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M. Hoza
G. I. Fann
D. F. Hopkins
C. A. LoPresti
G. F. Piepel
P. D. Whitney

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Pacific Northwest Laboratory
Richland, Washington 99352

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# OPTIMIZATION OF WASTE LOADING IN HIGH-LEVEL GLASS IN THE PRESENCE OF UNCERTAINTY

G.I. Fann, D.F. Hopkins, M. Hoza, C.A. Lo Presti, G.F. Piepel, and P.D. Whitney
Pacific Northwest Laboratory

## ABSTRACT

Hanford high-level liquid waste will be converted into a glass form for long-term storage. The glass must meet certain constraints on its composition and properties in order to have desired properties for processing (e.g., electrical conductivity, viscosity, and liquidus temperature) and acceptable durability for long-term storage. The Optimal Waste Loading (OWL) models, based on rigorous mathematical optimization techniques, have been developed to minimize the number of glass logs required and determine glass-former compositions that will produce a glass meeting all relevant constraints. There is considerable uncertainty in many of the models and data relevant to the formulation of high-level glass. In this paper, we discuss how we handle uncertainty in the glass property models and in the high-level waste composition to the vitrification process.

Glass property constraints used in optimization are inequalities that relate glass property models obtained by regression analysis of experimental data to numerical limits on property values. Therefore, these constraints are subject to uncertainty. The sampling distributions of the regression models are used to describe the uncertainties associated with the constraints. The optimization then accounts for these uncertainties by requiring the constraints to be satisfied within specified confidence limits.

The uncertainty in waste composition is handled using stochastic optimization. Given means and standard deviations of component masses in the high-level waste stream, distributions of possible values for each component are generated. A series of optimization runs is performed; the distribution of each waste component is sampled for each run. The resultant distribution of solutions is then statistically summarized.

The ability of OWL models to handle these forms of uncertainty make them very useful tools in designing and evaluating high-level waste glasses formulations.

## INTRODUCTION

In immobilizing high-level liquid waste for long-term storage, it is advantageous to produce the minimum volume of immobilized waste glass (for a given volume of waste), thus minimizing vitrification and disposal costs. The Optimal Waste Loading (OWL) models, based on rigorous mathematical optimization techniques, have been developed to determine the minimum number of glass logs required to immobilize Hanford high-level liquid waste. This optimization varies the glass-formers composition to maximize the waste loading (and minimize the glass volume), such that all processing and product (durability) constraints on the glass are satisfied.

Because there is uncertainty in both the property model predictions and in the waste feed composition for vitrification, the question naturally arises as to how the uncertainty affects the calculated waste loading and resultant number of logs required. In this paper we describe our approaches for addressing this question. We discuss techniques we are developing for addressing each of these uncertainties separately. As these techniques mature, we will integrate these and other techniques to handle all appropriate uncertainties.

In this paper, we briefly describe the general constrained optimization problem, which is the basis for the OWL model. We then present an overview of the OWL model formulation, followed by a discussion of the glass property models that are used in the constraints for optimization. We then describe our techniques for handling property model and waste composition uncertainties, and present some results for each. We end with some brief concluding comments.

THE GENERAL CONSTRAINED OPTIMIZATION PROBLEM

In the general constrained optimization problem, the objective is to find the multidimensional point $\boldsymbol{x}$ that produces the maximum (or minimum) value of some function f and meets a set of criteria called constraints. The problem would be stated as:

$$\begin{aligned} &\textit{maximize} && f(x) \\ &\textit{subject to} && \boldsymbol{h}(x) = 0 \\ & && \boldsymbol{g}(x) \le 0 \end{aligned}$$

where

- f is the objective function which we want to maximize (or minimize). It is a single (scalar) function of the unknown variables
- **x** is a vector $(x_1, x_2, \ldots, x_n)$ of the variables over which we optimize
- **h** is a vector function containing the equality constraints $(h_1(\mathbf{x}), h_2(\mathbf{x}), \ldots, h_p(\mathbf{x}))$
- **g** is a vector function containing the inequality constraints $(g_1(\mathbf{x}), g_2(\mathbf{x}), \ldots, g_q(\mathbf{x}))$

Each equality and inequality constraint is also a function of the unknowns. In general, the objective function and any or all constraints may be nonlinear.

