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POTENTIAL BENEFITS AND IMPACTS ON THE CRWMS TRANSPORTATION SYSTEM OF FILLING SPENT FUEL SHIPPING CASKS WITH DEPLETED URANIUM SILICATE GLASS

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ABSTRACT

A new technology,[1] the Depleted Uranium Silicate COntainer Fill System (DUSCOFS), is proposed to improve the performance and reduce the uncertainties of geological disposal of spent nuclear fuel (SNF), thus reducing both radionuclide release rates from the waste package and the potential for repository nuclear criticality events. DUSCOFS may also provide benefits for SNF storage and transport if it is loaded into the container early in the waste management cycle. Assessments have been made of the benefits to be derived by placing depleted uranium silicate (DUS) glass into SNF containers for enhancing repository performance assessment and controlling criticality over geologic times in the repository. Also, the performance, benefits, and impacts which can be derived if the SNF is loaded into a multi-purpose canister with DUS glass at a reactor site have been assessed. The DUSCOFS concept and the benefits to the waste management cycle of implementing DUSCOFS early in the cycle are discussed in this paper.

I. INTRODUCTION

The U.S. Department of Energy's (DOEs) Office of Civilian Radioactive Waste Management (OCRWM) has been developing the Civilian Radioactive Waste Management System (CRWMS) for the disposal of commercial spent nuclear fuel (SNF). The CRWMS may eventually include (1) taking possession of the SNF at commercial reactors, (2) storing the SNF on an interim basis, (3) disposing of the SNF in waste packages at a geologic repository, and (4) transporting the SNF in U.S. Nuclear Regulatory Commission (NRC) certified casks.

One concept being given serious consideration by the OCRWM is to use a multi-purpose canister (MPC) as the inner container in transportation, storage, and disposal systems. As currently envisioned, the MPC would be loaded with SNF, drained of water, sealed, and filled with a gas early in the CRWMS waste management cycle. The loaded MPC would then be used, without its being reopened, for:

1. transport from the reactor to a storage system,
2. storage in storage casks or other systems,
3. transport from the storage system to the repository surface facilities,
4. loading into the waste package at the repository site and transfer of this loaded waste package to final emplacement in the repository, and
5. all handling action associated with these activities.

In each case, the MPC would be placed in another element of the waste management system (e.g., into a shipping cask for transport, into a storage cask of other storage system for storage, or into a waste package for disposal).

[a] Managed by Lockheed Martin Energy Research Corp. for the U.S. Department of Energy under contract DE-AC05-96OR22464.

Relative to the disposal of the SNF, one of the major problems which must be addressed is the need to ensure prevention of criticality in the repository over geologic times. Should the repository designer and those addressing the performance assessment of the repository be forced (a) to assume failure of the waste package and any internal containers (such as an MPC) and (b) to assume that water enters the damaged packages, ensuring criticality safety may be difficult.

Criticality safety has also been an issue in the design efforts, to date, for the storage and transport systems being considered by the OCRWM, including the concepts that have been developed for storing and transporting SNF in an MPC. Specifically, the need to enhance transportation systems performance relying on burnup credit has been one of the design issues which OCRWM has been addressing for a number of years. For transport and storage, designers must also be concerned with adequate shielding to protect workers and members of the public from exposure to excessive amounts of penetrating radiation.

To address these issues, a new technology,[1] the Depleted Uranium Silicate COntainer Fill System (DUSCOFS) has been conceived and is proposed for application in SNF waste management systems. It is noted that, initially, the concept was called Depleted Uranium Silicate Container Backfill System (DUSCOBS). However, it has been recognized that the DUS glass is to be used as a fill for the waste packages and not as a backfill around the waste packages. Thus, the concept was renamed.

The use of DUSCOFS could improve the performance and reduce the uncertainties of geological disposal of SNF by reducing both radionuclide release rates from the waste package and the potential for repository nuclear criticality events. DUSCOFS could also provide benefits for SNF storage and transport if the DUS glass is loaded into the container early in the waste management cycle. Preliminary assessments have been made of the potential benefits of DUSCOFS. The DUSCOFS concept is briefly introduced, and the potential benefits and impacts of DUSCOFS on the SNF waste management system are discussed.

Finally, the potential of beneficially using the large inventory of depleted uranium, currently stored in the United States as uranium hexafluoride (560,000 t are currently the responsibility of DOE) for this application are presented; if useful applications for this depleted uranium inventory are not found, then ultimately extensive and costly efforts will be needed to dispose of the inventory. A systems approach to resolving this problem, using DUSCOFS as one of a number of possible applications within the SNF and other radioactive material waste management systems, is briefly discussed.

