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SIMULATION USED TO QUALIFY NUCLEAR WASTE GLASS FOR DISPOSAL

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ABSTRACT

A hypothetical vitrification system was simulated to provide quantitative estimates of the errors associated with controlling and predicting the composition of the nuclear waste glass produced in the system. The composition of the glass must fall within certain limits in order to qualify for permanent geologic disposal. The estimated error in predicting the concentrations of various constituents in the glass was 2% to 8%, depending on the strategy for sampling and analyzing the feed and on the assumed magnitudes of the process uncertainties. The estimated error in controlling the glass composition was 2% to 9%, depending on the strategy for sampling and analyzing the waste and on the assumed magnitudes of the uncertainties. This work demonstrates that simulation techniques can be used to assist in qualifying nuclear waste glass for disposal.

1. INTRODUCTION

In 1982, the United States Congress passed the Nuclear Waste Policy Act, assigning the U.S. Department of Energy (DOE) responsibility for permanently disposing of existing and future high-level nuclear wastes generated in the U.S. That same year, DOE assigned responsibility for developing technology for the treatment of civilian high-level radioactive wastes generated during the reprocessing of commercial nuclear fuel to the Nuclear Waste Treatment Program (NWTP) at Pacific Northwest Laboratory (PNL), which is operated for DOE by Battelle Memorial Institute. The reference process for treatment of this waste is to incorporate it into a glass matrix and bury it in a deep geologic repository.

In the vitrification process, the wastes are combined with appropriate glass-forming compounds, and the resulting slurry is fed to a ceramic-lined melter operated at 1100 to 1250°C. Water from the slurry boils off rapidly, and the remaining components gradually melt into a molten glass phase. The glass is poured from the melter into canisters, where it cools and solidifies, immobilizing the waste. The canisters will eventually be sealed and shipped to a geologic repository for permanent disposal.

The ability to control and to predict glass composition and to estimate the uncertainty in the composition is important because there are constraints on 1) the range of compositions that can be processed in a ceramic-lined melter and 2) the range of compositions acceptable for geologic disposal. The first set of constraints arises because glass composition affects key processing parameters such as melting point, melting rate, viscosity, and electrical conductivity of the glass. The second set arises because composition is an important parameter affecting durability of the glass against aqueous attack, a key measure of acceptability of the glass for disposal. Because of the relationship between durability and composition, nuclear waste glass producers will be required to qualify their glass for disposal by providing documented assurance that the glass falls within certain composition limits. Waste producers would prefer to provide this assurance without excessively sampling the radioactive glass, which poses difficulties.

2. THE VITRIFICATION SYSTEM

Because no commercial-scale nuclear waste vitrification systems are yet operating in the U.S., a hypothetical system (Figure 1) was chosen for demonstrating the applicability of simulation techniques to waste qualification (Reimus et al. 1986; Kuhn et al. 1987). The flow diagram, stream compositions, and process flow rates for the system were adapted from preliminary flowsheets for proposed vitrification plants. The system has five major tanks: the waste lag storage tank (WLST), waste concentration tank (WCT), feed preparation tank (FPT), melter feed tank (MFT), and the melter. The first four tanks are assumed to be well-agitated and thus ideally mixed. The melter, which is resistance-heated by passing electrical current through the glass, contains two distinct regions: the cold cap and the molten glass. The cold cap is a thin layer of unmelted material formed on the surface of the molten glass as slurry is fed to the melter. Both the cold cap and the glass are assumed to be ideally mixed. This assumption has been partially substantiated in tracer studies in experimental melter systems at PNL.

Briefly, the flow of material through the hypothetical system is as follows (Reimus et al. 1986). Dilute waste slurry is periodically batched to the WLST. From there it is transferred to the WCT, where the slurry is concentrated by evaporation to less than one-fourth of the original volume. Once the slurry is adequately concentrated, it is transferred to the FPT, where glass formers are added and additional evaporation occurs. After the slurry is further concentrated, it is transferred to the MFT. Up to this point, all slurry transfers have been batch operations. The flow rate of feed from the MFT to the melter, however, is continuous as long as the glass level and the cold-cap coverage in the melter stay within control limits.