GLASS FORMULATION OPTIMIZATION MODEL

In the simple glass formulation problem, frit (glass formers) is added to a single waste composition and the mass fraction of waste in the glass (waste loading) is maximized such that all the constraints are satisfied. The frit composition is varied as part of the optimization. The problem formulation is shown in Figure 1 and briefly described below. More detailed information on the model can be found in Hoza (1).

PLACE FIG. 1 HERE

The simple waste optimization problem can be generally stated as follows:

| | |
|---|---|
| minimize<br>or<br>maximize | *number of glass logs required*<br><br>*waste loading in the glass* |
| subject to | *mass balance constraints*<br>*property model component bounds*<br>*solubility constraints*<br>*glass property constraints* |

The objective function and constraints will be discussed in the next two sections.

Objective Function

The goal is to minimize the number of glass logs necessary to immobilize a waste of the specified composition. This can also be achieved by maximizing the waste loading (fraction of the glass that is waste).

Constraints

There are four classes of constraints in the model.

The first, the mass balance constraints, are equalities which define the relationships involved in the formation of glass from its components. These include an overall balance and component balances for all components.

The second, the property model component bounds, limit the range of the composition (mass fraction) values each component can have in the calculated glass composition. They reflect the composition region over which the glass properties were experimentally determined, as part of the Composition Variation Study (CVS) (2), and define a polyhedron in composition space that specifies the region over which the glass property models are considered valid. Compositions outside these limits will not necessarily produce unacceptable glasses. Rather, these compositions represent regions for which the glass property models must be extrapolated. Promising glasses outside these limits would have to be evaluated experimentally to determine their acceptability.

The third, the solubility constraints, limit the maximum value for the mass fraction of selected components ($Cr_2O_3$, F, $P_2O_5$, $SO_3$, and noble metals). They are intended to represent solubility limits for the specified components. These limits cover component species not included among the species used in the glass property models. Eventually these solubility limits will be replaced with thermodynamic calculations that predict insoluble species.

The fourth, the glass property constraints, utilize the glass property models developed in the CVS. These are discussed in the next section.

## GLASS PROPERTY MODELS

The glass property models are equations empirically fit to data, i.e., glass compositions and property values (viscosity, electrical conductivity, and durability in this work). Liquidus temperature models have also been developed but were not used in this work. The modeling approach and the calculation of uncertainty are addressed in the balance of this section.

Modeling of Properties

The property models are empirically fit linear and nonlinear (in composition) models. The models were developed as part of the Composition Variation Study (CVS) and are described in Hrma, Piepel, et al. (2). The CVS has been performed in five phases (CVS-I and CVS-II Phases 1-4). The models used in this work were based on data obtained through CVS-II, Phase 2.

The CVS used *statistical mixture experiment* design and *optimal experimental design* methods and software to select the glass compositions tested throughout the CVS. The glass composition region included is expected to contain glasses that might be made from various waste types expected to be processed at Hanford.

The model of each property is of the form

$$\ln(PropertyValue)=\sum_{i=1}^{10} b_i x_i+\sum_{i=1}^{10}\sum_{j\geq i} b_{ij} x_i x_j \qquad (1)$$

where $b_i$ and $b_{ij}$ are the coefficients of the first- and second-order terms, respectively; $x_i$ is the mass fraction of component i; and 10 is the number of components considered in the study. The components included in the models are $SiO_2$, $B_2O_3$, $Na_2O$, $Li_2O$, CaO, MgO, $Fe_2O_3$, $Al_2O_3$, $ZrO_2$, and Others, which accounts for all species other than the nine specifically included. For the linear property models, all $b_{ij}$ are zero.

The glass properties used in this work were viscosity, electrical conductivity, and durability (actually rate of release of boron) by either the Product Consistency Test (PCT) or Materials Characterization Center Test (MCC-1). The current version of OWL includes PCT Li and Na releases, and no longer uses MCC-1 releases.

