume is void space (Sadeghi, Pigford, Chambre and Lee, 1990).

3. URANIUM SILICATE GLASSES

There are several choices of uranium silicate backfill materials. The preferred choice will depend upon complex trade-offs on different performance characteristics and economics.[2] Table 1 provides the chemical composition of three uranium glasses that have been manufactured (Ramsey 1994; Chakrabarty 1969).

Loffler glass has a water leachability about three orders of magnitude lower than that of conventional HLW borosilicate glass. This leachability is noteworthy in terms of long-term durability of the backfill material and the ability to choose glasses with high uranium loadings and any desired solubility over a broad range of properties. Loffler glasses have been loaded up to 30 wt % uranium dioxide (UO_2) equivalent. Soda lime glass is used as bottle glass and for other purposes. Soda lime glasses with uranium loadings up to 50 wt % U_3O_8 equivalent have been made.

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[1]The method for filling the MPC (loaded with SNF) with DUS glass beads has not been explored; however, it is the view of the authors that a number of alternatives are possible. Trade-off studies and demonstration testing will be required to define the best method for filling the MPC with DUS glass beads.

[2]These tradeoff studies have not been undertaken.

Table 1. Example glass compositions

Compound	Loffler (5.0 g/cm³)		Soda lime (4.1 g/cm³)		Sodium silicate	
	mol %	wt %	mol %	wt %	mol %	wt %
Al_2O_3	18.0	14.5	0.0	0.0	0.0	0.0
B_2O_3	5.4	2.97	0.0	0.0	0.0	0.0
BaO	2.8	3.39	0.0	0.0	0.0	0.0
CaO	0.0	0.0	16.1	9.6	0.0	0.0
Na_2O	0.0	0.0	9.6	6.3	18.3	11.2
PbO	5.8	11.0	0.0	0.0	0.0	0.0
SiO_2	49.4	23.45	69.8	44.2	62.1	36.8
UO_2	**9.0**	**19.2**	0.0	0.0	**19.6**	**52.0**
U_3O_8	0.0	0.0	**4.5**	**40.0**	0.0	0.0
Rare earths (e.g., Nd_2O_3)	9.6	25.5	0.0	0.0	0.0	0.0

One other backfill material deserves special note. This material is UO_2 with a theoretical density of 10.96 g/cm^3 (Dean, 1973). Uranium dioxide has several advantages as a backfill material including its high density and well-known characteristics. It has two potential disadvantages. First, its chemical properties can not be modified to optimize SNF leach resistance or other desired characteristics. With glasses, there are a range of chemical and physical characteristics that can be optimized. Second, current fabrication techniques for UO_2 beads may be more expensive than glass beads. There are other DUS compounds that are also potential backfill materials. A better understanding of the system is required before decisions are made.

4. REPOSITORY BENEFITS AND CONSIDERATIONS

The goal of a repository is to prevent the release of radionuclides from the SNF to the open environment. Because radionuclides decay, over time, to nonradioactive isotopes, radionuclide release is prevented only if the radionuclides in SNF are isolated for a sufficient time for decay to nonradioactive materials. DUSCOBS may improve isolation in several ways.

4.1 REDUCED RADIONUCLIDE RELEASE RATE FROM WASTE PACKAGE

The dominant expected repository failure mode is radionuclide migration to the open environment by (1) water leaching of SNF, (2) dissolution of radionuclides and generation of colloids, and (3) transport of those radionuclides in dissolved or colloid forms to the open environment. DUSCOBS offers the potential to reduce radionuclide releases by two mechanisms: (1) reduced water flow through the MPC inside the disposal overpack and (2) reduced radionuclide release rates from the package.

4.1.1 Mechanisms of Degradation of SNF in a Geological Repository

LWR SNF consists of bundled sets of Zircaloy tubes, each of which contains UO_2 fuel pellets. The fission products and actinides are primarily located within the UO_2 fuel pellets. Most of the radionuclides can not escape until the UO_2 matrix is destroyed. The release of fission products and actinides from the SNF is limited by the rate of alteration and dissolution of the UO_2 fuel matrix (Finn, Hoh, Bates, and Wolf, December 1994).

Over geological time, the waste package and the SNF cladding will fail, and the UO_2 will be exposed to the repository environment. The initial chemical reactions with water in many cases form a protective layer that slows alteration of the UO_2 (Nicol and Needes, 1975; Nicol and Needes, 1977; Shoesmith, Sunder, Bailey, and Wallace, 1988). The detailed mechanisms of SNF degradation and radionuclide release are dependent upon the local groundwater chemistry—particularly the oxidation potential of the groundwater (Johnson and Shoesmith 1988).

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4.1.1.1 Oxidizing Groundwater Impacts on SNF

In an oxidizing groundwater environment such as at the proposed U. S. Yucca Mountain repository site, the uranium in the UO_2 will be oxidized, over time, from the +4 valence state to the +6 valence state. Uranium in the +6 valence state is more soluble in groundwater than is uranium in the +4 valence state. The actual solubility of the uranium is also dependent upon other chemical species in the groundwater. If silicates are dissolved in the groundwater, the uranium will usually undergo a series of chemical transformations to more stable, less soluble uranyl silicates (Bruton and Shaw, 1988; Finn, Hoh, Bates, and Wolf, December 1994). Silicates often control the solubility of uranium. It is found that silicates can lower the solubility of uranium in water by more than two orders of magnitude—presumably because of the lower solubility of uranium silicates compared to other uranium compounds.

The transformation process of UO_2 to other uranium compounds that can allow the escape of water soluble fission products and the formation of americium and curium colloids that can be transported by groundwater to the environment have been studied extensively. The transformation processes observed in laboratory tests of uranium oxide (Wronkiewicz, et al., 1992) and SNF (Finn, et al., December 1994) are similar to those transformations seen in uranium ore bodies over geological time (Murphy, 1995). The transformation processes are initially rapid, but they slow with time and the formation of protective uranyl silicate surfaces on the UO_2.

4.1.1.2 Reducing Groundwater Impacts on SNF

With chemically reducing groundwater, UO_2 does not change its chemical form. It is thermodynamically stable. The dissolution rate of UO_2 is determined by total water flow and the difference between the uranium dissolved in the groundwater and the solubility of uranium in groundwater. The solubility of uranium in reducing groundwater is low.

4.1.2 Chemical Effects of DUSCOBS on Repository Performance

Backfilling the waste package with a DUS glass creates a groundwater environment within the waste package saturated in uranium species and silicates. The small glass beads with their high surface areas ensure that the groundwater is saturated with this DU without fission products rather than uranium with fission products from the SNF. This uranium-silicate-saturated environment slows the transformation and dissolution of the UO_2 in the fuel elements via several mechanisms.