Samples are taken from two points in the system: the WLST and the FPT. The samples

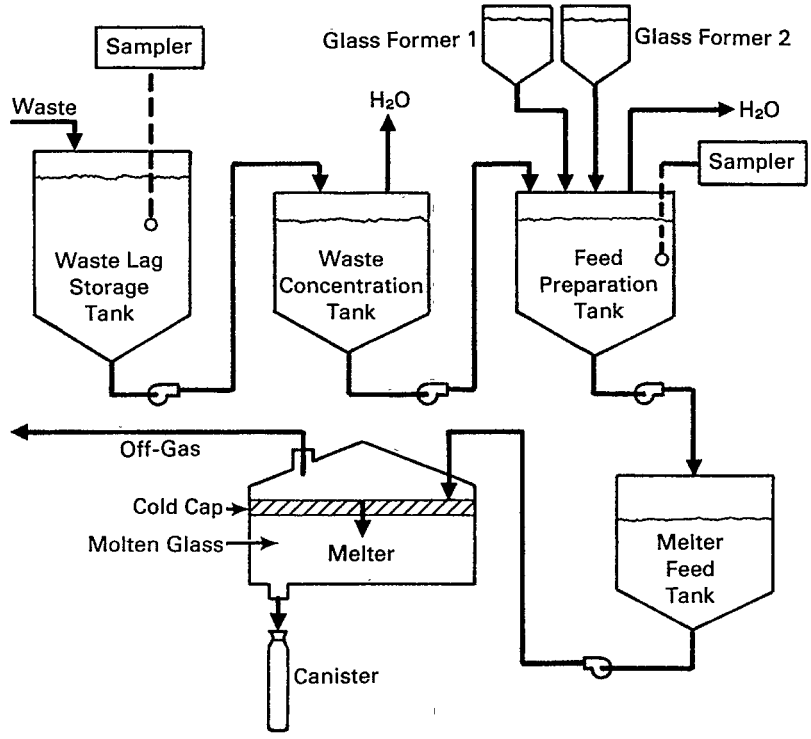

Figure 1: Schematic Flow Diagram of the Vitrification System

taken from the WLST are used to determine the amount of glass formers to add to the waste slurry in the FPT to produce glass having the target composition. The samples taken from the FPT (after the addition of glass formers) are used to predict the composition of the glass poured into the canisters.

In an actual vitrification system, there will be as many as 100 constituents in the glass. To simplify the demonstration, only the seven constituents listed in Table 1 were considered to be in the system (Reimus et al. 1986; Kuhn et al. 1987). All radioactive decay-heat-producing compounds in the waste were lumped into the constituent called "heat producers," all non-heat-producing compounds in the waste (other than water) were lumped into "other waste," and all glass-forming compounds other than Na_2O, B_2O_3, and SiO_2 were considered to be "other glass formers." The nominal compositions of the waste, glass formers, and glass assumed in the demonstration are listed in Table 1.

The use of two glass formers permits independent adjustment of decay heat loading and boron loading in the glass. Decay heat loading is often mentioned as an acceptance criterion for geologic disposal, and boron loading affects the melting rate of the cold cap and the chemical durability of the glass. As Table 1 indicates, one glass former is boron-rich while the other is boron-lean. A control algorithm was used to determine the amount of each glass former to add to each batch of waste.

3. PROPAGATION OF UNCERTAINTY

The ability to control and to predict the composition of the glass will depend on the magnitudes of the uncertainties in the vitrification system and the manner in which they propagate. The ability to control the glass composition (i.e., to add the correct amount of glass formers to the slurry in the FPT) will depend on the uncertainties associated with: 1) sampling and analyzing the contents of the WLST, 2) measurements of levels in the WCT and FPT, 3) the composition of the glass formers, and 4) the amount of glass formers added to the FPT compared with the amount the control algorithm prescribed. The ability to predict the glass composition will depend on the uncertainty associated with sampling and analyzing the contents of the FPT (after glass formers are added) and that associated with measurements of levels in the FPT, MFT, and melter.