Calculation of Uncertainty

Predictions made with a fitted property model are subject to uncertainty in the fitted model coefficients. The uncertainty results from the random errors in property values introduced during testing and measurement as well as minor lack-of-fit of the empirical model relative to the true relationship.

The uncertainty in a predicted property value for a given glass composition is defined as

$$Uncert=M[x^T Sx]^{0.5} \qquad (2)$$

where

M = multiplier, which is usually the upper 95th percentile of a t-distribution $[t_{.95}(n-p)]$, where n is the number of data points used to fit the model and p is the number of fitted parameters (coefficients) in the model
**x** = glass composition vector expanded in the form of the model
$\mathbf{x}^T$ = transpose of glass composition vector expanded in the form of the model
S = covariance matrix of the estimated parameters (coefficients)

For linear (first-order) property models, **x** is the usual glass composition vector. For nonlinear models, the vector is augmented by second-order terms. For example, if there are two second-order terms, $x_1^2$ and $x_2x_4$, the usual composition vector $(x_1, \ldots ,x_{10})$ becomes $(x_1, \ldots ,x_{10}, x_1^2, x_2x_4)$.

OPTIMIZATION WITH GLASS PROPERTY MODEL UNCERTAINTY

The method used to account for glass property model uncertainty in the glass optimization and results of optimization calculations with property model uncertainty are given in the next two sections.

Method

This model accounts for uncertainty in the glass property constraints by using uncertainty to narrow the feasible region determined by glass property models. This approach changes the form of the glass property constraint to

$$\ln(MinVal)+Uncert \le \sum_{i=1}^{10} b_i x_i + \sum_{i=1}^{10} \sum_{j \ge i} b_{ij} x_i x_j \le \ln(MaxVal)-Uncert \qquad (3)$$

When Uncert = 0, this constraint is the same as for the model that does not account for property model uncertainty. Figure 2 shows the effect on a ternary diagram (for a waste + frit + recycle mixture. The idea is the same for a waste + frit system, but a ternary diagram better helps visualize the concept). A single linear glass property constraint with upper and lower limits is shown on the figure. The regions in the triangle with dark shading are infeasible (the constraint cannot be satisfied in those regions). The unshaded region is feasible. The lightly shaded regions represent those compositions that become infeasible when property model uncertainty is considered. Alternately, it can be viewed as the shrinkage of the feasible region due to uncertainty. The shading around the glass composition point represents the uncertainty in the glass composition resulting from uncertainty in the waste composition. Methods for dealing with this uncertainty will be discussed in the section on waste composition uncertainty.

PLACE FIG. 2 HERE

Results

The effect of property model uncertainty on maximum waste loading was examined for four Hanford double shell tank waste types. Table I summarizes the results of calculations that explore this effect for two constraint sets.

PLACE TABLE 1 HERE (AT PAGE BREAK--FULL PAGE LANDSCAPE TABLE)

When the *full constraint set* is used (first and second rows in the Table I), there is no difference between the waste loading with uncertainty and the waste loading without uncertainty in the glass property models. This is not surprising. The uncertainty in the glass property constraints effectively tightens the glass property constraints, but not enough to make a difference. The binding constraint for each case is still the same as for the case without uncertainty, so the glass property constraints and their uncertainties are irrelevant (for these cases; this will not always be the result).

When *only the glass property constraints* (viscosity, electrical conductivity, and durability) are used (third and fourth rows in Table I), the following occurs:

- Waste loading is reduced. As expected, the uncertainty in the glass property constraints makes a difference. The percent reduction in waste loading as a result of considering the uncertainty is on the order of the uncertainty in the binding constraints.

- The uncertainty in the glass properties is much greater than it is for the full-constraint case. Because the calculated uncertainty is a function of where the point is located in composition space, this indicates that these points are in composition regions where less experimental data are available and may even be outside the experimental region. Examination of the glass compositions for these cases (which are not in the table) confirms this. Several of the component compositions are outside the upper and lower limits on the ten components (because those limits were dropped for these cases).