The rate of uranium dissolution is proportional to the difference between the uranium concentration in the water at the SNF UO_2 surface and the solubility of uranium in that groundwater. Uranium-saturated groundwater does not dissolve uranium fuel pellets. Silicates further reduce SNF transformation and dissolution by multiple mechanisms:

Solubility limits. Uranium silicate is much less soluble in most groundwaters than most other uranium compounds (Sadeghi, et al., 1990). This low solubility reduces the quantity of uranium that can dissolve in a unit of groundwater. It implies much longer times until the uranium in the waste package is dissolved and transported. This in turn implies better radionuclide isolation.

Mass transfer barriers. The uranium-silicate-saturated solution also helps form insoluble layers of uranium silicates over the UO_2 crystals in the SNF. This phenomenon slows uranium alteration and dissolution. This mechanism would be expected to be particularly important in oxidizing groundwater environments. In such environments other soluble radionuclides may be released and colloids may be formed during chemical transformations. The transformations may not result in much uranium transport, but they can create the potential for release of other radionuclides.

4.1.3 Physical Effects of DUSCOBS on Repository Performance

The proposed DUSCOBS backfill serves several other functions. It lowers the hydraulic conductivity within the package and thus ensures low groundwater flow rates inside the waste package. Water flow is the primary mechanism for the transport of radionuclides to the open environment. The release of many radionuclides is controlled by solubility limits (Sadeghi et al., 1990). The slower the flow of groundwater, the slower the transport of radionuclides.

The backfill may also reduce colloid transport. Experimental evidence indicates that much of the americium and curium are transported as colloids rather than as dissolved in the water (Finn, Gong, Bates, Emery, and Hoh, 1994). Particle sizes typically exceed 1 nm. The beads may act together as a barrier for these colloids. For optimum removal capabilities, appropriate glass-bead sizes, distributions of sizes, and surface characteristics for colloid retention are required.

Last, the backfill helps maintain the form of package for long periods of time. The total radionuclide releases from a package by groundwater flow depends upon groundwater flow in and around the waste package. The cylindrical shape of a waste package has a relatively small surface-to-volume ratio, which in turn minimizes the interactions of water with the package. However, the waste package contains significant void space. If the package is not backfilled, it will ultimately collapse and further disturb the local geology and produce more flow of water by the radioactive materials.

4.2 AVOIDANCE OF NUCLEAR CRITICALITY

Nuclear criticality should be avoided in a geological repository to prevent the release of radionuclides to the environment. Naturally occurring nuclear reactors in the geological past [Cowan, 1976; International Atomic Energy Agency (IAEA), 1975; IAEA, 1977; and Smellie, 1995] indicate that such events generate both added radioactivity and heat over time periods of

hundreds of thousands of years. The heat generated creates higher repository temperatures that (1) drive chemical reactions, which in turn, degrade SNF packages and SNF; (2) cause water movement within a repository that may transport radioactivity to the environment (Buscheck and Nitao, 1993); and (3) create major uncertainties in repository performance. Water movement can be accelerated in both unsaturated (Buscheck, Nitao, and Wilder, 1993) and saturated geological environments by heat. In this context, it is important to emphasize that the concern is not that nuclear criticality occurs or that some radioactivity is added to the repository, but rather that it may occur sufficiently often for a sufficient time to generate significant amounts of heat. Heat is a driver for groundwater movement and hence radionuclide transport.

After SNF is disposed of, there is significant radioactive decay heat. To minimize the potential impacts of heat on repository performance, the SNF is packaged in long-lived waste packages. The radioactive decay heat decreases to low levels before the waste packages degrade significantly. The package provides waste isolation during the periods of high heat release. Nuclear criticality, should it occur, would occur after loss of waste package integrity.

There are two types of repository criticality concerns (Fig. 2): nuclear criticality involving a single waste package with a single MPC and nuclear criticality involving fissile material from multiple MPCs.

4.2.1 Package Criticality

Over time, the waste package and the MPC will degrade. Water will selectively leach components from the MPC. In particular, it is known (Vernaz and Godon, 1992) that boron and certain other neutron poisons will leach preferentially from a waste package (Fig. 2). Subsequently, if the MPC contains sufficient fissile material, it may become critical. This type of nuclear criticality is primarily a problem associated with large MPCs loaded with PWR SNF which have the highest total fissile content within a single MPC.

DUSCOBS prevents this type of nuclear criticality because DU is uranium that is also a neutron poison. The uranium in the SNF and the DU in DUSCOBS transform to the same chemical compounds with the same chemical characteristics; hence, they do not separate from each other (Sect. 4.1). The use of DUSCOBS to prevent repository package nuclear criticality was investigated for the MPC using a very conservative set of assumptions:

> *Fuel characteristics.* Fresh PWR nuclear fuel with no burnup credit.

> *Basket characteristics.* Neutron poison (boron) in the MPC grid structure to be fully leached from the package.

> *Glass characteristics.* DUSCOBS glass to be uranium-loaded soda lime glass in which the only major neutron absorber is DU. The alternative Loffler glass has a higher neutron absorption capability because of its boron and rare earth content.

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INITIAL CONDITIONS $(T=T_0)$