Each process uncertainty in the system was defined in terms of its mean and a standard deviation (assuming that the uncertainties were described by normal distributions). The assumed magnitudes of the uncertainties are listed in Table 2 (Reimus et al. 1986). Except for the variability in the concentration of waste constituents from one waste batch to the next, all uncertainties represent measurement errors or errors in control actions. The variability in the concentration of waste constituents is an inherent process fluctuation that requires the control algorithm be used to keep the

Table 1: Compositions Considered in the Simulation (Mass Fractions)

Constituent	Waste	Glass Former 1	Glass Former 2	Glass
Heat Producers	0.00113	0.0	0.0	0.00844
Other Waste	0.03887	0.0	0.0	0.29
B_2O_3	0.0	0.012	0.092	0.0912
Na_2O	0.0	0.0446	0.0354	0.0703
SiO_2	0.0	0.2988	0.2372	0.47
Other Glass Formers	0.0	0.0446	0.0354	0.0703
Water	0.96	0.6	0.6	0.0

Table 2: Uncertainties in Process Variables

Variable	RSD†
Concentration of Waste Constituents from One Waste Batch to the Next	5%
Tank Level Measurements	1%‡
Concentrations of Constituents in Glass-Former Holding Tanks	2%
Amount of Glass Formers Added to FPT Compared to that Prescribed by Control Algorithm	2%
Sampling Error for Waste Constituents	5%
Analytical Error for Waste Constituents	5%
Sampling Error for Glass-Former Constituents	4%
Analytical Error for Glass-Former Constituents	4%

†Standard deviation relative to the mean (relative standard deviation).
‡ Percentage of full-scale reading.

glass composition as close as possible to the target. In Table 2 "sampling error" is the difference between the true concentration in a tank and the concentration in a sample from it. "Analytical error" refers to the difference between the true concentration in a sample and the apparent concentration as determined by an analytical procedure. The uncertainties listed in Table 2 do not necessarily reflect all uncertainties in vitrification systems but are thought to be the most important or largest ones. Also, the relative standard deviations defining the uncertainties are currently estimates for which there are few supporting data.

In principle, the relationships between the uncertainties associated with controlling or predicting the glass composition and the basic uncertainties in the process (i.e., those in Table 2) can be obtained by conducting a classical propagation of uncertainty analysis. In practice, however, it is difficult to obtain analytical expressions for the propagation of uncertainty because of 1) the combined discrete/continuous behavior of the system and 2) the interrelationships between some of the uncertainties. The classical approach, at best, would require many simplifying assumptions.

Simulation of the vitrification system offers a different method of conducting the propagation of uncertainty analysis. A stochastic simulation can provide data for directly estimating the confidence limits associated with controlling and predicting glass composition. This approach is conceptually straightforward and requires no simplifying assumptions.

4. THE SIMULATION

The SIMAN simulation language (Pegden 1985) was chosen to simulate the vitrification system because it can model simultaneously occurring discrete and continuous changes in the system. SIMAN can also incorporate stochastic variability into the simulation, an important requirement if the simulation is to be used for propagating uncertainties.

4.1 Continuous Aspects

The continuous changes in the state of the vitrification system are described by differential and state equations that correspond to mass balances around the tanks in the system. Assuming that the tanks are ideally mixed, the general equation for the rate of change of the total mass in any given tank is

$$dM/dt = \sum_{i=1}^{L} F_i - \sum_{j=1}^{N} G_j \qquad (1)$$

where M = total mass in tank
F_i = mass flow rate into tank from stream i
G_j = mass flow rate out of tank from stream j
L = number of streams flowing into tank
N = number of streams flowing out of tank

The equation for the rate of change of the mass of an individual constituent in a tank is

$$dM_k/dt = \sum_{i=1}^{L} F_i * X_{ki} - \sum_{j=1}^{N} G_j * X_{kj} \quad (2)$$

where M_k = mass of constituent k in tank
X_{ki} = mass fraction of constituent k in stream i

Since ideal mixing is assumed, the composition of any stream in the system is the same as that in the tank from which it originated. The mass fraction of a constituent in any given tank is calculated by dividing the mass of the constituent in the tank by the total mass in the tank,

$$X_k = M_k/M \quad (3)$$

Equations (1), (2), and (3) were written for each tank and each constituent in the system. The flow rates in all cases were either zero or some maximum value at various times during the simulation, depending on the conditions in the system. The conditions under which the flow rates were "turned on" or "turned off" are discussed in the next section. One exception to the on/off flow rate behavior was the flow rate from the cold cap to the molten glass in the melter; this actually corresponds to a melting rate rather than a controlled flow rate. This rate was assumed to be a function of both the boron mass fraction in the cold cap and the total mass of the cold cap:

$$\text{Melting rate} = K * X_{bc} * M_c \quad (4)$$

where K = constant
X_{bc} = mass fraction of B_2O_3 in the cold cap
M_c = mass of the cold cap

