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Project Technical Information

Preliminary Feed Test Algorithm for the Hanford Waste Vitrification Plant Product Composition Control System

M.F. Bryan
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Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest National Laboratory
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SUMMARY

The Feed Test Algorithm (FTA) will test the acceptability (conformance with requirements) of process batches in the Hanford Waste Vitrification Plant (HWVP). Although requirements and constraints will be imposed on properties of the material in the melter and the resulting glass, the FTA must test acceptability while the batch is still in the Slurry Mix Evaporator (SME), i.e., *before* material is transferred to the Melter Feed Tank. Hence, some properties upon which requirements will be imposed must be estimated from data available on the feed slurry. The major type of data to be used in this estimation is feed composition, usually expressed in terms of nine oxide mass fractions and a catchall tenth category, Others.

Uncertainties are inherent in the HWVP process. The two major types of uncertainty are composition uncertainty (that related to measurement and estimation of feed composition and other quantities) and model uncertainty (uncertainty inherent in the models developed to relate melt/glass properties to feed composition). Types of uncertainties, representation of uncertainty, and a method for combining uncertainties are discussed.

The FTA must account for these uncertainties in testing acceptability; hence it must be statistical in nature. Three types of statistical intervals (confidence, prediction, and tolerance) are defined, and their roles in acceptance testing are discussed.

A *reference constraint set* containing the currently recognized requirements to be imposed on HWVP is identified. Aspects of requirements that affect the form of statistical acceptance tests are discussed. Three distinctions are used to identify the appropriate statistical method for each requirement in the reference constraint set:

- Direct constraints vs. constraints on modelled properties -- This distinction determines whether or not model uncertainty must be taken into account in statistical tests. Direct constraints are those that apply directly to measured quantities or to *known* functions thereof. For testing of direct constraints, only composition uncertainty is required.

For testing of constraints on modelled properties, an estimate of model uncertainty is also required.

- Single-batch constraints vs. multiple-batch constraints --This distinction determines the type of composition uncertainty required by the statistical test. Single-batch constraints are those that apply to single process batches; multiple-batch constraints apply over several batches (e.g., all batches within a single waste type). Testing of single-batch constraints requires the use of an estimate of within-batch variability, while testing of multiple-batch constraints also requires the use of an estimate of batch-to-batch variability.
- Target of inference -- This affects the type of statistical interval to be used in the acceptance test. If the target of inference is a single fixed true value (e.g., oxide mass fraction or true property value in a single batch), a confidence interval is appropriate. If the target of inference is the proportion of values conforming to the requirement, a tolerance interval is appropriate.

Several technical issues and possible modifications to the FTA are discussed:

- considerations in choosing multipliers used in construction of statistical intervals,
- the role of the normal (Gaussian) distribution in statistical inference,
- separate and simultaneous control of confidence (error rates) in statistical testing,
- an alternate method for treating model uncertainty and the relationship of this method to the Qualified Composition Region, and
- the possible applicability of Bayesian methods to the FTA.

Testing of the FTA via its implementation in the Plant Simulation Code should assist in resolving some of these issues and in suggesting worthwhile modifications of the algorithm.

Finally, lists of inputs, outputs, and supporting algorithms for the FTA are presented.

GLOSSARY

Acceptable--A batch or composition for which all applicable requirements will be met (with some degree of statistical confidence, as discussed in the body of the document).

Analytical uncertainty--Uncertainty among analytical results from the same sample. This is a composite form of uncertainty, made up of *variability* induced during sample preparation and the inherent *error* of the measurement process itself.

Batch--A discrete quantity of material (waste, frit, recycle, or a combination of the three) to be processed by the Hanford Waste Vitrification Plant (HWVP).

Batch-to-batch variability--Heterogeneity between *batches* made from the same *waste type*.

Bias--Consistent departures of measured or estimated quantities from the true value; compare *error*.

Components of covariance--*Covariance matrices* representing hierarchical levels of uncertainty for multivariate data.

Components of variance--*Variances* representing hierarchical levels of uncertainty in univariate data.

Composition--The proportions of each chemical species in a batch of material to be processed by the HWVP; usually expressed as mass fractions of nine major oxides (SiO_2, B_2O_3, Na_2O, Li_2O, CaO, MgO, Fe_2O_3, Al_2O_3, ZrO_2) and a catchall tenth category, Others. In some cases, individual species normally included in Others may be segregated.

Composition uncertainty--Uncertainty in measured or estimated quantities stemming from *variability* in material and/or sampling and analytical *error*.

Compositional data--A type of multivariate data in which the numerical values in each datum are the proportions (or percentages) of the individual components of the material or characteristic being represented by the datum. From their nature as proportions (percentages), these numerical values must lie between 0 and 1 (0 and 100%), inclusive, and they must sum to 1 (100%).

Confidence--A measure of the long-run performance of a statistical procedure, expressed as the probability that the procedure produces the advertised result. For example, the procedure for producing a 95% confidence interval for the mean of a population has a 95% chance of producing an interval that traps the mean. Note that *confidence* pertains to the procedure and not to any particular result.

Confidence interval--A type of statistical interval designed to trap, with specified *confidence*, a single fixed true value, such as the mean of a random variable.

Correlation--A standardized *covariance* which must lie between -1 and 1, *correlation* is computed by dividing the covariance between two random variables by the product of the standard deviations of the two variables.

Correlation matrix--A standardized representation of the interrelationships between individu quantities that make up a multivariate datum, the *correlation matrix* is a symmetric matrix with 1's on the diagonal and the pairwise *correlations* in the off-diagonal positions.

Covariance--A measure of the tendency of two random quantities to vary together, *covari* is defined as the expected value of the product of the deviations of the two random quant from their respective means, i.e., Covariance(X,Y) = $E(X - \mu_X)(Y - \mu_Y)$. Positive covarian indicates that the two quantities tend to increase or decrease together. Negative covarian indicates that one quantity tends to increase while the other decreases (or vice versa). Covariance can be estimated from a sample of n pairs (X_i, Y_i), i = 1, ..., n, with the form

$$\mathrm{Cov}(X,Y) = \sum_{i=1}^{n} \frac{(X_i - \overline{X})(Y_i - \overline{Y})}{n-1} .$$

Covariance components--See *components of covariance.*

Covariance matrix--A representation of the uncertainties and interrelationships betwee individual quantities that make up a multivariate datum, the *covariance matrix* is a s matrix with the variances of the individual quantities on the diagonal and the pairwi *covariances* in the off-diagonal positions.