The study reported in this paper was undertaken with the support of the DOE Environmental Restoration and Waste Management (EM) Office, which has been given responsibility for defining beneficial uses of the depleted uranium hexafluoride which is the responsibility of DOE. EM funded the evaluation because the DUSCOFS concept may provide a beneficial use for much of this inventory. The details of the results of these initial studies are published.[1]

II. MECHANICAL AND CHEMICAL DESCRIPTION

DUSCOFS was initially conceived to resolve the criticality issues for an SNF repository. To perform preliminary analytical assessments to determine whether benefits to the CRWMS repository performance assessment could result by enhanced criticality safety, it was necessary to assume characteristics of the DUS glass and that operationally a loaded MPC or other container with SNF could be filled with DUS glass. Specifically, the basic DUSCOFS concept relies upon loading the repository waste packages with SNF and then filling them with depleted uranium (0.2 wt % ^{235}U) silicate (DUS) glass beads. As conceived, the DUS glass beads would be sufficiently small (<1 mm) to be poured in and fill coolant channels in the SNF assemblies and other small volumes in and around the fuel basket support in the waste package. It has further been assumed that the glass can be readily manufactured to contain depleted uranium sufficient to lower the total fissile concentration in the waste package below 1 wt % heavy metal. Although it was initially assumed that the filling would occur at the repository before final disposal, it has been assumed that the filling of the container with DUS glass could also occur at the reactor site upon initial loading of the MPC with SNF elements or at a later stage in the waste management process such as at the storage. In both cases, additional benefit to the waste management system might accrue.

A high performance DUS glass has been chosen for the concept, where the glass could be tailored to have dissolution kinetics similar to SNF. Assessment of glass technologies indicates that the conversion of the depleted uranium from uranium hexafluoride to

DUS glass is feasible, and, once developed on a pilot basis, the technology could be readily transferred to the private sector.[2,3] A number of glass compositions are possible. For this study, three were chosen as examples: (1) Loffler glasses, (2) soda lime glasses, and (3) sodium silicate glasses. It is expected that these glasses can be readily produced having significant quantities of uranium loadings (30 to 50 wt % U_3O_8 equivalent). The average densities of these glasses should range between 3 and 5 g/cm^3. Ultimately, the preferred choice will depend upon complex trade-offs among different performance characteristics and economics.

III. REPOSITORY SYSTEM BENEFITS

A. Reduced Radionuclide Release

The addition of DUS glass offers the potential of reducing the long-term radionuclide release rate from the waste package in the repository. Most fission products and actinides in SNF are incorporated into uranium dioxide (UO_2) pellets. These fission products and actinides can not escape until the UO_2 dissolves or is transformed into other chemical species. It is postulated that the DUS glass in waste packages would saturate the groundwater in the waste package with uranium and thereby slow the SNF dissolution process. Uranium-saturated groundwater can not dissolve added SNF uranium.

The silicate in the glass would lower the solubility of uranium in groundwater; thus, more water must flow through a waste package to remove a unit quantity of uranium. This will delay the dissolution of the SNF UO_2. Uranium silicate also tends to form a coating on the UO_2 which acts as an additional barrier to the dissolution and transformation of UO_2.

Another potential benefit of the DUS glass is that as water enters the waste package, uranium oxides and silicates in oxidizing groundwater will eventually evolve into hydrated uranium silicate minerals. These silicate minerals have much lower densities than uranium oxides and glasses. Thus, the fill materials, once exposed to water entering the package, will expand and potentially seal the waste package from additional water flow.

All of the above mechanisms will work together to reduce the long-term radionuclide release rate from the waste package in the repository.

B. Reduced Potential For Repository Nuclear Criticality

DUSCOFS could reduce the potential for both package and zonal repository nuclear criticality events in a repository. In a repository, long-term, low-power nuclear criticality events are a major concern because they generate heat that (1) accelerates degradation of the waste packages and (2) accelerates water movement that can transport radionuclides to the environment.

Nuclear criticality in a repository is prevented by neutron absorbers and geometric spacing of fissile materials. Neutron absorbers include ^{238}U, boron, gadolinium, and other materials. Neutron absorbers (except ^{238}U) will leach from waste packages and travel at different rates through the geological media than the SNF uranium because of their different chemistries in groundwater. This in turn will create the potential for two types of repository criticality events if the fissile concentration in the repository SNF is sufficiently high:

Package Criticality. Large SNF waste packages contain inventories of fissile materials such that nuclear criticality may occur within the package if the neutron absorbers are preferentially leached from the waste package.