× NEUTRON POISON
○ FISSILE MATERIAL

WATER

FLOW

MULTIPURPOSE
CANISTER
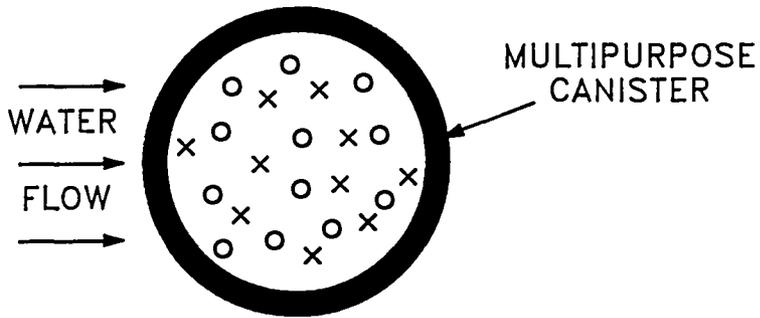
PACKAGE CRITICALITY $(T=T_0+T_1)$

WATER

FLOW
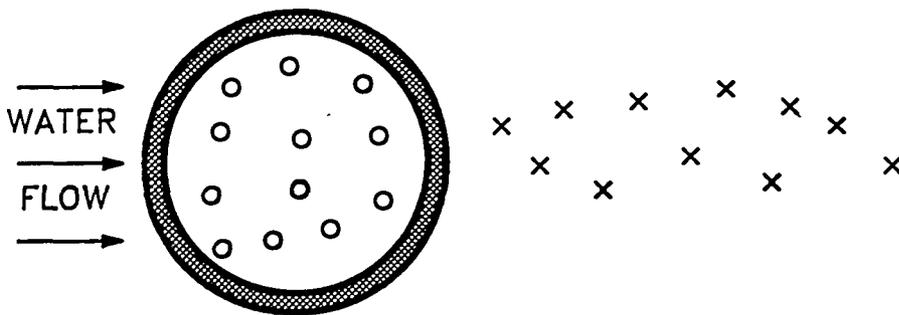
ZONE CRITICALITY $(T=T_0+T_1+T_2)$

WATER

FLOW

FIGURE 2. Alternative repository criticality scenarios.

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The addition of DU eliminates package criticality concerns. The calculated neutron multiplication constant (k_{eff}) is <1 (nuclear criticality occurs at $k_{eff}=$ 1) and below the regulatory requirements of k_{eff}<0.95. Table 2 shows the neutron multipl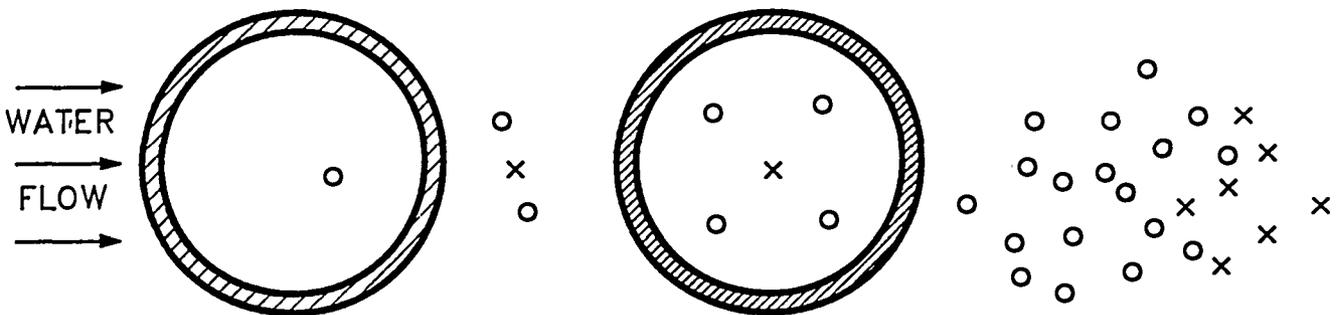ier (k_{eff}) for several examples of backfill in the MPC: air, water, DUSCOBS with air between glass beads, and DUSCOBS and water between glass beads (Forsberg et. al., 1995).

Table 2. Package criticality: neutron multiplication factor (k_{eff}) for fully loaded MPCs in which all boron has been removed from the waste package[a]

Filler material	Fresh fuel enrichment levels	
	3.75%	5.0%
Air	0.4059	0.4690
DUSCOBS with air[b]	0.4337	0.5057
Water	1.1988	1.2643
DUSCOBS with water[b]	0.8756	0.9347

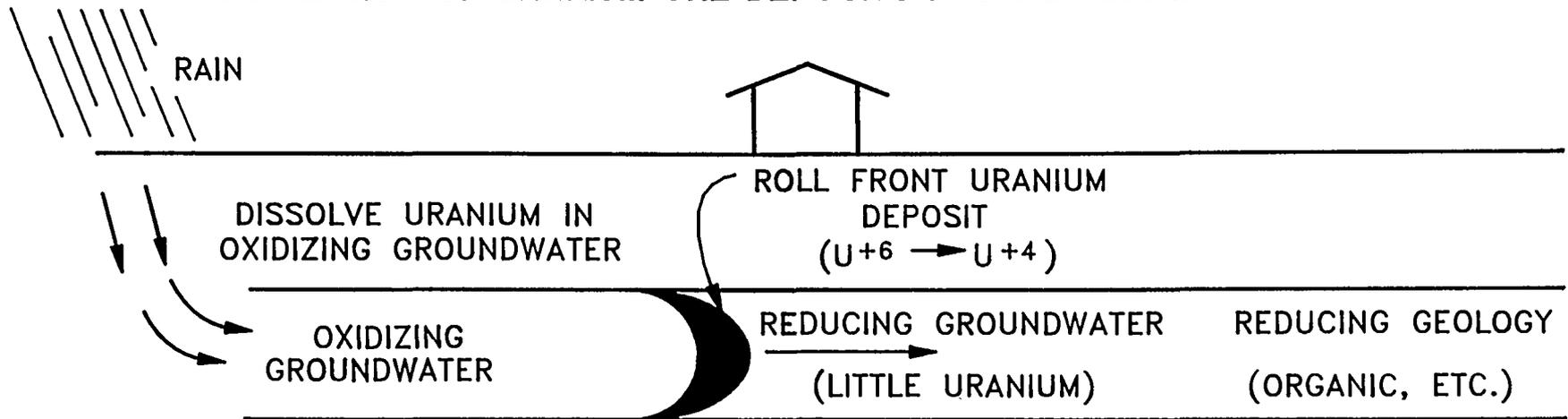[a]Regulatory requirement is k_{eff}+ uncertainty + bias <0.95.

[b]Assumes backfill material is 65 vol % glass and 35 vol % void space. The assumed glass density is 4.1 g/cm^3 with 25 wt % DU with 0.2 % ^{235}U.

4.2.2 Zone Criticality

4.2.2.1 Mechanisms of Zone Criticality

In a geological environment, chemical neutron poisons (boron, rare earths, cadmium, etc.) will separate from the uranium, and the uranium will dissolve in groundwater, migrate and then redeposit. Uranium dissolves in oxidizing groundwater and then precipitates under chemically reducing conditions (Wronkiewicz et. al., 1992; Smellie, 1995). Uranium may also be precipitated by the formation of less soluble uranium species in the same uranium oxidation state. These are the chemical mechanisms that created natural uranium ore bodies. In a repository, the same geological mechanisms will operate and may concentrate and purify uranium (Fig. 3). Some of these mechanisms may be accelerated within repositories with oxidizing conditions by the inclusion of chemical reducing agents in the repository (i.e., iron in waste packages) and tunnel support systems (i.e., rock bolts, etc.) that create local chemical reducing conditions for buildup of uranium deposits. This is a much longer term phenomena (Fig. 2) than package criticality and occurs later in time ($T = T_0 + T_1 + T_2$).