Critical component constraints--Constraints imposed by HWVP on the mass fraction several minor chemical species that may (e.g., for reasons of solubility) impair melt function or product acceptability.

CVS region constraints--Constraints imposed by HWVP on mass fractions of the n oxides and Others in the feed material. These constraints are related to the extent Composition Variability Study (CVS) database and are intended to discourage extr CVS property models to compositions outside the region studied by CVS.

Direct constraints--Requirements and constraints on HWVP material (feed compo and glass) that pertain directly to measured quantities (e.g., oxide mass fractions) functions of these measured quantities.

Error--The random deviation of a measured or estimated quantity from the true v to the imperfection of the sampling or analytical procedure.

Feed--Though technically referring to material after processing in the Slurry Mi *feed* or *feed material* will here be used as a generic term to refer to any materi processed by HWVP upstream of the melter itself; compare *melt.*

Long-term variability--Heterogeneity in material over waste types.

Melt--Material being processed by HWVP in the melter or before it has cooled and solidified into glass. Before reaching the melter, this material will be referred to as *feed.*

Model uncertainty--Uncertainty in an estimated property value stemming from imperfection of the model used to relate feed composition to the property.

Modelled properties--Properties of HWVP feed, melt, or glass for which statistical models are being developed to relate feed composition to the property values.

Multiple-batch requirement or constraint--A requirement or constraint imposed over a set of batches to be processed by the HWVP; e.g., a property for which the requirement is imposed on an entire *waste type*, rather than on the individual batches constituting the waste type. Compare *single-batch requirement or constraint.*

Nuisance uncertainty--Uncertainties that may be quantified and removed from a statistical procedure in order to increase the efficiency of the procedure.

Prediction interval--A type of statistical interval designed to trap, with specified *confidence*, a single random true value, such as a new observation of a random variable.

Processability properties and requirements--Properties of and requirements on HWVP feed material that are related to the ability to process the material effectively, efficiently, and without damage to equipment.

Reference constraint set--The current set of requirements and constraints to be imposed on HWVP feed, melt, and glass. This set will be used to target and identify acceptable batches and to identify remediation strategies for unacceptable batches.

Sampling uncertainty--Uncertainty among samples from the same parent material; this is a composite form of uncertainty, made up of *variability* (heterogeneity) in the parent material and the inherent *error* of the sampling process itself.

Single-batch requirement or constraint--A requirement or constraint imposed on each individual batch to be processed by the HWVP, with no reference to the characteristics of preceding or succeeding batches. Compare *multiple-batch requirement or constraint.*

Standard deviation--Defined as the square root of the *variance*, the *standard deviation* is a measure of uncertainty on the same scale as the original quantity. Roughly, the standard deviation is the average distance of an observed value from the mean.

Stand-in constraints--Constraints imposed on mass fractions (and functions thereof) of the ten major glass components (nine oxides and Other) that are intended to control crystallinity of the glass.

Tolerance interval--A statistical procedure designed to trap, with specified *confidence*, a specified proportion of the distribution of a random variable. The proportion of the distribution to be trapped is termed the *content* of the tolerance interval. For example, a 95%/99% tolerance interval traps 99% of the distribution with 95% confidence.

Uncertainty--A general term used to refer to any of several measures of the random behavior of some quantity; for example, see *composition uncertainty*, *model uncertainty*, *variability*, and *error*.

Variability--Uncertainty related to heterogeneity in material under examination; for example, see *batch-to-batch variability* and *within-batch variability*.

Variance--A statistical measure of the random behavior of some quantity, *variance* is defined as the expected value of the squared deviation of a random variable, X, from its mean, μ, i.e., Variance(X) = $E(X - \mu)^2$. Variance can be estimated from a sample, X_i, i = 1, ..., n, with the formula

$$s^2 = \sum_{i=1}^{n} \frac{(X_i - \overline{X})^2}{n - 1}$$

Variance components--See *components of variance*.

Variance-covariance matrix--See *covariance matrix*.

WAPS properties and requirements--Properties of and requirements on glass produced by HWVP, as detailed in the *Waste Acceptance Product Specifications* (WAPS; DOE, 1993). These properties and requirements are related to the performance of the glass in the repository.

Waste loading--The mass fraction of waste in a batch of feed or in the resulting glass.

Waste type--A relatively homogeneous stream of waste to be processed by the HWVP. Several to many *batches* will be made from a single waste stream.

Within-batch variability--Heterogeneity in a single batch of material.

ACRONYMS

CVS--Composition Variability Study

DWPF--Defense Waste Processing Facility

EA--Environmental Assessment

FTA--Feed Test Algorithm

HWVP--Hanford Waste Vitrification Plant

LCB--Lower confidence bound

LTB--Lower tolerance bound

MEM--Measurement Error Model

PCC--the system to be used by HWVP for product composition control

PCT--Product Consistency Test

PHTD--Pacific Northwest Laboratory (PNL) HWVP Technology Development

PPMD--Process/Product Model Development

QCR--Qualified Composition Region

SME--Slurry Mix Evaporator

UCB--Upper confidence bound

UTB--Upper tolerance bound

WAPS--Waste Acceptance Product Specifications

CONTENTS

1.0 INTRODUCTION

The Hanford Waste Vitrification Plant (HWVP) will immobilize transuranic and high-level radioactive waste in borosilicate glass. Similar operations will be performed in the Defense Waste Processing Facility (DWPF) at the Savannah River Site. DWPF has developed a Product Composition Control System for controlling feed slurry composition (which affects glass properties) and for checking and documenting product quality (Postles and Brown, 1991). The *HWVP Project Waste Form Qualification Program Plan* (Randklev, 1993) calls for the development of a product composition control-type system to perform these functions for the HWVP. No name for the HWVP product composition control system has yet been generally agreed upon. PCC (from product composition control) will be used here to refer to the system under development for HWVP.