Zonal Criticality. Mechanisms that create natural uranium ore bodies—dissolution and precipitation of uranium—can concentrate uranium and separate fissile ^{235}U from all neutron poisons except ^{238}U. These mechanisms create the potential of zonal criticality events during which uranium from multiple-breached waste packages dissolves in groundwater, is transported some distance, and reprecipitates, thus forming large deposits of uranium.

The fissile content of light-water reactor SNF is sufficient to cause nuclear criticality by the previous mechanisms. This phenomenon can be demonstrated by comparing the expected fissile concentrations in the repository with fissile concentrations that have caused criticality in the past.

Fissile Concentrations. The average fissile content of SNF in the United States is 1.47 wt %. This average will increase in time because (1) higher burnup fuels with higher end-of-life fissile contents are now being used and (2) end-of-reactor-life SNF discharges with partially burnt SNF. Highly enriched SNF and excess plutonium (which decays to ^{235}U) may further increase enrichment levels. The ultimate average fissile enrichments in the repository may approach 1.7 wt %.

Conditions for Nuclear Criticality. In a homogeneous environment,[4] criticality may occur at ^{235}U equivalent enrichment levels as low as 1 wt %. Natural reactors occurred on the earth two billion years ago in the Oklo mining district of Gabon, Africa. When these natural reactors shut down, uranium enrichments[5] were as low as 1.3 wt %. French studies[6] of natural reactors indicate that such phenomena may occur at enrichments as low as 1.3% and become likely when enrichments approach 1.7 wt %. A repository is a man-made uranium mine and is expected to behave in a similar fashion over geological times.

Whether nuclear criticality will, in fact, occur in a specific repository depends upon the evolution of the chemical conditions, over time. The potential for criticality exists with current designs. The addition of DUS glass as an SNF waste package fill lowers the uranium enrichment in each waste package and, consequently, in the repository as a whole to a level below 1 wt %. Nuclear criticality can not occur with such low uranium enrichment levels in a geological environment. The depleted uranium and the SNF uranium have the same aqueous uranium chemistry. Once an SNF waste package degrades and water enters the package, chemical isotopic exchange, over time, mixes the SNF uranium with the depleted DUSCOFS uranium. Uranium-238 is the only neutron absorber that does not separate from ^{235}U over geological time frames.

IV. TRANSPORTATION SYSTEM BENEFITS

To fully assess the viability of the DUSCOFS concept, it was desirable to better understand the system implications including benefits and impacts of filling an MPC at the reactor site, and having the DUS glass present during all further handling, transport, and storage activities preceding emplacement in a repository. It has been assumed that the reactor operators would be capable of adequately drying and then filling the casks with the DUS glass and that they would be able to handle the casks should they be slightly heavier than the conceptual design cask,[7] that is, that the systems impacts of performing these tasks would not be unacceptable.

A. Analyses

A preliminary set of analyses was performed to determine whether criticality, burnup credit, shielding, and heat transfer in the storage and transport environments would be positively or negatively impacted by inserting the DUS glass during MPC loading at the reactor.

The studies undertaken considered the MPC and transportation cask concept developed by the DOE in 1994.[7] Efforts were not made to adapt these designs, to evaluate structural changes which would be needed, or to initiate new designs. The concepts were used only to provide a basis for making top-level estimates of system performance and defining general impacts on the system if the DUSCOFS concept were used with the MPC concept in transport and disposal. Various properties of DUS glass are possible, and if the concept is pursued, optimization of the glass used will be necessary. The properties of the glass were assumed as follows:

- 29.5 wt % U_3O_8,
  11.2 wt % CaO,
  7.4 wt % Na_2O, and
  51.9 wt % SiO_2;
- 0.2% depleted uranium (i.e., 0.0014 wt % ^{234}U, 0.2000 wt % ^{235}U, 0.0009 wt % ^{236}U, and 99.7977 wt % ^{238}U);
- DUS glass density = 4.1 g/cm^3, with an effective density of 2.7 g/cm^3 assuming 65% of space in the MPC is DUS glass beads; and
- Thermal conductivity (same as dry sand) = 0.33 W/m°C

B. Results of Analyses

Shielding, criticality, and heat transfer analyses, and weight trade-off studies were performed. From the brief analyses performed, the following preliminary conclusions were reached.

- Relative to criticality performance of the transport cask, loading the MPC with DUS glass at the reactor could eliminate all need to consider burnup credit in the design and certification and could significantly cut the costs associated with designing, certifying and operating casks for the CRWMS program.

