Scenarios for the Evaluation of the Criticality Potential of High Actinide Glasses

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SCENARIOS FOR THE EVALUATION OF THE CRITICALITY POTENTIAL OF HIGH ACTINIDE GLASSES

INTRODUCTION AND SUMMARY

Vitrification is one of the leading options for immobilization of actinide-containing materials no longer needed for national defense. For these glasses to be suitable for disposal, it must be established that a significant potential for a nuclear criticality involving these glasses does not exist.

The vitrification working group within the nuclear materials disposition program has been given the responsibility for developing scenarios to be evaluated. In this report, potential bounding scenarios for disposal of high actinide glasses in a geologic setting are described. These scenarios are being provided to the Department of Energy's Office of Civilian Radioactive Waste Management (OCRWM) so that the potential for criticality can be evaluated. If the evaluation of these scenarios by OCRWM reveals a significant potential for criticality then a sensitivity analysis to numerical values should be used to determine whether more precise definitions of any parameter is warranted. It is anticipated that there will need to be extensive interactions between the working group and the personnel performing the criticality evaluations.

INITIAL CONDITIONS AND ASSUMPTIONS

The scenarios described below are all based on a common set of initial conditions and assumptions. The first is that the actinide of interest at the time of production of the glass is Pu-239. Pu-239 is mixed with nuclear poisons and melted to produce a glass in which the plutonium and any nuclear poisons added to prevent criticality are uniformly distributed. The molten glass is poured into a 304L stainless steel canister of the type used by the Defense Waste Processing Facility (DWPF), see Attachment 1. It is then welded closed. It is assumed that the glass has been formulated so that criticality of a semi-infinite array of canisters is not credible.

For all of the scenarios presented, the release of the actinides and the neutron poisons in the glass is initiated by reaction between the glass and groundwater. This presupposes that a crack has formed in any overpack placed around the DWPF canister by the repository operator, and that a crack has also formed in the DWPF canister itself. Thus, the time required before the reaction between the glass and the groundwater begins to occur is very long, most likely after the thermal pulse due to radioactive decay has begun to decay (possibly several thousand years - it is the responsibility of the OCRWM personnel to determine the time at which reaction begins). Thus, the decay of Pu-239 to its fissile daughter, U-235, must also be taken into account.

It is assumed that the glass dissolves congruently in the groundwater. Some elements, such as boron, remain in solution, and are transported away from the canister with the groundwater. Other elements, such as Pu-239, U-235, and rare earth elements, precipitate to form sparingly soluble hydrous oxides or silicate species. It should be assumed that groundwater leaving the canister will contain these sparingly soluble species at least at their saturation concentrations. These species may also be transported out of the canister as colloidal material. The reaction between the glass and the groundwater will depend on the composition of the water (dissolved solids, pH, redox potential), the amount of water, the composition of the glass, the surface area of the glass, and the temperature. Since the rates of reaction are relatively low, a significant portion of the Pu-239 will have decayed to U-235 before the glass is completely reacted. Thus, again, the properties of both Pu-239 and U-235 must be considered. This is especially important because scoping calculations performed by the group indicate that if uranium contacts reductants in the waste package, it may precipitate in a relatively concentrated form.

Table 1 contains a matrix of initial conditions and assumptions for use in evaluating the scenarios.

Each of these parameters, and the basis for their selection, is described below. The vitrification working group has provided only those properties specific to the glass. The working group assumes that OCRWM will use appropriate computer codes, and values of important parameters such as saturation concentrations, consistent with other OCRWM efforts. The working group can provide additional assistance to OCRWM in these efforts if necessary.

Canister Contents

The reference amount of Pu-239 oxide in the high actinide glass is 10 wt%. The compositions of two glasses currently being tested are given in Table 2. Thus, each canister will initially contain 170 kg of PuO_2 (assumed to be Pu-239). Again, it should be assumed that the actinides and the neutron poisons are uniformly distributed throughout the glass. Each DWPF canister should be assumed to contain 1700 kg of glass in a cylinder 2 feet in diameter.

It should be assumed that the glass produced is not a monolith. The difference in the coefficients of thermal expansion of the glass and the 304L stainless steel canisters leads to cracking of HLW glass which significantly increases the surface area compared to that of a monolith. Data developed by Savannah River Technology Center indicates that not all of this additional surface area is involved in the dissolution process, at least initially. However, as the glass corrosion process proceeds, more of the internal surface area is likely to be exposed. For this reason, two surface areas are provided: 5 m^2, which corresponds to the surface area of a HLW glass monolith; and 100 m^2, which corresponds to the maximum surface area which has been reported for HLW canisters.