The major objective of the Process/Product Model Development (PPMD) cost account of the Pacific Northwest Laboratory HWVP Technology Development (PHTD) Project is the development of a PCC system. Briefly, control of HWVP operations and product quality will be achieved by a series of mathematical/statistical algorithms. Bryan and Piepel (1993) discuss the statistical strategy for the product composition control system and the major algorithms being developed for the HWVP. One of the algorithms to be incorporated in this system is the Feed Test Algorithm (FTA). This document describes the FTA. The remainder of this section defines the objective of the FTA, introduces terminology, and sketches the issues to be addressed in subsequent sections.

As currently envisioned, the HWVP will process material in batches by combining three input streams (waste, frit, and recycle) in a tank known as the Slurry Mix Evaporator (SME). The SME is the last stage at which feed composition can be adjusted. From the SME, a batch of slurry will be passed to the Melter Feed Tank, then to the melter. The molten material (or melt) from the melter will then be poured into canisters for cooling and eventual disposal in a geologic repository.

Some definitions and terminology are now in order. *Composition* will be used to refer to the chemical species (or proportions thereof) in a given material (e.g., waste, frit, recycle, and combinations of these). Compositions are usually expressed as mass fractions of nine individual oxides (SiO_2, B_2O_3, Na_2O, Li_2O, CaO, MgO, Fe_2O_3, Al_2O_3, ZrO_2) and a catchall tenth category, Others. This convention was adopted for studies which are developing melt and glass property models based on composition. *Batch* denotes a discrete quantity of material to be processed. The main focus of the HWVP PCC will be SME batches (material residing in the SME), since this is the last stage at which feed composition can be modified. Batches will fall rather naturally into groups, with the batches in each group deriving from a relatively homogeneous waste stream. This homogeneous waste stream and the batches made from it will be referred to as a *waste type.*

Various properties of the melt and the resulting glass influence effectiveness of processing and disposal. These properties are largely determined by the composition of the melter feed, the slurry from which the melt and glass are derived. PCC algorithms other than the FTA will choose a target mixture (of waste, frit, and recycle) for the current SME batch and will estimate the SME composition that results from the mixing operations. The task of the FTA is to use the estimated composition of the SME batch to decide whether the batch will be *acceptable*, i.e., whether the batch will yield a melt and glass meeting all requirements and constraints imposed on composition and glass/melt properties. This task would be quite simple if each property of the melt and glass were known or could be measured without error: we would simply compare the known property value to the requirements on that property. Unfortunately, as the remainder of this document discusses, the situation is not this simple. At least two complications arise in certifying acceptability.

First, since the SME is the last stage of the process at which composition can be modified, and since several of the most important requirements are imposed on stages of the process following the SME (melt and glass), these *downstream properties* must be calculated from the estimated SME composition. Glass/melt properties, requirements, and the models used in calculating glass/melt properties are discussed in Section 2.

The second complication in certifying acceptability arises from the existence of uncertainty in all real-world measurement and estimation procedures and in the mathematical/statistical modelling used to calculate downstream properties. Fortunately, methods exist for quantifying these uncertainties. Section 3 discusses various uncertainties in measurement, estimation, and modelling, and the methods used to estimate and to represent these uncertainties.

A reasonable approach to certifying acceptability must take into account the inevitable uncertainties discussed above. Rather than asking whether the estimated value of an attribute falls within requirements, we must ask whether it is reasonable to conclude that the *true* value falls within requirements, given the estimated value *and the magnitude of the uncertainties in the estimate.* This drives us inexorably into the realm of statistics. Section 4 discusses the statistical methods used to draw conclusions about unknown true values given estimates of the values and estimates of the uncertainties in these values.

Section 5 draws upon the material in Sections 2, 3, and 4 to derive methods for testing compliance with each of the requirements and constraints imposed on HWVP material. The FTA described in this document is preliminary, because design of the HWVP process and knowledge of the product are evolving. This evolution may require some modification of the techniques described in Section 5. Section 6 discusses some possible modifications and alternative approaches, as well as some technical issues that should be revisited as knowledge accumulates. Section 7 lists possible inputs and outputs of the FTA, as well as supporting algorithms.

2.0 ATTRIBUTES[a], REQUIREMENTS, AND MODELS

Requirements and constraints will be imposed on many attributes of the HWVP feed, melt, and glass. Models are being developed to relate feed composition to properties of the resulting melt and glass. The development of property models is one of the objectives of the Composition Variability Study (CVS), in which glasses of known composition are fabricated, and properties of the resulting melts and glasses are measured. The CVS is described in detail by Hrma, Piepel, et al. (1992). Requirements and constraints that will be imposed fall into three broad categories:

- Requirements related to performance of the final glass in the repository -- These requirements are imposed by the *Waste Acceptance Product Specifications* (WAPS; DOE, 1993), so these will be referred to as *WAPS requirements.*
- Requirements and constraints related to the processability of the material -- These requirements are imposed by the HWVP project; they will be referred to as *processability constraints and requirements.*
- The *CVS composition region constraints* -- These constraints are essentially the limits of the composition region explored by CVS and are necessary because of the danger inherent in extrapolating models developed from the CVS database (i.e., applying these models to compositions outside the range of compositions examined by CVS). CVS region constraints apply directly to the ten major components of feed, melt, and glass.

The major WAPS requirement is imposed by WAPS 1.3 on "product consistency," as measured by the Product Consistency Test (PCT; Jantzen, 1992b). The PCT measures the quantities of elements released from ground glass in deionized water. The WAPS requires that "the mean concentrations of lithium, sodium and boron in the leachate ... shall each be

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(a) Established usage reserves the word *property* for characteristics of the melt and glass (which will usually be estimated via models based on feed composition), but requirements and constraints will also be imposed on feed slurry composition (oxide mass fractions and functions thereof). To avoid confusion, the word *attribute* will be used to refer to any characteristic upon which a requirement or constraint is imposed.

less than those of the" Environmental Assessment (EA) benchmark glass, described in Jantzen (1992a). The FTA will treat Li, Na, and B separately; i.e., WAPS 1.3 will be interpreted as establishing three separate requirements on HWVP material.