FORMATION OF URANIUM ORE DEPOSITS FROM URANIUM IN ROCK

RAIN

DISSOLVE URANIUM IN
OXIDIZING GROUNDWATER

ROLL FRONT URANIUM
DEPOSIT
$(U^{+6} \longrightarrow U^{+4})$

OXIDIZING
GROUNDWATER

REDUCING GROUNDWATER

(LITTLE URANIUM)

REDUCING GEOLOGY

(ORGANIC, ETC.)

FORMATION OF URANIUM ORE DEPOSITS FROM SPENT NUCLEAR FUEL URANIUM

RAIN

OXIDIZING
GROUNDWATER

URANIUM
DEPOSIT
$(U^{+6} \longrightarrow U^{+4})$

DISSOLVED URANIUM

IN GROUNDWATER

REDUCING GROUNDWATER

(LITTLE URANIUM)

DEGRADED SNF
WASTE PACKAGE

IRON (WASTE PACKAGE,
ROCK BOLTS, ETC.)
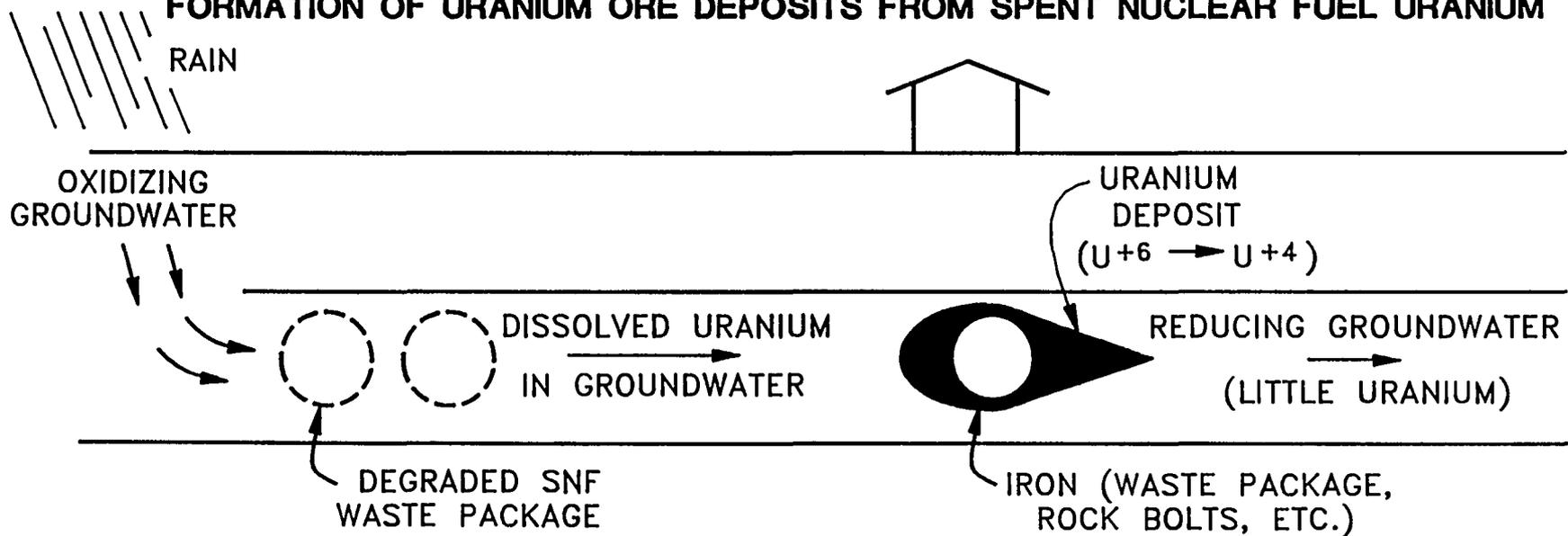
FIGURE 3. Natural and man-made formation of uranium ore deposits.

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If (1) the uranium enrichment is sufficiently high, (2) the quantities of uranium are sufficient, and (3) the uranium deposit has the appropriate geometry, then nuclear criticality will occur. Historical, experimental, and theoretical information indicates the conditions under which this may occur. Together this information suggests that if the uranium enrichments are above 1.3 wt %, nuclear criticality in a geological repository is a possibility:

History. The historical geological record (Brookins, 1990; Cowan, 1976; and Smellie, 1995) shows that nuclear criticality has occurred in natural uranium ore bodies because of these mechanisms in the past. At the Oklo, Africa, site, 15 fossil, natural, nuclear reactors have been identified which operated when the ^{235}U enrichment of natural uranium on earth was approximately 3.6%. After operation with the generation of heat and fission products, ^{235}U enrichments of the uranium were as low as 1.3%—equivalent to the fissile enrichment of full-burnup LWR SNF. Today natural uranium deposits have a ^{235}U enrichment level of 0.71 % due to decay of ^{235}U over time. Nuclear criticality can no longer occur in natural uranium ore bodies due to these low enrichment levels.

Geochemical modeling of uranium ore deposits. The French atomic energy commission has studied the conditions under which natural nuclear reactors form (Naudet, 1977). Their analysis indicates that nuclear criticality may occur at enrichments as low as 1.28%; but, criticality becomes reasonably probable in some geological environments as enrichments approach 1.64%.

Engineering experiments and analysis. Criticality calculations [American National Standards Institute (ANSI), 1983] and laboratory experiments (Paxton and Pruvost, 1987) with the types of materials found in the natural environment indicate that nuclear criticality could, in theory, occur with fissile enrichment concentrations as low as 1% ^{235}U, but there is no experimental evidence of this having occurred in nature. Such criticality in a natural system would require nearly incredible conditions.

Environmental analysis. An analysis (DOE, October 1995) for disposition of excess uranium concluded that the uranium enrichment should be below 0.9% for high assurance that nuclear criticality will not occur in a disposal system.

Repository performance assessments. Modeling studies for disposal of high-enriched SNF in repositories using waste packages not filled with DUS glass beads show nuclear criticality to be the major technical issue for disposition of such fuels (Rechard, 1993; Patric, 1992). The models conclude that criticality may occur in a repository as has occurred in the natural environment. The uncertainties associated with geochemical evolution of a repository with time make predictions of the future highly uncertain.