All differential equations in the system were simultaneously solved using the Runge-Kutta-Fehlberg algorithm included in the SIMAN subroutine library. This fourth-fifth-order method adjusts the size of each time step to satisfy truncation error criteria defined by the user. Since all differential equations in the system were time derivatives, the initial values of the state variables in the system had to be specified. The variables were always initialized to their target or design values.

4.2 Discrete Aspects

During operation of the system, many events occur at discrete points in time (i.e., discrete events). These events are triggered either by a condition on time or by a condition on one or more of the state variables in the system. An example of the former is the sampling of a tank one hour after filling. An example of the latter is turning on a pump to transfer slurry into a tank whose level has dropped below a certain limit.

Table 3 summarizes the discrete events built into the simulation and the conditions under which they were triggered (Reimus et al. 1986). Many of these events correspond to operational decision points or control actions in the system. The SIMAN framework permitted incorporating these events into the simulation with minimal programming effort.

4.3 Stochasm in the Simulation

The simulation was designed to calculate both the "true" and the "predicted" compositions of the glass poured from the melter as a function of time. The predicted composition was expected to differ from the true one because of uncertainties associated with process measurements. The true composition was expected to differ from the target composition because of uncertainties associated with control actions in the system. These uncertainties (see Table 2) were simulated using the stochastic features of SIMAN, which include a random number generator and algorithms for translating random numbers into representative values from statistical distributions. Every time a process measurement or control action was simulated, a normal distribution was sampled. The distribution had a mean equal to the true or target

Table 3: Discrete Events in the Simulation

Event	Condition(s)
Turn off flow of waste into WLST	Level in WLST exceeds 70,000 kg
Turn on flow of waste from WLST into WCT, start evaporation from FPT	Level in WCT drops below 2,000 kg
Turn off flow of waste from WLST into WCT, start evaporation from WCT	(1) Level in WCT exceeds 27,000 kg (2) Level in WLST drops to 2,000 kg
Stop evaporation from WCT	Level in WCT drops to 7,808 kg
Turn on flow of slurry from WCT into FPT, add glass formers to FPT	(1) Level in FPT drops below 2,000 kg (2) Level in WCT is at 7,808 kg
Stop evaporation from FPT, start sampling the FPT	Level in FPT drops to 12,467 kg
Turn on flow of slurry from FPT into MFT	(1) Level in MFT drops below 5,000 kg (2) FPT sampling is complete
Turn off flow of slurry from MFT into Melter	(1) Level in MFT drops below 2,000 kg (2) Mass of cold cap exceeds 500 kg (3) Mass in Melter exceeds 4,800 kg
Turn on flow of slurry from MFT into Melter	(1) Level in MFT rises above 2,050 kg (2) Mass of cold cap drops to 450 kg (3) Mass in Melter drops to 4,500 kg
Stop pouring glass into Canister	(1) Mass in Canister exceeds 1,500 kg (2) Mass in Melter drops to 2,800 kg
Start pouring glass into Canister	(1) It is 15.37 hr since last pour (2) Mass in Melter rises above 3,551 kg
Take sample from WLST	It is 1 hr after filling the WLST
Sampling of FPT is complete	It is 0.2 hr after sampling started
Analyses of WLST samples are complete	It is 24 hr after sampling WLST
Turn on flow of waste into WLST	Level in WCT exceeds 27,000 kg and the WLST was not filled the last time this happened
Calculate the amounts of glass formers 1 and 2 to add to the FPT	It is 0.1 hr after analyses of WLST samples were completed
Turn off flow of glass former 1 into FPT	The amount of glass former 1 prescribed by the glass-former control algorithm has been added
Turn off flow of glass former 2 into FPT	The amount of glass former 2 prescribed by the glass-former control algorithm has been added

value of the measured or controlled variable and a standard deviation equal to the value specified in Table 2. The value returned from the distribution was then substituted for either the true or target value of the variable in all subsequent calculations. This substitution, in effect, simulated the error in either the process measurement or control action.