The WAPS also imposes requirements on attributes of the canistered waste form, and it requires *reporting* other glass attributes (i.e., no requirements are imposed on the attributes themselves). Attributes of the canistered waste form upon which requirements are imposed include free liquid, gas, explosiveness, pyrophoricity, combustibility, organic materials, chemical compatibility, heat generation, maximum dose rate, and subcriticality. These attributes are not expected to be limiting and are not considered further here. Reporting requirements include chemical composition, crystalline phases, radionuclide inventory, phase stability information (glass transition temperature and time-temperature-transformation diagrams), and results of the Toxicity Characteristic Leaching Procedure. Since only reporting these attributes is required, they play no role in identification of acceptable compositions and are not considered further here.

Processability requirements include those on viscosity at 1150°C, electrical conductivity at 1150°C, liquidus temperature (possibly separate requirements for different crystalline phases), redox state (Fe^{++}/Fe), phase separation, melt rate, and critical components. CVS has constructed satisfactory models for viscosity and electrical conductivity. Preliminary liquidus temperature models have been developed, but they are not considered satisfactory at this time. Redox rate may be directly measured. Work is underway on phase separation and melt rate, so these properties will not be further addressed here. *Stand-in constraints* on functions of the ten major glass components (nine oxide species and Others) have been used to address crystallinity of the glass, which is related to liquidus temperature. When satisfactory models become available for liquidus temperature and crystallinity behavior, the stand-in constraints may still be active as CVS region constraints, due to the role the stand-in constraints have played in defining the CVS experimental program. *Critical component constraints* are upper bounds on mass fractions of several minor species that may impair melter function for some reason (e.g., solubility).

The current *reference constraint set* appears in Table 1. Some of these constraints will not be included in the preliminary FTA, due to unavailability of satisfactory models or other difficulties. This set is subject to modification as requirements change, as the CVS database grows (which may relax some of the CVS region constraints), and as new CVS property models become available. As discussed in Section 6.6, incorporation of new requirements into the FTA and modification of existing requirements will present no new technical difficulties.

Table 1. Current Reference Constraint Set

Category	Constraints	Lower Limit[a]	Upper Limit[a]
WAPS	PCT for Li	n/a	4.8 g/m^2 [b]
WAPS	PCT for Na	n/a	6.6 g/m^2 [b]
WAPS	PCT for B	n/a	8.2 g/m^2 [b]
Processability	Viscosity at 1150°C	2 Pa·s [c]	10 Pa·s [c]
Processability	Electrical conductivity at 1150°C	18 S/m [c]	111 S/m [c]
Processability	Liquidus temperature	n/a	1050°C
Processability	Redox state (Fe^{++}/Fe)	0.005 [c]	0.23 [c]
Processability, Stand-in	SiO_2 / Al_2O_3	3.0	n/a
"	MgO + CaO	n/a	0.08
"	Fe_2O_3 + Al_2O_3 + ZrO_2 + Others	n/a	0.225
"	Al_2O_3 + ZrO_2	n/a	0.14
"	MgO + CaO + ZrO_2	n/a	0.18

(a) Limits are expressed as mass fractions unless otherwise specified.

(b) WAPS does not specify limits. These limits are based on PCT testing of the EA glass by the Savannah River Technology Center (WSRC, 1993). It is envisioned that these limits will be applied by HWVP to models of Li, Na, and B PCT release from quenched glass as well as canister centerline cooled glass.

(c) Units on viscosity are Pascal-seconds. Units on electrical conductivity are Siemens per meter (Siemens = Ohm^{-1}). Redox state is expressed as a unitless ratio of Fe^{++} to total Fe.

Table 1. Current Reference Constraint Set (continued)

Category	Constraints	Lower Limit[a]	Upper Limit[a]
Processability Critical Component	Cr_2O_3, SO_3	n/a	0.005
"	P_2O_5	n/a	0.010
"	F	n/a	0.017
"	Rh_2O_3, PdO, Ru_2O_3	n/a	0.025
CVS Region	SiO_2	0.42	0.57
"	B_2O_3	0.05	0.20
"	Na_2O	0.05	0.20
"	Li_2O	0.01	0.07
"	CaO	0	0.10
"	MgO	0	0.08
"	Fe_2O_3	0.005	0.15
"	Al_2O_3	0	0.17
"	ZrO_2	0	0.13
"	Others	0.01	0.10

(a) Limits are expressed as mass fractions unless otherwise specified.
(b) WAPS does not specify limits. These limits are based on PCT testing of the EA glass by the Savannah River Technology Center (WSRC, 1993). It is envisioned that these limits will be applied by HWVP to models of Li, Na, and B PCT release from quenched glass as well as canister centerline cooled glass.
(c) Units on viscosity are Pascal-seconds. Units on electrical conductivity are Siemens per meter (Siemens = Ohm^{-1}). Redox state is expressed as a unitless ratio of Fe^{++} to total Fe.

The classification given above is useful for understanding the origins and roles of these requirements and constraints, but, for identifying the proper approaches to statistical testing of these requirements, two other distinctions are more important. These distinctions relate to the uncertainties that must be considered by the statistical tests.

The first distinction is that between *direct constraints* and *constraints on modelled properties*. Direct constraints are those that apply to directly to measured quantities (e.g., mass fractions of certain components) or to *known* functions of these measured quantities. Examples include the stand-in constraints, the critical component constraints, the CVS region constraints, and the constraint on redox state. The only uncertainties to be taken into account in testing these constraints are *composition uncertainties* (see Section 3.1 for more information on composition uncertainty).