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4.2.2.2 Fissile Content of SNF

An analysis was made of the fissile content of the total LWR SNF inventory in the United States using the U. S. Department of Energy spent nuclear fuel data base (1993). LWR SNF contains several fissile materials—^{235}U, ^{239}Pu, and other isotopes. In time, fissile materials such as ^{239}Pu decays to ^{235}U, and ^{241}Pu decays to ^{233}U; thus, fissile plutonium in the SNF ultimately is converted to ^{233}U or ^{235}U. Therefore, long-term avoidance of nuclear criticality is avoidance of criticality from uranium enriched in ^{233}U and ^{235}U. For this analysis, the fissile content was defined as the sum of the ^{235}U, ^{239}Pu, and ^{241}Pu divided by the total uranium and plutonium content of the SNF. The results of the analysis are shown in Fig. 4. The average fissile content is 1.47%. The average fissile content of the geological repository is expected to increase with time because of five factors:

High burnup SNF. LWR fuels designed for high burnup are being developed and deployed. These fuels reduce (a) natural uranium requirements per unit energy produced and (b) the quantities of SNF to the repository, but the average fissile content (plutonium and uranium) of the SNF is higher. For example, increasing SNF burnup (Ludwig and Renier, 1989) from 33,000 megawatt-days (MWd)/t to 50,000 MWd/t increases the fissile content of the SNF by 0.1%. This directly follows from the higher fission product and burnable neutron poison levels in the SNF that, in turn, necessitate a higher fissile content in the discharged SNF.

Power reactor shutdown. Typically, when a power reactor is refueled, it replaces a fourth to a third of the reactor core. When the reactor is decommissioned, the full reactor core is discharged. Two-thirds to three-fourths of the SNF will not be fully burned and will consequently have higher enrichments. The quantities of excess fissile material in this SNF depend upon how far in advance the shutdown is known. The ordering of nuclear fuel is typically 6 to 8 years before the SNF is discharged. If a decision to shutdown a reactor is made fewer than 8 years in advance, there will be significant excess fissile material in the SNF. Only a few reactors to date have been shut down, and these have been shutdown only after a few years or less notice.

Low-cost nuclear fuel. Nuclear fuel is cheap, but nuclear power plants are expensive. Therefore, strong incentives exist to improve plant availability in a highly competitive environment. This implies that if a power plant is shut down to repair some failed equipment, it may be refueled at the same time to avoid a later refuelling outage. Refueling while a plant's equipment is being repaired is economically efficient, but it increases the fissile content of SNF.

Disposition of plutonium. With the end of the cold war have come plans to dispose of excess plutonium from nuclear weapons. This material may go into a geological repository (1) after it is mixed with HLW and converted to glass or (2) after it is fabricated into mixed-oxide nuclear fuel and then used as power reactor fuel. Its fissile enrichment levels, particularly with plutonium in HLW glass, will be significantly higher than that of LWR SNF.
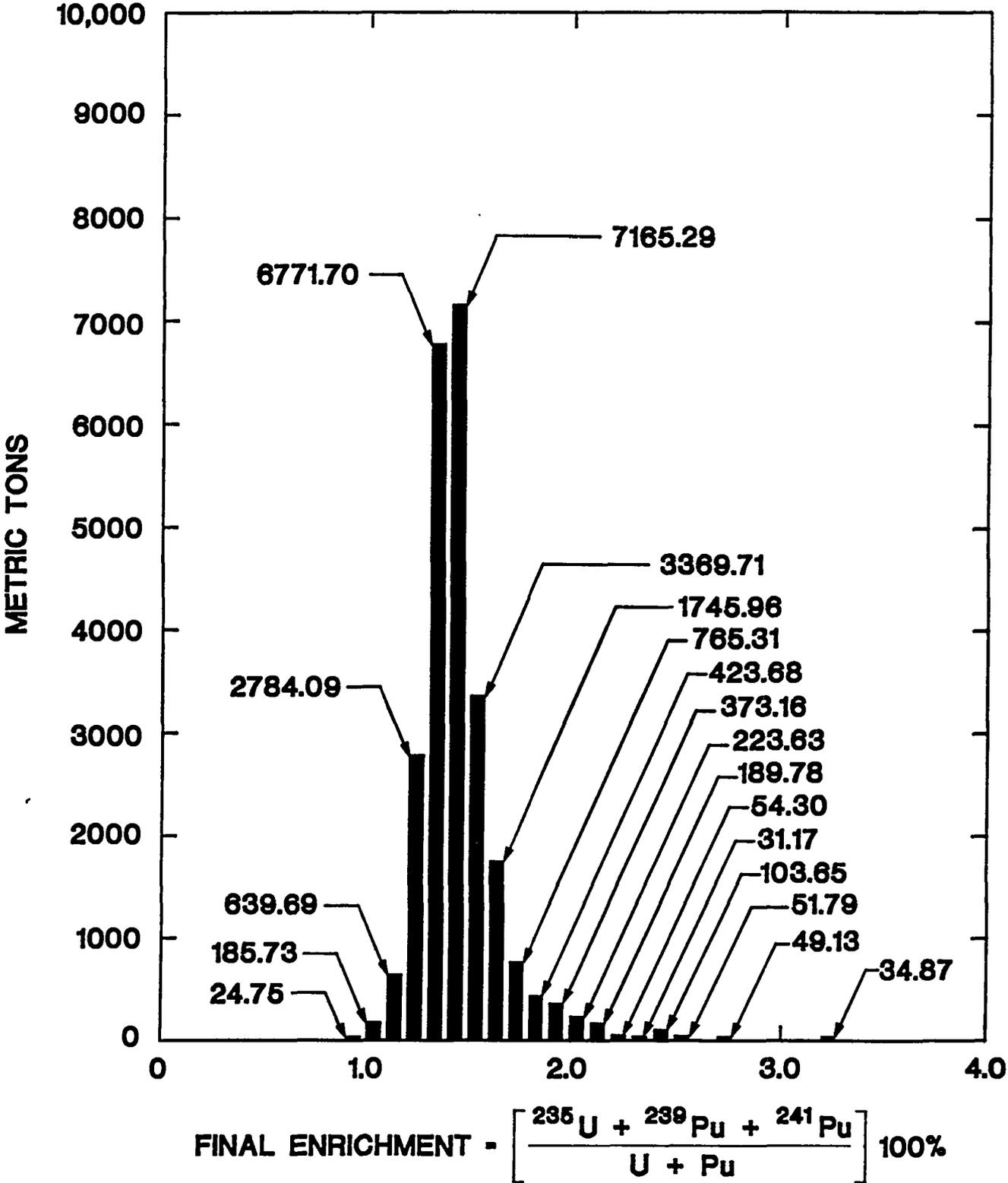
FIGURE 4. LWR SNF fissile enrichment levels.

$$\text{FINAL ENRICHMENT} = \left[\frac{^{235}U + {}^{239}Pu + {}^{241}Pu}{U + Pu} \right] 100\%$$

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Disposition of navy and research reactor SNF. There are significant quantities of these high-enriched SNFs which, if sent to the repository, will raise the average fissile enrichment of the repository.