- Relative to cask shielding, based upon the large MPC/cask concept, significant amounts of the gamma shielding could be removed from the cask body. It was estimated that all of the lead shielding and 15% of the depleted uranium shielding in the cylindrical portion of the cask body could be eliminated. Additional shielding might be eliminated from the ends of the casks, but further studies will be needed to verify this conjecture.

- Relative to heat transfer, adding DUS glass has a negative impact of virtually eliminating radiative and convective exchange of heat between fuel pins. However, the preliminary assessment showed that the loss of these heat transfer mechanisms will be approximately offset by a significant increase in the thermal conductivity between the fuel pins. As a result, the thermal performance of SNF in an MPC/cask configuration loaded with DUS glass is approximately the same as that for an MPC/cask configuration filled with a gas.

- Relative to the mass of the fill material, it was estimated that some of the added mass introduced by the DUS glass would be offset by the reduction in gamma shielding in the cask body.

For a design with fixed geometry and fixed number of SNF assemblies (the large conceptual MPC/cask), additional mass would result but the mass growth was projected not to be sufficient to eliminate the concept. The mass of the large MPC/cask system was projected to increase from 109 to 114 t (an increase of approximately 4,300 kg), an increase which was within the projected limit for the 125-t MPC/cask.

For a design in which the geometry is modified from that of the large, 125-t MPC/cask concept, the mass of the system was projected to decrease. In this case, in order to accommodate the added weight of the DUS glass, it was assumed that the MPC wall thickness would need to increase from 2.54 to 5.08 cm (from 1 to 2 in.) and that the number of assemblies loaded into the MPC would decrease from 21 to 17 [pressurized water reactor (PWR) assemblies] and from 40 to 32 [boiling water reactor (BWR) assemblies]. On this basis it was projected that:

1. although the loaded weight of the MPC would increase slightly, the overall MPC/cask system weight would decrease by approximately 10 t (8,809 kg);
2. the number of shipments would increase by approximately 24% (from about 7,211 to 8,954); and
3. the amount of depleted uranium utilized would increase from about 110,000 t to 147,000 t (about 99,800 to 133,400 tonne).

Thus, a number of subsidiary benefits could be derived from filling the void spaces in an MPC with DUS glass for controlling criticality in the repository and improving repository performance assessment if (a) the glass is added at the reactor site and (b) the system is designed with that in mind.

V. SYSTEMS APPROACH TO DEPLETED URANIUM DISPOSITION

For the proposed Yucca Mountain repository, a minimum of about 100,000 t of depleted uranium at 0.2 wt % ^{235}U is required for DUSCOFS. There are about 560,000 t of uranium hexafluoride, resulting in more than 400,000 t of depleted uranium in the United States for which DOE has responsibility. In addition, the U.S. inventory continues to grow as a result of the United States Enrichment Corporation's uranium enrichment activities. The current and future inventory of depleted uranium has limited uses. The NRC has previously stated that some type of deep disposal of this material will be required if it is declared a waste.

Using DUS glass may provide an economic and beneficial method for its disposal. Indeed, this application can be viewed as one of a number of potential beneficial reuse options available to DOE. In addition to utilizing the depleted uranium as fill material (as described above for the DUSCOFS concept), the depleted uranium can also be used (a) both in the form of oxides [e.g., depleted uranium oxide concrete[8] (DUCRETE)] for storage cask and storage system shielding, and potentially as shielding material in single-use, low-level waste shipment/disposal packages and (b) as metal for shielding in the lids of MPCs and in the lids and bodies of SNF casks for both storage and transport. It is speculated that, if this multiple application, system approach to the beneficial reuse of depleted uranium is followed, potentially all of the current and possibly the future inventory of depleted uranium can be beneficially reused, thereby solving SNF and other radioactive material disposal problems.

VI. ADDITIONAL STUDIES NEEDED

It must be emphasized that should the DUSCOFS concept be pursued seriously, studies of many factors will be needed to ensure important systems issues are adequately addressed. These include:

- developing and demonstrating the ability to produce the DUS glass;
- performing leaching tests on the DUS glass;
- defining a preferred method for loading the DUS glass into MPCs after they have been loaded with SNF assemblies;
- performing design alternative studies and defining costs and benefits of the various alternatives, including assessments of MPC, transportation cask, storage cask, and waste package alternatives; and
- assessing trade-offs for and defining systems and interfaces for applying the DUSCOFS concept to the CRWMS.

VII. CONCLUSIONS

Use of DUS glass has the potential to (1) reduce radionuclide release rates from waste packages, (2) reduce criticality events in the repository, (3) solve current burnup credit problems associated with SNF cask designs for the CRWMS, and (4) dispose of significant quantities of excess depleted uranium. This is a new concept and further studies are required.

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