## OPTIMIZATION WITH WASTE COMPOSITION UNCERTAINTY

The method used to account for waste composition uncertainty in the glass optimization and results of optimization calculations with waste composition uncertainty are given in the next two sections.

### Method

The basic approach taken to address the optimization in the presence of waste composition uncertainty problem relies on the stochastic modeling method (3). Using this method, the strategy is to generate a large number of possible waste compositions based on the composition error structures, and for each of these, to generate a waste loading. The distribution of waste loadings can then be analyzed. The main steps in this method, as applied to this problem, are:

- Develop probability distributions for the masses in the high-level vitrification feed of each of the components followed in the OWL models, based on estimates of means and standard deviations. For this work, all mass distributions were assumed to be normal (Gaussian).
- Sample the above distributions and developing N waste composition input sets (mass fraction basis). Sampling the distributions provided masses for each of the species tracked. Latin Hypercube Sampling (4) was used because it provides better coverage of the composition distributions than simple random sampling with fewer samples. Given these masses and the total mass of the waste, the mass fractions of all species were determined and normalized to 1.000.
- Run the N waste composition sets through the OWL glass formulation model to calculate the optimal waste loadings for each waste composition set.
- Analyze the resulting distribution of waste loadings for the N input sets.

For this work, uncertainties in waste components were assumed to be statistically independent (i.e., uncorrelated). This is likely an unrealistic assumption, but knowledge of composition uncertainty correlations was insufficient to account for them in this work. Future efforts will account for them once they are adequately quantified.

### Results

The method described above takes distributions in the masses of all relevant species, performs a series of calculations, and produces a distribution of waste loadings. This section looks at how the waste loading distribution is related to the input distributions, and what one can conclude from the output distribution?

The relationship between the output distribution and the input distribution depends on the constraints--which constraint(s) is/are binding and whether the same constraint is binding for all cases or the binding constraint changes for different runs. The following situations are possible; they are listed in order of increasing complexity.

- The same single-component constraint is always binding.
- The same multiple-component constraint (e.g., durability) is always binding.

- The binding constraint is different for different runs.
- No feasible solution is possible for some runs.

The waste selected for the sample calculation represents the least complicated situation. For this case, the binding constraint was consistent over all generated waste composition sets. This binding constraint was the upper limit on a single waste component ($P_2O_5$). As expected for this case, the waste loading varied inversely with the mass fraction of $P_2O_5$. Because the upper bound on $P_2O_5$ was the binding constraint for optimization, higher concentration of $P_2O_5$ causes a lower maximum waste loading fraction (WLF).

What can one conclude for this single-component-limited case? If the generated waste component mass distributions reflect reality, and if **N** is set appropriately high, then the sample input sets are increasingly likely to cover the range of possible waste composition sets. Each WLF is the highest WLF that will produce glass meeting the property constraints for an input waste composition set. Therefore, the distribution of optimal WLFs represents the possible range of optimal WLFs given the uncertainty defined for the input high-level waste stream.

Figure 3 shows the distribution of the optimal WLF and the reverse cumulative distribution of the optimal WLF, with cumulative probability increasing as WLF decreases. The cumulative distribution can be interpreted as follows: for any WLF calculated by maximizing the WLF subject to constraints as per OWL optimization, the cumulative distribution represents the probability that that WLF can be achieved given the waste composition and its associated uncertainty and error structure. For example, if the WLF is 0.038, the probability of being able to achieve that WLF is 0.85.

PLACE FIG. 3 HERE

The above analysis was for the simplest case; the same single-component constraint is always binding. How would the results change for binding multiple-component constraints or for changing binding constraints? Subsequent work will have to examine this issue, but a cumulative distribution (as in Figure 3) could still be developed and used as described above.

<u>Issues</u>

Because this was a preliminary look at the application of stochastic modeling, many assumptions were made to simplify calculations. These assumptions, which are addressed below, will be revisited in future work.

- Two independent (and inconsistent) determinations of the total mass are available, the sum of the sampled masses and the measured mass (actually measured volume and density). Some technique to reconcile the two is needed.
- The distributions for each component were assumed to be independent. This is unlikely to be true for several reasons (e.g., relationships of components in frit, waste, and recycle; correlations in analytical uncertainties; and imposed correlations among component mass fractions because they must sum to one).
- The simple case examined had the same binding constraint for all N samplings. This will not generally be the case. Accounting for statistical dependence between components may also change binding constraints.