Once the canister is breached, and water enters, the amount of water in direct contact with the glass will be limited to that within the canister. For DWPF glass, the volume of void space above the glass in the canister is nominally 165 L. However, some of these high actinide glasses are at least twice as dense as DWPF glass. For this reason, two volumes of canister void space (assumed to equal the volume of water in direct contact with the glass) have been provided - 165 L and 330 L.

Glass Dissolution Rates

As noted above, the rate of glass dissolution will depend on the glass composition, the surface area of the glass, the groundwater composition, the volume of water in contact with the glass, and the temperature. For natural groundwaters of the type expected in an underground repository (e.g., at Yucca Mountain), the glass dissolution rates in deionized water will bound those in groundwater. The rates recommended for use in this document are primarily based on experiments performed at 90°C. Thus, their use presupposes that the thermal pulse in the repository has passed. They should overestimate the rate of glass-groundwater reaction at lower temperatures.

Two values are give in Table 1 for glass dissolution rates — 0.0001 and 0.1 g/(m^2·d). The lower value is 1% of that found for typical DWPF glasses, but has been measured for the first composition in Table 2 (containing actinide simulants).[1] The upper value corresponds to a rate which might be seen for a typical DWPF glass in a 28 day MCC-1 type test, and thus is probably an overestimate of the long-term dissolution rate.

SCENARIOS

The high actinide glass will contain neutron poisons to ensure safety during production and interim storage. These poisons, which are uniformly distributed in the glass with the actinides, may be either soluble or insoluble in groundwater. Thus, the key question to be answered for safe disposal is whether a scenario exists in which the plutonium and the neutron poisons are separated as a result of reaction of the glass with groundwater resulting in a criticality event. Criticality can

only occur if the scenario leads to the accumulation of plutonium to critical levels, in a geometry favorable for criticality. If such a scenario is credible, then a means to prevent this scenario from occurring must be found, such as changing the glass composition or the waste package design.

In the following scenarios, processes which will separate the actinides from neutron poisons in the glass are discussed. The numerical values assumed for the various processes are believed to be generally biased toward those more favorable for causing a criticality. However, some of these parameters are not precisely known. If, after evaluation, one of these scenarios has a significant criticality potential, an analysis of the sensitivity of the criticality to the numerical values should be used to determine whether more precise definitions of numerical values is warranted. Alternatively, the vitrification working group may elect to refine the scenario so that it is not as conservative.

In the scenarios which follow, it is assumed that the species of interest are the fissile materials (Pu-239 and its daughter, U-235), and the neutron poisons (boron and the rare earth elements). It is further assumed that boron from the dissolving glass is soluble, and transported away from the glass with the groundwater. The scenarios are differentiated by the assumed behavior of plutonium, uranium and the rare earth elements.

Scenario 1

In the first scenario, it is assumed that separation of actinides and rare earth poisons occurs only because of the difference in solubilities. The actinides and rare earth elements are transported away from the canister in groundwater which is saturated with each of them. Based on the solubilities of plutonium and the rare earth hydrous oxides, the proportion of each of these in the groundwater will be different from that in the glass, thus providing a mechanism for separation. Since the solubility of plutonium will differ from that of uranium, decay during transport also needs to be considered. As the actinides are transported away from the canister, dilution and dispersion effects must be considered. The potential for a criticality event should be evaluated for both the actinides transported away from the canister, as well that portion which remains behind in the canister.

Scenario 2

This scenario adds an additional separation mechanism to the first scenario. Once the saturated groundwater leaves the canister, it will experience a further "chromatographic" effect due to differential sorption of the rare earths and the actinides. This may affect the potential for criticality of actinides which are transported away from the canister, but should not affect those remaining behind in the canister. As the actinides are transported away from the canister, dilution and dispersion effects must also be considered.

Scenario 3

In this scenario, the actinide is transported away from the canister as a colloid. Thus, the groundwater leaving the canister is both saturated with actinide and rare earths as well as containing the rest of the actinide as a colloidal sol. The rare earths are present in the groundwater only at their saturation concentrations. In this case, there is no need to evaluate the criticality potential for material remaining in the canister, because there is no enrichment of actinide. As the actinides are transported away from the canister, dilution and dispersion effects must again be considered.