Modelled properties include those properties for which CVS is developing empirical models, as functions of composition, with parameters estimated from the CVS database. Examples of modelled properties include viscosity at 1150°C, electrical conductivity at 1150°C, and PCT for B, Li, and Na. (Constraints on liquidus temperature and other properties will be added to the FTA as models become available.) The property models being developed by CVS are *second-order mixture models*, the general form of which is

$$\phi_k = \sum_{i=1}^{10} a_{ik} x_i + \sum_{i=1}^{9} \sum_{j>i}^{10} b_{ijk} x_i x_j \qquad (1)$$

where ϕ_k is the k-th melt/glass property (or, in some cases, a simple mathematical transformation thereof), the x_i and x_j are the mass fractions of the i-th and j-th oxides, and the a_{ik} and b_{ijk} are the coefficients of the relation between the oxide mass fractions and ϕ_k (to be estimated from the CVS database). The oxide mass fractions used in a mixture model must sum to 1, that is,

$$\sum_{i=1}^{10} x_i = 1$$

Several of the models developed by CVS are *first-order*, meaning that, for some properties (k), $b_{ijk} = 0$ for all i and j. CVS may employ theoretical models of liquidus temperature; these models may differ in form from the mixture models developed for other properties. The details of the CVS database, models, estimation techniques, and validation techniques are

discussed by Hrma, Piepel, et al. (1992).

Each of the models developed by CVS has some associated *model uncertainty*, because the coefficients of the models are estimated from experimental data. Both composition uncertainty and model uncertainty must be taken into account in statistical testing of constraints on modelled properties, with the result that the testing methods are somewhat more complicated for constraints on modelled properties than for direct constraints. Model uncertainty is expressed as a covariance matrix for the estimated coefficients, a_{ik} and b_{ijk}. Model uncertainty is discussed further in Section 3.1, covariance matrices are discussed in Section 3.2, and methods for combining composition and model uncertainties are discussed in Section 3.3.

The second distinction important in identifying proper statistical methods is that between *single-batch requirements* and *multiple-batch requirements*. Single-batch requirements apply to the material within a single batch; i.e., these requirements are imposed on a batch-by-batch basis, so that the quality of preceding and succeeding batches does not affect the acceptability of the current batch. In contrast, a multiple-batch requirement is one imposed on a set of batches (e.g., all batches derived from a single waste type). For example, a requirement that the value of Property A be less than 10 for each batch is a single-batch requirement, whereas a requirement that the *mean value* of Property A over some set of batches be less than 10 is a multiple-batch requirement.

Most requirements imposed on HWVP material are of the single-batch type. The WAPS 1.3 PCT requirements are the only members of the reference constraint set that will be tested as multiple-batch requirements, and they will also be tested as single-batch requirements (see Section 4.4 for discussion). Statistical methods used for single-batch requirements must account for uncertainty within a single process batch, but methods for multiple-batch requirements must also account for uncertainty *between* process batches. (The various sources of uncertainty in the HWVP process are discussed in Section 3.1.)

All of the direct constraints identified above are also single-batch constraints; statistical testing for single-batch direct constraints is discussed in Section 5.1. Statistical methods for single-batch constraints on modelled properties are discussed in Section 5.2. Statistical methods for multiple-batch constraints on modelled properties (the WAPS 1.3 PCT requirements) are discussed in Section 5.3. There are no multiple-batch direct constraints in the reference constraint set -- the statistical method for such constraints would be a simplified version (omitting model uncertainty) of the method for multiple-batch constraints on modelled properties.

3.0 UNCERTAINTY: SOURCES, REPRESENTATION, AND ESTIMATION

Uncertainty arises in several places and forms in the HWVP process and the data therefrom. The various sources of uncertainty arise from different causes and may be best handled in different ways. Therefore, it is useful to distinguish between several broad categories of uncertainty. Terminology becomes a problem here. In this document, uncertainty (without a preceding modifier) will be used to refer to all sources. When uncertainty is used to refer to a more specific source, it will be preceded by a qualifier, e.g., model uncertainty.

3.1 TYPES AND SOURCES OF UNCERTAINTY

Uncertainty in the HWVP process can be divided into two broad categories: model uncertainty and composition uncertainty[(b)]. The latter category can be subdivided into *variability* (heterogeneity) in material, and *error* in sampling and analytical procedures. (In fact, a similar subdivision of model uncertainty is possible, but will not be considered here.)

As discussed in Section 2, models will be used to calculate melt and glass properties

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(b) Composition uncertainty might also be called *data uncertainty*, since it exists to some degree in virtually any process used to collect data. However, the main type of data to be used in HWVP product control will be compositional data, so the more specific term will be used here.

from feed composition. These models will not be perfect representations of the true relationships between feed composition and melt/glass properties. One strength of the CVS program is that it will yield not only models, but also estimates of the uncertainty inherent in the models. This model uncertainty must be considered when judging the quality of a process batch.

The input streams (waste, frit, and recycle) and the contents of the SME will all be heterogeneous to some degree. Given this fact, even if we could obtain an exact measurement from a single place and time, we would not be guaranteed that we know the truth at all other places and times. This spatial and temporal variability is inevitable (though, of course, a well-designed process can minimize it). Variability in the HWVP process can be broken down into several components:

- long-term variability, e.g., between waste types or batches of frit (over the life of the HWVP);
- batch-to-batch variability, i.e., heterogeneity between process batches made from the same waste type and frit batch; this type of heterogeneity might also be called between-batch variability or within-waste type variability;
- within-batch (or within-tank) variability, i.e., heterogeneity within a single process batch (and the streams used to make the batch); and
- within-sample variability, i.e., heterogeneity within a sample of material.

Even if the input streams and the tank contents were perfectly homogeneous (zero variability), it would still be quite difficult to know the exact compositions, for the simple reason that most sampling and analytical procedures are imperfect. Samples drawn from a single homogeneous tank or stream will usually differ slightly from each other and hence from the true (but usually unknown) composition. This may occur because of differential efficiency of the sampling technique with respect to the various physical phases or chemical species in the sampled entity. Multiple analyses of the same sample will usually yield results that are not in perfect agreement, even if the sample itself is perfectly homogeneous.

Unpredictable departures of samples and analytical results from the true but unknown state of nature will be referred to here as sampling and analytical error.