The combination of these factors suggests that the average enrichment of the nuclear materials in the repository will likely exceed 1.6% and may be significantly higher.

4.2.2.3 Occurrence of Nuclear Criticality Based on Geological History

The fissile concentration of LWR SNF (1.47%) exceeds the minimum fissile concentration that was observed upon shutdown of natural reactors in the Oklo mining district in Gabon, Africa. The fissile concentrations of SNF are expected to increase with time. The enrichment of some SNF is much higher; this implies that part of the repository will have higher enrichments than the average enrichment. The total quantities of fissile material in projected repositories far exceeds the fissile content of any natural uranium ore body or a uranium mining district. The history of uranium ore deposits indicates a significant probability that nuclear criticality will occur, in time, within a geological repository for SNF. Geological history does not provide any information about when such events may happen and provides only limited information about the consequences.

Nuclear criticality concerns may be eliminated by backfilling the MPC with DU glass. Uranium in the fuel assemblies will isotopically exchange (Benedict, 1957) with uranium in the backfill, over geological time, as the uranium that has dissolved in water migrates within the waste package. Uranium ions in solution continuously exchange places with uranium ions in the various solids. There is no large energy barrier to isotopic exchange, and the large isotopic entropy of mixing effect encourages this reaction (Moore 1962). This mechanism allows isotopic dilution of the SNF uranium to levels below 1.3% creating a low enriched uranium (<1% ^{235}U) geological environment where nuclear criticality can not occur. The geological experience shows no examples of criticality with equivalent uranium enrichments below 1.3%.

Table 3 shows the average MPC enrichments for different backfill materials with different uranium densities—assuming that the DU has an enrichment of 0.2% and that the MPC is filled with PWR SNF. For uranium densities of the backfill material of 1 to 2 g of uranium per cubic centimeters, average package enrichments will be near 1%. It is noted that these package enrichments are for the MPC, which was designed for minimum free volume; other designs with greater free volume would result in even lower average package enrichments. The option exists to expand the size of the MPC or use an overpack with added DU glass to lower package enrichments to much lower levels.

In recent years, speculation has arisen that other types of criticality events might occur in geological repositories (Bowman and Venneri 1994) in addition to those demonstrated to have occurred at Oklo. However, these postulated criticality events appear to require very special conditions that are unlikely or perhaps impossible. It is noted that use of DU backfill would also

eliminate these theoretical criticality concerns because of the isotopic dilution of ^{235}U with large quantities of ^{238}U.

TABLE 3. Average MPC enrichments (wt %) for different PWR SNF enrichment levels and different uranium density backfill materials with 0.2% ^{235}U[a]

Uranium density in backfill (g/cm³)	Average SNF equivalent enrichment level (wt %)				
	1.4	1.6	1.8	2.0	2.2
0.5	1.15	1.31	1.46	1.62	1.78
0.75	1.06	1.20	1.34	1.49	1.63
1.0	0.98	1.11	1.24	1.38	1.51
1.25	0.92	1.04	1.16	1.28	1.40
1.5	0.87	0.98	1.09	1.20	1.31
1.75	0.82	0.92	1.03	1.13	1.24
2.0	0.78	0.88	0.98	1.07	1.17
2.25	0.75	0.84	0.93	1.02	1.11

[a]Assuming 9.689 t of uranium in 21 PWR SNF assemblies and 5.155 m³ void space in loaded MPC. For highly loaded uranium glasses, the glass densities are 4–5 g/cm³. Voids will occur between glass beads; hence, effective packing density will be 2.5–4 g/cm³. Uranium densities in glass vary from 20 to 50 wt %. Packing density is dependent upon the number of glass-bead sizes used. With optimum sizing, higher densities are possible.

4.2.2.4 Impacts of Plutonium

The foregoing analysis is based on the assumption that plutonium remains with the uranium until the major plutonium isotope (^{239}Pu) decays to ^{235}U and can be isotopically diluted by the DU. This is assured if the rate of plutonium decay to uranium is faster than the rate of dissolution and transport of uranium within the natural environment. The primary plutonium isotope, ^{239}Pu, has a half-life of 24,000 years (i.e., the decay rate is 3×10^{-5}/year).

The regulatory requirement (NRC, 1995) for waste package system performance is that the maximum allowable radionuclide release rate from the engineered barrier system shall not exceed 10^{-5}/year (1 part in 100,000/year) of the inventory of that radionuclide calculated to be present at 1000 years following permanent repository closure. For a waste package system to meet regulatory requirements, it would be expected that most of the plutonium would have had to decay to uranium and that the fissile decay-product uranium would be isotopically diluted by the DU before significant quantities of uranium or plutonium leave the package.

The experimental field data for naturally occurring reactors (Brookins, 1984; McCombie and McKinley, 1995; Brookins, 1990; Von Maravic, 1993; IAEA, 1975; IAEA, 1977) indicate that for most environments the plutonium will decay to uranium before the uranium or plutonium can be transported from the package. The uranium and plutonium within the package may, however, convert to more stable chemical forms within the package over the time frames of plutonium decay. Specific modelling studies of the proposed Yucca Mountain repository show very low release rates of plutonium and uranium in SNF (Sadeghi, Pigford, Chambre and Lee, October 1990). With such release rates, the plutonium will have converted to uranium long before significant releases of plutonium or uranium occur from the waste package system.

The inverse premise that plutonium will not decay to uranium before uranium transport in the local geological environment must also be considered. In a geological repository using DUSCOBS, the local geological environment will become saturated with DU. This has longer term implications. If it is assumed that, in a particular waste package because of some special local conditions, the uranium partly separates from the plutonium before complete plutonium decay to uranium, then residual plutonium will decay to uranium (primarily ^{235}U) and isotopically mix with any remaining uranium in the near-package environment. As this uranium is transported within the geological environment, it will most likely follow the flowpath of the uranium that was leached from the waste package earlier in time. This movement will bring it into contact with the uranium that was transported earlier. It will then be isotopically diluted by the DU in the local geological environment below enrichment levels at which nuclear criticality can occur.