## CONCLUSIONS

The techniques presented here address the uncertainty in property models (which are used in specifying constraints in the optimization model) and in waste feed composition. The latter technique needs further development to address the issues identified. Combining the two techniques would allow formulation of glasses in the presence of both types of uncertainty.

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FIGURE CAPTIONS:

Fig. 1. Simple Waste Optimization Problem

Fig. 2. Uncertainty in Glass Property Models and Waste Composition

Fig. 3. Empirical Probability vs Optimal Waste Load Fraction

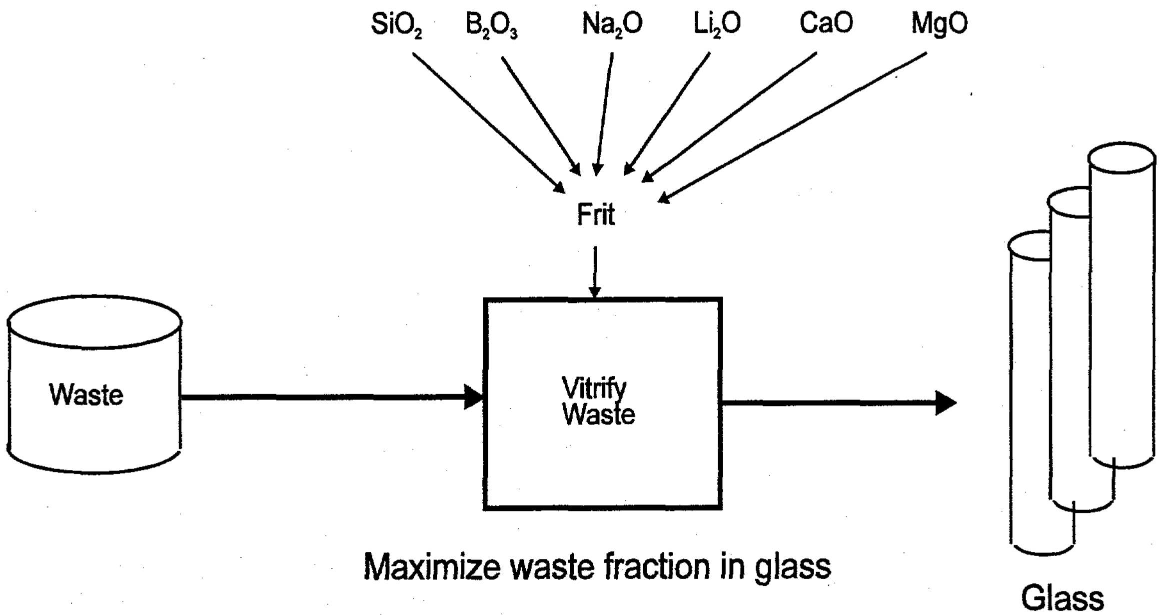


**Figure 1.** Simple Waste Optimization Problem

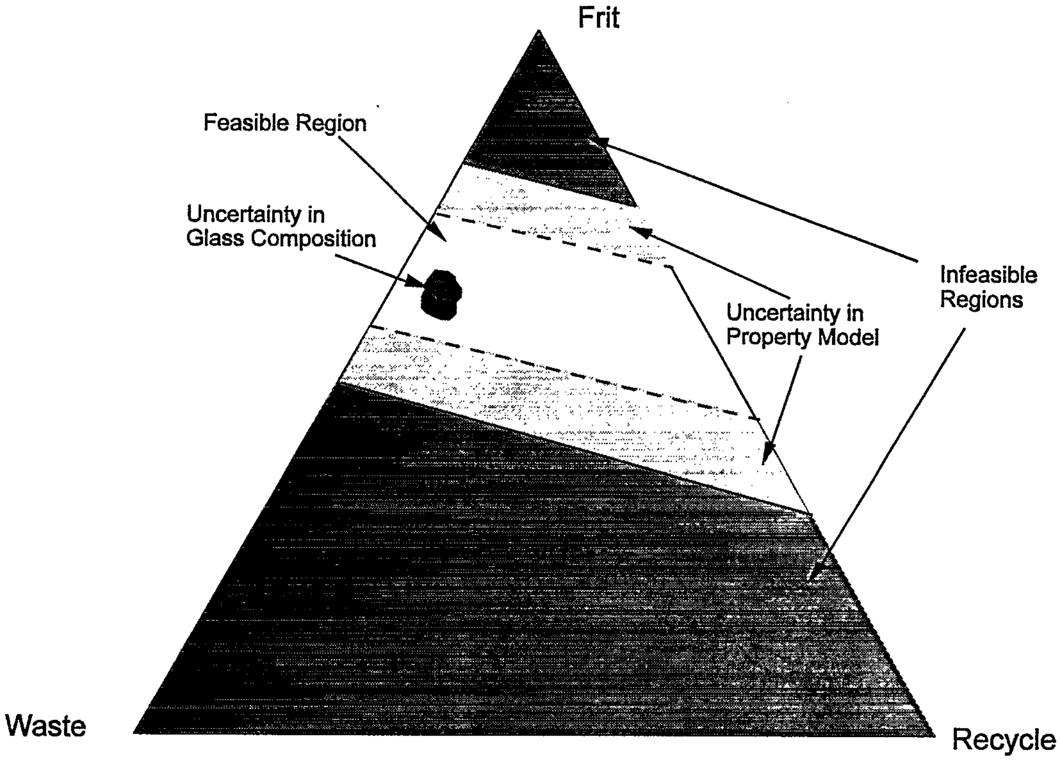


**Figure 2.** Uncertainty in Glass Property Models and Waste Composition

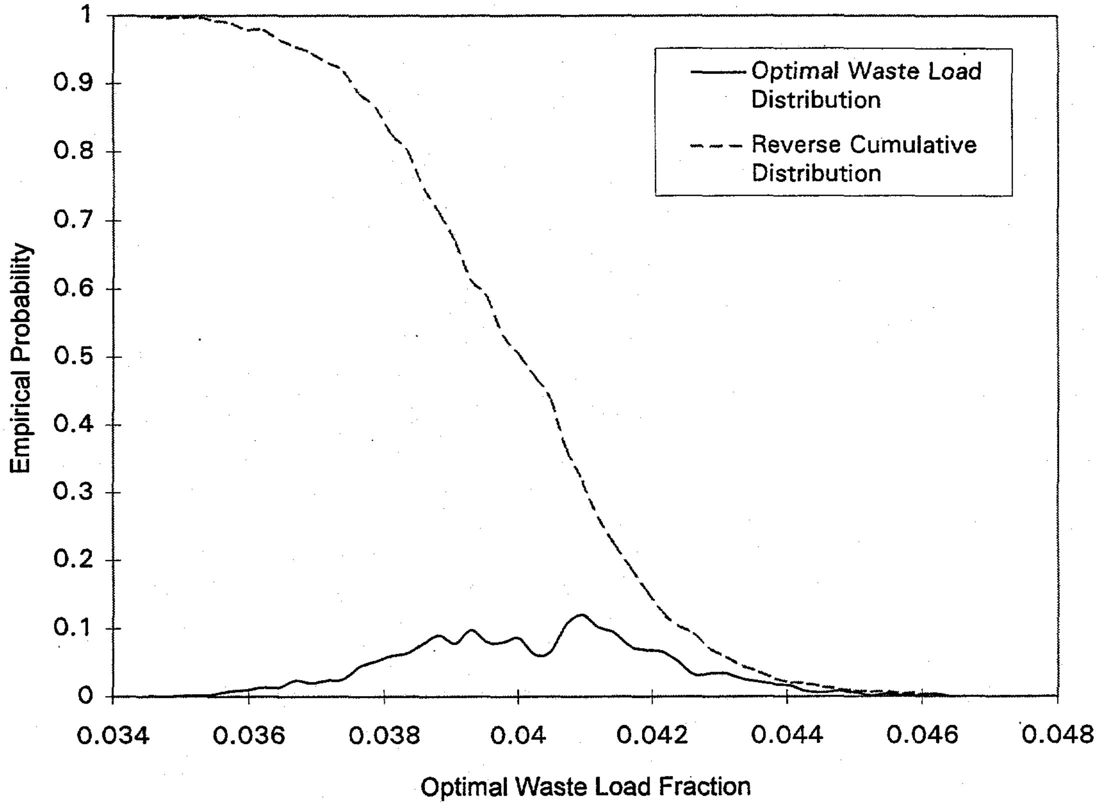


**Figure 3.** Empirical Probability vs Optimal Waste Load Fraction

**Table I.** Effects of Uncertainty on Waste Loading

| Waste -> <br> V Test Case | Key to table entries | NCAW | NCRW | PFP | CC |