For the sake of completeness, it should be noted that a third source of uncertainty might exist in some stages of HWVP measurement processes: *bias*, which is here used to refer to consistent, predictable departures from reality in sampling and analytical results. The consistent, predictable nature of bias distinguishes it from what has been termed sampling and analytical error. For example, a sampling technique may be known (or may be shown) to regularly underrepresent the amount of one or more chemical species in the sampled tank or stream. Similarly, an analytical procedure may result in consistent underestimation of some chemical species. Bias should be controlled operationally (by improving the procedure) or therapeutically (by correcting known deficiencies when reporting or using results), but the detection, documentation, and correction of sampling and analytical biases is beyond the scope of this document. Possible biases in sampling and analytical methods should be investigated in future efforts (see Bryan and Piepel, 1993, for more information). In all that follows, it will be assumed that bias has been eliminated.

The two components of composition uncertainty, variability and error, may sometimes be confounded at a given point in the process. For example, both within-batch variability and sampling error contribute to observed differences between multiple samples drawn from a single tank or stream. This *combination* of within-batch variability and sampling error will be referred to here as *sampling uncertainty*. Similarly, the phenomenon commonly known as *analytical uncertainty* is actually a combination of variability induced during sample preparation and the inherent error of the measurement process itself. Dissection of these composite types of uncertainty would be quite difficult (and expensive); therefore, sampling uncertainty is usually treated as an indivisible source of uncertainty, as is analytical uncertainty.

Some sources of uncertainty described above are not relevant to the problem of judging acceptability of process batches. For example, long-term variability is important in

plant design and in establishing the range of compositions included in the CVS database, but this source of uncertainty is not relevant to judging the processability of a single batch, nor is it relevant to judging the quality of glass made from a single waste type (i.e., compliance with WAPS 1.3). Therefore, long-term variability is not discussed further here.

Two sources of uncertainty must be considered in judging acceptability of a process batch:

- Within-batch variability -- This is important in judging compliance with single-batch requirements.
- Batch-to-batch variability (within a waste type) -- This is important in judging compliance with WAPS 1.3 (and any other multiple-batch constraints that may be added to the reference constraint set).

Statistical testing requires estimates of these sources of uncertainty, but, as noted above, measured quantities include other uncertainties, such as within-sample variability, sampling error, and analytical error. If present, these *nuisance uncertainties* decrease the efficiency of statistical tests. In some cases, it may be possible to isolate and eliminate these nuisance uncertainties. This issue is related to estimation and use of *components of variance* and *components of covariance*, which are discussed in more detail by Bryan et al. (1994b).

3.2 REPRESENTATION OF UNCERTAINTY

The standard method of representing uncertainty in a single (univariate) measured or estimated quantity is in terms of the *standard deviation* (sometimes called the *standard error*) of the estimate. In the following, it will often be more convenient to refer to the *variance* of the estimate. The variance is simply the square of the standard deviation and is defined as the expected value of the squared deviation of an observation from the mean of the distribution of the quantity. The standard deviation, being the square root of the variance, has units identical to the quantity itself and hence is more easily used in judging the uncertainty in the quantity. Both terms, variance and standard deviation, will be used below without

further comment.

HWVP product control will be achieved by controlling feed composition, which is usually expressed as a ten-component vector (nine individual oxides and a catchall tenth category, Others). Thus, feed composition is a *multivariate* form of data, in which each observation consists of a vector of individual measurements or estimates. This complicates the representation of uncertainty, in that we must account for *covariance* between components of the composition. The covariance of two random quantities is defined as the expected value of the product of the deviations of the two quantities from their respective means. Covariance is a measure of the tendency of the two quantities to vary together. A positive covariance indicates that the two quantities vary directly (one tends to be greater than its mean when the other is), while negative covariance indicates that the two vary inversely (one tends to be greater than its mean when the other is smaller than its mean) and zero covariance indicates that the two vary independently (here used in the lay sense, rather than in the technical sense of probability and statistics). The (Pearson product-moment) *correlation* between two random quantities is simply the covariance divided by the square roots of the variances. This *correlation coefficient* is a standardized covariance that must lie between -1 and 1, inclusive.

For multivariate data, the standard method of representing uncertainty is the *variance-covariance matrix*, a square (symmetric) matrix with the variances of the individual components on the diagonal and the pairwise covariances in the off-diagonal positions. (For brevity below, the variance-covariance matrix will be referred to as the *covariance matrix*.) For example, if Σ represents the covariance matrix of a vector of length three, then

$$\Sigma = \begin{pmatrix} \sigma_1^2 & \sigma_{12} & \sigma_{13} \\ \sigma_{21} & \sigma_2^2 & \sigma_{23} \\ \sigma_{31} & \sigma_{32} & \sigma_3^2 \end{pmatrix}$$

where σ_i^2 is the variance of the i-th element of the vector and $\sigma_{ij} = \sigma_{ji}$ is the covariance between the i-th and j-th elements. (The covariance of a random variable with itself is simply

the variance of the random variable.) The relationship of the covariance matrix to the vector of estimated quantities is analogous to the relationship of the variance to a single estimated value. Related to the covariance matrix is the *correlation matrix*, a square (symmetric) matrix with 1's on the diagonal and the pairwise correlations in the off-diagonal positions.

The specific nature of the multivariate data involved in HWVP product control further complicates the representation and interpretation of uncertainty. Ideally, the mass fractions used to represent a single composition should sum to one (or 100%). Thus, the data involved in HWVP product control are *compositional data*, a type of multivariate data in which the numerical values in each datum are the proportions (or percentages) of the individual components of the material or characteristic being represented by the datum. From their nature as proportions (percentages), these numerical values must be lie between 0 and 1 (0 and 100%), inclusive, and they must sum to 1 (100%). This *unit-sum restriction* is both the defining feature of compositional data and the source of technical difficulties associated with compositional data. Compositional data are discussed in depth by Aitchison (1986); the implications of the compositional nature of the HWVP data are largely beyond the scope of this document but are discussed briefly in the Section 6.2.