Scenario 4

In this scenario, the rare earth is transported away from the canister as a colloid. Thus, the

groundwater leaving the canister is both saturated with actinide and rare earths as well as containing the rest of the rare earth released from the glass as a colloidal sol. The rare earths are present in the groundwater only at their saturation concentrations. In this case, evaluation of the criticality potential of actinides remaining in the canister is necessary, because the amount of poison remaining is significantly depleted.

TABLE 1
INITIAL CONDITIONS FOR CRITICALITY SCENARIOS

Condition	Value(s)
Plutonium loading of glass	10 wt%
Glass composition	See Table 2
Glass content of canister	1700 kg
Glass surface area	5 or 100 m2
Glass dissolution rate	0.0001 g glass/(m^2·d) or 0.1 g glass/(m^2·d)

TABLE 2
COMPOSITIONS OF HIGH ACTINIDE GLASS

GLASS 1

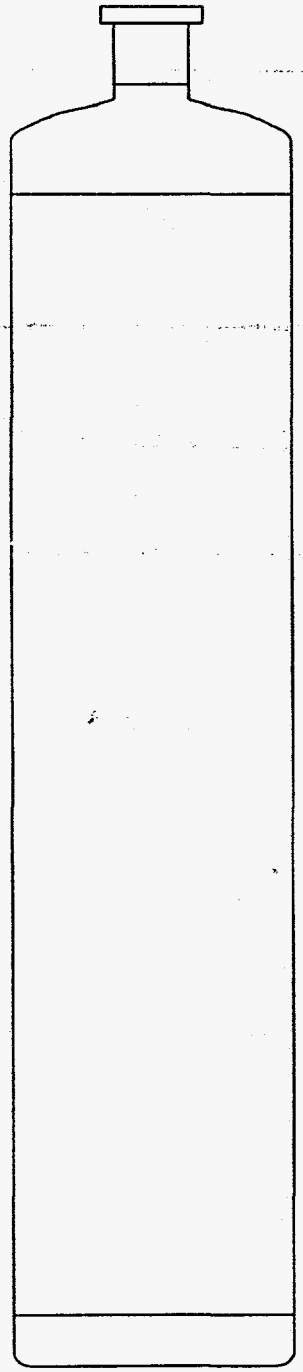
COMPONENT	AMOUNT (wt %)
SiO_2	31.4
B_2O_3	5.5
BaO	3.2
Al_2O_3	9.9
ZrO_2	0.2
PbO	13.6
La_2O_3	8.5
Sm_2O_3	17.7
PuO_2	10.0

GLASS 2

COMPONENT	AMOUNT (wt %)
SiO_2	44.6
B_2O_3	11.7
Li_2O	3.9
Na_2O	8.8
K_2O	5.0
Cs_2O	0.9
Al_2O_3	2.2
Gd_2O_3	3.2
ZrO_2	5.0
SnO_2	2.6
TiO_2	2.0
PuO_2	10.0

Figure 1. DWPF canister dimensions

Figure 1. DWPF canister dimensions



	Average	Std.Dev.
Overall length	117.98 in (299.67 cm)	0.03 in (0.08 cm)
Top end diameter	23.97 in (60.88 cm)	0.02 in (0.051 cm)
Top diameter	24.02 in (61.01 cm)	0.02 in (0.05 cm)
Middle diameter	24.04 in (61.06 cm)	0.03 in (0.08 cm)
Bottom diameter	24.04 in (61.06 cm)	0.02 in (0.05 cm)
Bottom end diameter	24.02 in (61.01 cm)	0.03 in (0.08 cm)
Flange tilt from horizontal	0.31°	0.19°
Upper head mis-match	-0.07 in (-0.178 cm)	0.09 in (0.23 cm)
Lower head mis-match	-0.03 in (-0.08 cm)	0.02 in (0..05 cm)
Bow (middle)	0.007 in (0.02 cm)	0.080 in (0.20 cm)
maximum bow (middle) - -0.141 in (-0.358 cm)		
Bow (top)	-0.008 in (-0.020 cm)	0.09 in (0.22 cm)
maximum bow (top) - 0.172 in (0.437 cm)		

Change in diameter after filling*:	Maximum	Std.Dev.
Top end diameter	0.110 in (0.279 cm)	0.037 in (0.094 cm)
Top diameter	0.053 (0.135 cm)	0.022 (0.056 cm)
Middle diameter	0.057 (0.145 cm)	0.027 (0.067 cm)
Bottom diameter	0.060 (0.152 cm)	0.022 (0.056 cm)
Bottom end diameter	0.161 (0.409 cm)	0.034 (0.086 cm)

* measurements from six canisters

REFERENCES

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