Two sets of calculations were performed in parallel in the simulation: one for the "predicted" composition of the glass and the other for its "true" composition. The same equations were used in both sets of calculations; only the values in the calculations differed. In the calculations for the predicted composition, all values returned from the distributions describing

uncertainties in process measurements and control actions were substituted into the simulation. In the calculations for the true glass composition, only the values returned from the distributions describing uncertainties in control actions were substituted into the simulation. The predicted composition, therefore, reflected both measurement errors and errors in control actions, while the true composition reflected only errors in control actions.

5. RESULTS OF THE SIMULATION

The simulation was exercised many times to learn the effects of different strategies for sampling and analyzing the contents of the WLST and FPT on the ability to control and predict glass composition. Various sampling and analytical strategies are possible, ranging from a single analysis on a single sample to replicate analyses of replicate samples. The error in determining the composition in a tank was expected to decrease as both the number of samples and the number of analyses of each sample increased. Since the apparent composition in the WLST was used to determine the amount of glass formers to add to the FPT, the ability to control glass composition was expected to depend on the number of samples and the number of analyses of each sample taken from the WLST. Similarly, since the apparent composition in the FPT is used as the basis for predicting the glass composition, the ability to predict the composition was expected to depend on the number of samples and the number of analyses of each sample taken from the FPT.

The following sampling and analytical strategies were investigated:

Case 1: One sample and one analysis of both the WLST and the FPT.

Case 2: One sample and one analysis of the WLST, five samples and five analyses of each sample of the FPT.

Case 3: Five samples and five analyses of each sample of the WLST, one sample and one analysis of the FPT.

Case 4: Five samples and five analyses of each sample of both the WLST and the FPT.

For each case, a total of 40,000 hours of operation of the vitrification system was simulated. Results from one 4000-hour run representing case 1 and from a 4000-hour run representing case 4 are illustrated graphically in Figure 2 (Reimus et al. 1986; Kuhn et al. 1987). A quantitative measure of the ability to predict the glass composition in each case was provided by the 95th percentile of the absolute differences between the true and predicted glass compositions over the 40,000 hours (expressed as a percentage of the target composition). Similarly, a quantitative measure of the ability to control the glass composition was provided by the 95th percentile of the differences between the true and target compositions. The results are presented in Table 4 for the "heat producers" and B_2O_3 in the glass (the two controlled variables) (Reimus et al. 1986). This table shows that the ability to control glass composition depends primarily on the WLST sampling and analytical strategy, while the ability to predict glass composition depends primarily on the FPT sampling and analytical strategy. The values in Table 4 could be used to construct 95% confidence limits about both the true and predicted glass compositions.

6. CONCLUSIONS

By simulating the operation of a hypothetical vitrification system, it was possible to quantitatively estimate the errors associated with controlling and predicting the composition of the nuclear waste glass. Given the assumed magnitudes of the process uncertainties in the simulation, the 95th percentile of the absolute differences between the true and predicted compositions (relative to the target composition) ranged from roughly 2% to 8%, depending on the strategy for sampling and analyzing the feed. The 95th percentile of the absolute differences between the true and target