|---|---|---|---|---|---|
| Full Constraint Set, WITHOUT uncertainty | waste loading | .306 | .195 | .121 | .196 |
| | binding constraint | UL Cryst3 | UL $P_2O_5$ | UL $Cr_2O_3$ | UL $SO_3$ |
| | visc, PaS | 6.18 | 2.89 | 2.0 | 2.00 |
| | e-cond, S/m | 22.1 | 41.4 | 39.0 | 44.5 |
| | dur-B-PCT, $g/m^2$ | 1.09 | 1.24 | 0.405 | 2.45 |
| | dur-B-MCC, $g/m^2$ | 14.4 | 13.5 | 8.23 | 17.0 |
| Full Constraint Set, WITH uncertainty | waste loading | .306 | .195 | .121 | .196 |
| | binding constraint | UL Cryst3 | UL $P_2O_5$ | UL $Cr_2O_3$ | UL $SO_3$ |
| | visc/uncert | 5.91/6.4% | 2.58/10.5% | 2.21/10.1% | 2.21/10.2% |
| | e-cond/uncert | 23.0/7.6% | 24.9/10.3% | 19.8/9.5% | 19.9/10.0% |
| | dur-B-PCT/uncert | 1.09/36.1% | 1.44/47.8% | 0.78/47.3% | 0.50/56.0% |
| | dur-B-MCC/uncert | 14.0/17.7% | 16.7/23.5% | 10.1/26.9% | 20.6/30.6% |
| Glass Property Constraints only, WITHOUT uncertainty | waste loading | .789 | .685 | .713 | .864 |