In such scenarios there is separation of fissile uranium and plutonium; thus, the quantities of fissile material in any one location are decreased. This further reduces the potential of nuclear criticality.

These types of secondary mechanisms do not provide absolute avoidance of nuclear criticality, but they do decrease the probability of nuclear criticality in the repository. More importantly, these mechanisms limit the maximum possible heat generation from nuclear criticality events. A local geological environment saturated with DU ensures that most of the enriched uranium is diluted below concentrations at which nuclear criticality can occur. This limits potential heat generation by limiting the potential quantities of higher fissile enrichment materials in the repository. These considerations suggest that it is beneficial to dispose of any excess DU (beyond what is needed within the waste package for DUSCOBS) within the same repository area where the SNF is being disposed of.

4.3 REDUCED RADIATION INTERACTION WITH WASTE PACKAGE, WASTE PACKAGE CONTENT, AND NEAR-FIELD GEOLOGY

Radiation degrades the performance of a repository because of multiple mechanisms. The most important mechanisms may be radiolysis of air and groundwater components (Finn, et al., 1994). Radiation interactions with air create nitrogen oxides that react with water to generate acids. Radiation interactions with water generate hydrogen that can embrittle and weaken SNF clad. These materials accelerate SNF and package corrosion. These mechanisms create incentives to minimize the potential air and groundwater content in the waste package as the package degrades and to provide internal shielding material to absorb the radiation within the waste package.

Backfilling the waste canister with DUS materials would provide an internal radiation shield. The DUS glass has a solid density between 4.5 and 7 g/cm^3 with an effective particulate density between 3 and 5 g/cm^3. This density provides good gamma-shielding capabilities that reduce the radiation interactions with air and water. The backfill would also displace open space in the SNF assemblies, thus minimizing the volume available for air and groundwater available for interaction with radiation.
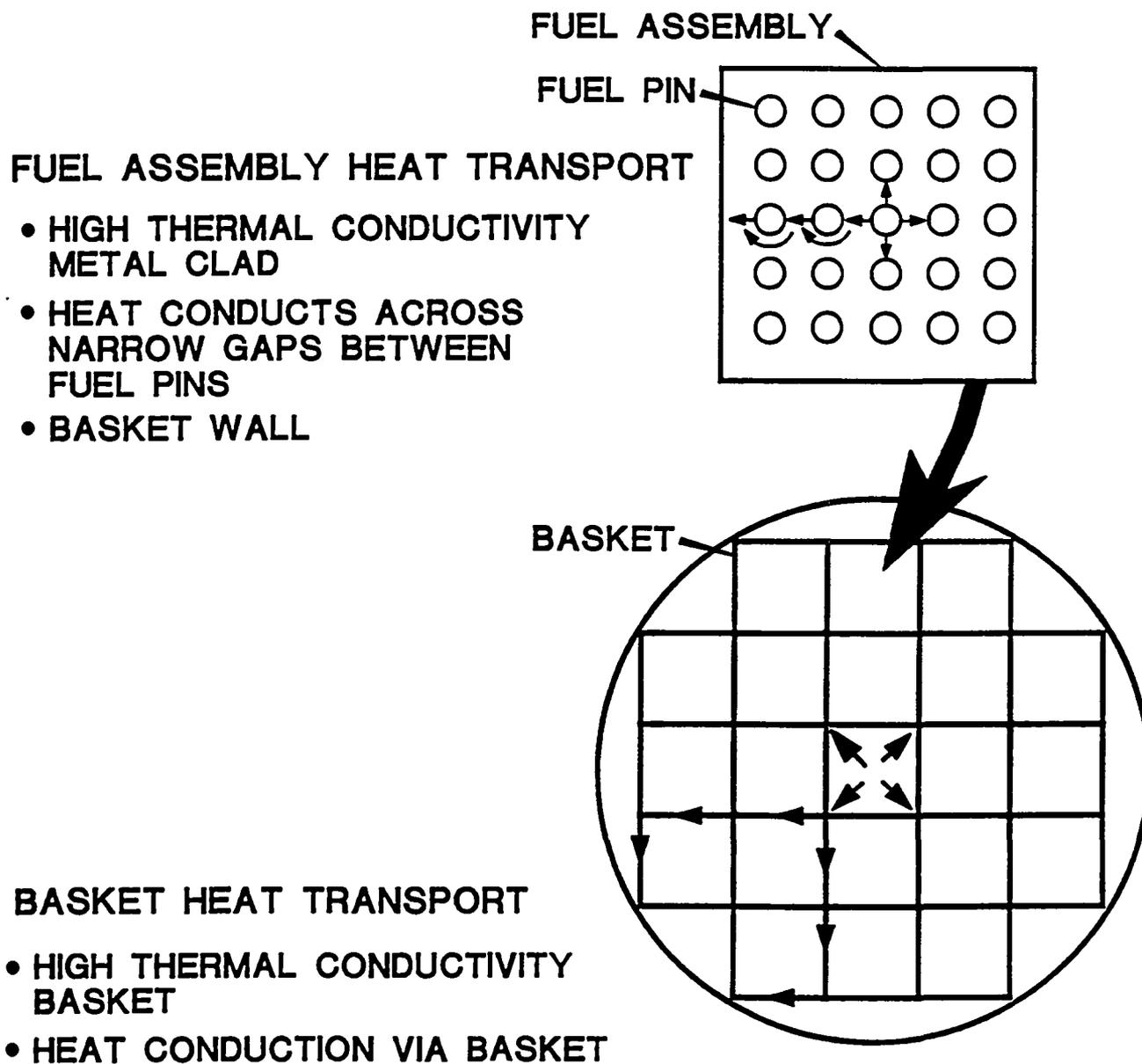
4.4 CONTROLLED INTERNAL PACKAGE TEMPERATURE

A major design constraint for the MPC is the maximum allowable SNF cladding temperature. The use of DUSCOBS does not significantly change the temperature within a waste package. However, it provides higher assurance of what those temperatures are (Forsberg, November 1995) and that unexpected events will not alter those temperatures. This is a result (Fig. 5) of several factors.:

> *Basket design.* The heat generated by the SNF must be transported from the SNF to the outside wall of the MPC. Although the fill material (air, water, or glass beads) transfers heat from the fuel pins to the inner MPC structure, it is not the dominant mechanism for heat transfer to the outer MPC wall because of the high thermal conductivity of the materials of which the basket is fabricated (boral and aluminum)

> *High conductivity SNF clad.* Within the fuel assembly, much of the heat is transferred by the fuel-pin metal cladding because of its high thermal conductivity. Heat is transferred from the cladding of one pin to the cladding of the next pin, across the narrow gaps between pins to the basket walls. Within a range of thermal conductivities, the impact of the fill material on the fuel pin temperature is low. The temperature drop across the fuel pin gaps is small compared to all of the other temperature drops across different materials from the UO_2 to the exterior of the cask.