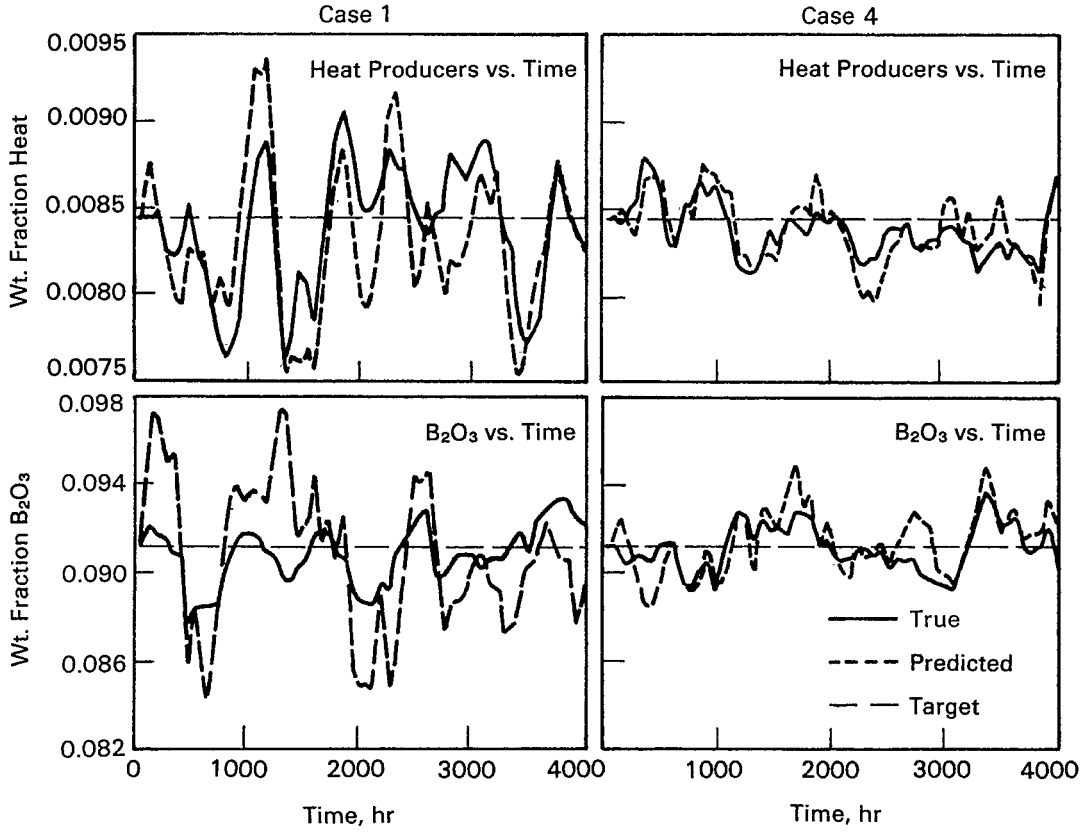

Figure 2: Comparison of Two Sampling/Analytical Strategies

Table 4: Comparisons of True, Predicted, and Target Glass Compositions

Constituent	Case	95th Percentile of TDiff†	95th Percentile of PDiff‡
Heat Producers	1	8.8§	7.7
	2	8.5	2.6
	3	4.3	7.6
	4	3.7	2.5
B_2O_3	1	3.3	6.0
	2	3.8	2.1
	3	2.3	6.1
	4	2.0	2.1

†Absolute difference between true and target compositions.
‡Absolute difference between true and predicted compositions.
§Percent of target composition.

compositions ranged from roughly 2% to 9%, depending on the strategy for sampling and analyzing the waste. The former percentile values provide a measure of the ability to predict glass composition, while the latter values provide a measure of the ability to control it.

The implication of this work for producers of nuclear waste glass is that they should be able to use simulation techniques to estimate both the composition of their glass and the uncertainty in its composition, assuming they can adequately characterize their process uncertainties. This approach would be preferable to conducting a classical propagation of uncertainty analysis, which is difficult even with simplifying assumptions, and it would minimize the need for directly sampling the radioactive glass.

Simulation techniques could also be used to 1) evaluate various strategies for controlling product quality in the vitrification process and 2) investigate the effects of the magnitudes of various uncertainties on the ability to control and predict the glass composition (i.e., a sensitivity analysis).

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REFERENCES

Reimus, P. W., et al. (1986). *Demonstration of an Approach to Waste Form Qualification Through Simulation of Liquid-Fed Ceramic Melter Process Operations.* PNL-5919, Pacific Northwest Laboratory, Richland, Washington.

Kuhn, W. L., et al. (1987). A method for qualifying canisters of vitrified high-level waste for disposal. In: *Proceedings of the ANS Topical Meeting on High-, Intermediate- and Low-Level Waste Management and Decontamination and Decommissioning* (J. M. Pope, ed.). American Nuclear Society Spectrum '86 meeting, Niagara Falls, New York.

Pegden, C. D. (1985). *Introduction to SIMAN.* Systems Modeling Corporation, State College, Pennsylvania.

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