| | binding constraint | UL-DBM | LL-visc & e-cond | UL-visc & e-cond | UL-visc & e-cond |
| | visc | 2.39 | 2.0 | 10.0 | 10.0 |
| | e-cond | 24.0 | 18.0 | 50.0 | 50.0 |
| | dur-B-PCT | 3.69 | .07 | .0003 | .078 |
| | dur-B-MCC | 28.0 | 1.00 | .27 | 2.82 |
| Glass Property Constraints only, WITH uncertainty | waste ldg/% change | .660/-16.3% | .509/-25.7% | .674/-5.5% | .843/-2.4% |
| | bindg constr | LL e-cond, UL-DBM | LL & UL visc, LL e-cond | UL visc & e-cond | UL visc & e-cond |
| | visc/uncert | 4.74/27.1% | 4.47/80.5% | 6.89/37.2% | 8.62/14.9% |
| | e-cond/uncert | 22.6/22.9% | 22.7/23.0% | 34.5/37.2% | 44.2/12.4% |
| | dur-B-PCT/uncert | 1.64/118% | 0.05/122% | 0.003/229% | .077/64.5% |
| | dur-B-MCC/uncert | 15.1/62.0% | 1.66/71.7% | 0.40/119% | 3.17/33.6% |

KEY:

| Abbreviations | Constraints | | | |
|---|---|---|---|---|
| LL - Lower Limit | Viscosity: | $2 < visc < 10$ | Durability-Boron-PCT: | DBP < 10 |
| UL - Upper Limit | E-conductivity: | $18 < e\text{-}cond < 50$ | Durability-Boron-MCC: | DBM < 28 |