Several of the sources of uncertainty discussed in Section 3.1 are hierarchical. For example, uncertainty exists among analyses on a single sample, among samples within a single batch or input stream, among batches within a waste type, and among waste types. Each estimated feed composition includes uncertainty introduced at each level of this hierarchy. As discussed in Bryan et al. (1994b), proper estimation of uncertainty in attribute values requires recognition of this nested structure. Briefly, the uncertainty at each level in this hierarchy may be represented by a variance (for a single measured quantity) or a covariance matrix (for a multivariate quantity). These hierarchical representations of uncertainty are known as *components of variance* and *components of covariance.*

3.3 ESTIMATION OF UNCERTAINTY FOR MODELLED PROPERTIES

Estimates of model uncertainty will be obtained from CVS. The estimates of composition uncertainty required by the FTA will be supplied by the existing algorithm known as the Measurement Error Model (MEM) or as the Measurement Correction Algorithm. This algorithm reconciles several sources of information[(c)] on tank contents and produces optimized estimates of feed composition, other data, and the associated uncertainties (covariance matrices). It is possible that some uncertainty estimates may have to be modified, combined, or updated during plant operation (e.g., during processing of a single batch or from batch to batch). These and other issues relating to estimation of uncertainty are discussed by Bryan et al. (1994b). In what follows, the relevant covariance matrices are assumed to be available. The section focuses on derivation of an estimate of uncertainty for a modelled property value, using the available covariance matrices for feed composition and the coefficients of the model.

Derivation of an estimate of uncertainty for a modelled property value is an application of the general procedure known as *propagation of error*. Briefly, one method of propagation of error works as follows. Let y represent a property of interest[(d)], and assume that $y = f(\mathbf{z})$, where $\mathbf{z}$ is a random vector with mean μ_z and covariance matrix Σ_z. Then, using a Taylor series expansion about μ_z to approximate $f(\mathbf{z})$, an approximation to the variance of the estimated y, σ_y^2, can be derived:

$$\sigma_y^2 \approx \mathbf{d}_z' \Sigma_z \mathbf{d}_z,$$

(c) Among the sources of information used by the MEM are pressure differences and gauge heights in various tanks, measured concentrations of various components, masses of samples, and constants relating oxide masses to cation masses. Reconciliation done by the MEM relates to several types of mass balances for material in and transferred between tanks. In addition to estimates of feed composition, the MEM produces optimized estimates of tank levels and densities of material.

(d) Standard statistical/mathematical practice is to place a caret ("hat") over the symbols for estimated quantities (e.g., data, parameters, and uncertainties). Since all the quantities to be employed in the FTA will be estimated, the "hats" will be omitted in much of this document, thereby preventing a potentially bewildering proliferation of such symbols.

where d_z is the gradient (i.e., the vector of partial derivatives) of f with respect to z. The partial derivatives are to be evaluated at the observed value of z.

Two distinct sources of uncertainty enter into the propagation of error for calculation of a melt or glass property, y, from an estimated feed composition, x. First is composition uncertainty, the uncertainty in the estimate of feed composition. For simplicity of presentation, it is assumed here that a single covariance matrix for feed composition is available. The case of several covariance components for feed composition is briefly discussed at the end of this section.

The second source of uncertainty entering into the calculation of y is model uncertainty. Let β be the vector of estimated coefficients (parameters) in the model relating y to x. Model uncertainty is quantified by a covariance matrix associated with β; denote this covariance matrix by Σ_β. The method of propagation of error outlined above can be applied to the special case of $y = f(x,\beta)$. Denote the gradients of $f(x,\beta)$ with respect to x and β by d_x and d_β, respectively. If x and β are *uncorrelated* random vectors (a reasonable assumption unless x is part of the data used to estimate β), the approximate variance of y divides neatly into two parts, one attributable to the uncertainty in x (i.e., composition uncertainty), the other attributable to uncertainty in β (i.e., model uncertainty):

$$\sigma_y^2 \approx d_x'\Sigma_x d_x + d_\beta'\Sigma_\beta d_\beta \qquad (2)$$

For the special case where the function $f(x,\beta)$ is linear in both the data, x, and the parameters, β, this formula takes on an even simpler form. For this case,

$$y = f(x,\beta) = x'\beta$$
$$d_x = \beta$$
$$d_\beta = x$$

$$\sigma_y^2 \approx \beta'\Sigma_x\beta + x'\Sigma_\beta x \qquad (3)$$

The role of this estimated variance in constructing tests for the acceptability of a feed batch is discussed in Sections 5.2 and 5.3.

If several covariance components relevant to feed composition are available, these covariance components can be propagated separately and the resulting variance components combined to form an overall estimate of uncertainty in y arising from the various covariance components. This subject is beyond the scope of this document but is covered in greater detail in Bryan et al. (1994b).

4.0 STATISTICAL INTERVALS AND DECISION-MAKING

The purpose of the FTA is to construct statistical tests for acceptability of a feed batch with respect to the various requirements imposed on the HWVP process and product. The statistical tests to be used by the FTA are intimately linked with statistical *intervals*. In this section, three types of statistical intervals will be discussed, as will the general principles underlying the applicability of each interval type to the requirements imposed on HWVP feed, melt, and glass. Specific applications of these interval types to specific requirements are discussed in Section 5. Technical issues surrounding the construction of statistical intervals are discussed in Section 6.

A statistical interval is, roughly, a range of values in which an unknown true value is believed (or expected) to lie. The interval is defined by a lower bound, an upper bound, or both. A *two-sided* statistical interval has both a lower bound and an upper bound. A *lower one-sided* interval is bounded only below (no statement is made about an upper limit), while an *upper one-sided* interval is bounded only above (no statement is made about a lower limit). The bounds themselves are often referred to as two-sided or one-sided. Due to the simultaneity required of two-sided bounds, they are farther apart (for a given statistical confidence level) than the two corresponding one-sided bounds (i.e., a two-sided interval is often wider than the intersection of two one-sided intervals).