> *Compensating heat transfer effects.* Heat is transported between pins within the fuel assembly by thermal convective air currents, thermal radiation, and thermal conduction. Adding glass beads reduces the heat transfer by the first two phenomena, but since the

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FUEL ASSEMBLY

FUEL PIN

FUEL ASSEMBLY HEAT TRANSPORT

- HIGH THERMAL CONDUCTIVITY
  METAL CLAD
- HEAT CONDUCTS ACROSS
  NARROW GAPS BETWEEN
  FUEL PINS
- BASKET WALL

BASKET

BASKET HEAT TRANSPORT

- HIGH THERMAL CONDUCTIVITY
  BASKET
- HEAT CONDUCTION VIA BASKET

FIGURE 5. Heat transfer in cask system

thermal conductivity of the glass is higher than the air, the net effect is calculated to result in about the same fuel pin temperature.

Based on the preliminary analysis, DUSCOBS reduces heat-transfer uncertainties. Heat transfer within the current design of MPC is partly by conduction, partly by radiation, and partly by gas convection. Radiation heat-transfer is strongly dependent upon SNF surface conditions. Gas convection heat transfer depends upon MPC orientation. In contrast, conduction heat transfer with glass beads is not impacted significantly by random factors such as SNF surface conditions or cask orientation.

4.5 REPOSITORY DESIGN TRADEOFF CONSIDERATIONS

The optimum glass composition and particle size for DUSCOBS will depend upon specific design trade-offs. For example, there is a trade-off in the size of the waste package. Large waste packages, such as the one consisting of an MPC with 21 PWR fuel assembles and an overpack, have the advantages of economics and of lower radionuclide release rates from the waste package over geological time. The lower release rates are possible because only so much water can flow by a single waste package. The dissolution of many fission products is solubility limited in groundwater. Furthermore, the dissolution and transport of uranium are limited by its solubility. The bigger waste package has less water flow by the package per metric ton of SNF.

However, large packages have two disadvantages: greater potential for nuclear criticality and potentially higher waste package temperatures resulting from higher SNF loadings. The advantages of a large package exist only if temperatures can be managed and nuclear criticality avoided. If large packages are chosen, then the optimum glass should have a high thermal conductivity to minimize package temperature, and the glass should be optimized to minimize the risk of nuclear criticality. The reverse is true for small packages. Controlling radionuclide releases may become more challenging than control of criticality and package temperature. MPC backfills can address multiple repository concerns, but an emphasis on only one characteristic of the backfill will likely degrade capabilities in other areas.

There are also trade-offs in the choice of glass or uranium oxide. A glass optimized for low radionuclide release rate from the waste package may have different properties than a glass optimized for avoidance of nuclear criticality. To optimize avoidance of nuclear criticality, the glass should have dissolution kinetics similar to SNF. To minimize radionuclide release rates, the DUS glass may be chosen to have faster dissolution kinetics than the SNF UO_2.

5. MANAGEMENT OF DU

At uranium enrichment plants in the United States, there are about 4×10^5 t of DU, which is a by-product of nuclear fuel enrichment processes and weapons production. For LWR fuel production, natural uranium with a [235]U content of 0.71% is separated into a DU fraction and an enriched

uranium fraction. The enriched uranium (3-4% ^{235}U) is fabricated into fuel. Typically, 4 to 6 tons of DU are produced per ton of enriched uranium nuclear fuel with a fissile content of 0.2-0.35 wt. % ^{235}U (Hertzler, Nishimoto, and Otis, May 1994).

The U.S. Department of Energy has responsibility for management of this material and is examining options for its beneficial use. If this material is declared a waste, existing regulatory requirements indicate that some type of geological disposal will be required (NRC, January 1994). Shallow land burial is unacceptable (NRC, September 22, 1992). The use of DU as a backfill material eliminates the cost of disposal of these materials. This provides a credit for use of DU package backfill material. Based on the analysis herein, a minimum of 25% of the inventory would be required to implement DUSCOBS—assuming that the MPC is designed with minimum internal volume.

Many of the costs for management of DU are identical for any option. For example, in the United States the depleted uranium is primarily in the form of uranium hexafluoride. The uranium must be converted to the oxide form first before chemical conversion to glass. The glass production step from oxide is inexpensive; but, the conversion of UF_6 to uranium oxide is more costly. However, the conversion of depleted UF_6 to oxide form is required (NRC, September 22, 1992) for long-term safe storage or disposal by any route. In Europe, the depleted uranium is stored as uranium oxide thus this conversion step is not required for implementation of DUSCOBS.

If so desired, the MPC could be expanded to incorporate the total excess DU - about 4 to 6 t per metric ton of SNF. It is noted that the use of added DU would further improve repository performance both in terms of reduced release of radionuclides from SNF and reduced potential for nuclear criticality.

6. ECONOMICS

No economic assessment of this option has been conducted. There are economic benefits and costs. The potential areas for cost savings include improved repository performance, simplified and more robust repository licensing case, and avoidance of costs for disposal of DU. The costs are for those associated with implementation of DUSCOBS. Because (1) DUSCOBS uses empty space within the waste package that is now not used and (2) the disposal costs of DU may be high, there is a significant potential for systems cost savings.

7. UNCERTAINTIES

The preliminary analysis herein is based on literature sources and limited calculations. No experiments have been conducted. Such information sources can provide a scientific basis for consideration of an advanced backfill material but by themselves can not quantify how much repository performance may improve. Benefits are strongly dependent on specific design details

such as the glass composition, costs to implement, time to implement, and other factors. Detailed analysis and experimental work are required to address these issues.

8. CONCLUSIONS

The use of DUS glass as a backfill material for MPCs containing LWR SNF has multiple potential advantages. First, there is improved repository performance with the potential for simplified repository licensing and higher assurance of system performance. This is a consequence of (1) reduced radionuclide release rates from the waste package and (2) reduced potential for nuclear criticality in the repository. Second, this provides a method for disposition of excess DU.

Detailed analysis and experiments will be required to quantify these potential benefits, identify any liabilities, and costs.

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