With each of the types of intervals to be discussed in this document is associated a quantity called the statistical *confidence*. Confidence is a measure of the success rate of the procedure by which a statistical interval is constructed, i.e., how often the procedure produces an interval that actually traps the true value. For example, a procedure to produce a 95% confidence bound has a 95% chance of producing a bound that traps the unknown true value. Technically, the confidence actually rests in the *procedure* used to construct the bound or interval, *not* in the bound or interval itself. Confidence refers to the long-run performance of the procedure, not the performance of any particular calculated interval. The interval itself, once produced, either does or does not trap the true value, and, though we do not know which is the case, it is not technically correct to say that the interval has a 95% chance of trapping the unknown true value. A method other than confidence for expressing strength of belief is discussed briefly in Section 6.5.

4.1 CONFIDENCE INTERVALS

A *confidence interval*[e] is designed to trap a *single fixed true value* with specified confidence. For example, a 95% confidence interval for the mean of a population is designed to trap the mean of the population with 95% confidence. Lower and upper confidence bounds (LCB and UCB, respectively) are of the form:

$$\begin{aligned} LCB &= \hat{y} - (m_{lc} \cdot s) \\ UCB &= \hat{y} + (m_{uc} \cdot s) \end{aligned} \qquad (4)$$

where $\hat{y}$ is the estimated value, s is the estimated standard deviation of $\hat{y}$, and m_{lc} and m_{uc} are multipliers, the exact definition of which depends on the statistical distribution of the data, the confidence to be associated with the interval, the desired nature of the bounds (one-sided or two-sided), and the strength of information about s. (In practice, for two-sided intervals, the two multipliers are usually equal, but this need not be the case.) The technical details

(e) This name, though endowed on the procedure by its creator, is somewhat unfortunate--as stated above, statistical confidence is associated with each of the types of statistical interval to be discussed here. The distinctions among the interval types lies in the nature of the unknown quantities they are designed to trap.

entailed in the choice of the multipliers are discussed in Section 6.1.

4.2 PREDICTION INTERVALS

A *prediction interval* is designed to trap a *single random true value* with specified confidence. This differs from the confidence interval in that the target entity of a prediction interval is a random quantity, while the target entity of a confidence interval is a fixed quantity. A prediction interval must account for the random behavior of the target entity, in addition to all the sources of uncertainty accounted for by a confidence interval. Therefore, all else being equal, a prediction interval will be wider than a confidence interval. Hence, use of prediction intervals in testing glass/melt properties is more likely to result in decisions to reject a process batch.

Perhaps some examples will make clearer the distinction between confidence and prediction intervals. If we believe that a given attribute is an exact function of true composition, then the true attribute value is a fixed quantity, all our uncertainty stems from uncertainty about the true composition and model uncertainty, and a confidence interval is appropriate. If we believe that the true attribute value is a function of true composition *plus some random effect* (i.e., that the true attribute value may differ between two batches with exactly the same composition), then the uncertainty about the true value must account for this random effect in addition to the uncertainties about the true composition and the model, and a prediction interval is appropriate. In the context of the FTA, confidence intervals are more appropriate, since we expect that batches with exactly the same composition should have the same properties, and we are interested in drawing inferences about these fixed but imperfectly known quantities. Although not applicable to the FTA, prediction intervals may be appropriate in other areas of HWVP process control, for example, in model validation and process monitoring.

The formulae for lower and upper prediction bounds are very similar to those given in Equation (4) for lower and upper confidence bounds, but the definitions of the statistical multipliers and/or the uncertainty estimate differ. Since prediction intervals will not be used

in the FTA, the details will not be given here.

4.3 TOLERANCE INTERVALS

A *tolerance interval* is designed to capture, with specified confidence, a predefined proportion of some statistical distribution of values. For example, consider testing whether some proportion of process batches from a single waste type have attribute values below some specified upper limit. In this application, individual batch attribute values are thought of as arising at random from an underlying population (the population of all batches that could have been made from this waste type), and the inference to be drawn concerns the underlying population rather than the mean or some other single characteristic of this population. The proportion of the population to be captured is termed the *content* of the tolerance interval. As with confidence and prediction intervals, this capturing is to be done with a specified level of confidence. Thus, two percentages (confidence/content) are usually used to specify a tolerance interval; for example, a 95%/99% tolerance interval is one designed to capture 99% of the underlying population with 95% confidence. Like other statistical intervals, tolerance intervals may be two-sided (an attempt to capture the central portion of the distribution), lower one-sided (an attempt to capture the upper portion of the distribution), or upper one-sided (an attempt to capture the lower portion of the distribution).

The formulae for lower and upper tolerance bounds (LTB and UTB, respectively) look very similar to those for confidence bounds, but the definitions of the items involved differ from those for confidence bounds:

$$\begin{aligned} LTB &= \hat{y} - (m_{lt} \cdot s) \\ UTB &= \hat{y} + (m_{ut} \cdot s) \end{aligned} \qquad (5)$$

where $\hat{y}$ is the estimated *mean* value and s is the estimated standard deviation *of the distribution*, and m_{lt} and m_{ut} are multipliers, the exact definition of which depends on the statistical distribution of the data, the confidence *and content* to be associated with the interval, the desired nature of the bounds (one-sided or two-sided), and the strength of

information about s. (Again, in practice, for two-sided intervals, the two multipliers are usually equal, but this need not be the case.) The technical details entailed in the choice of the multipliers are discussed in Section 6.1.

4.4 APPLICATION OF STATISTICAL INTERVALS TO THE FTA

To illustrate the role of statistical intervals in acceptance testing, assume that we wish to test whether it is reasonable to conclude that a fixed, unknown true attribute value is less than some specified upper limit, U. To perform this test, we obtain estimates of the attribute value ($\hat{y}$) and the uncertainty therein (s), choose a desirable confidence level (often 95%), and calculate the UCB corresponding to this confidence level. If UCB $\leq$ U, we conclude that the requirement is satisfied. Otherwise, we conclude that the requirement is violated, leading to rejection and remediation of the process batch.

The confidence level of this procedure controls the probability of concluding that a requirement is met (i.e., the true attribute value is "good") when in reality it is violated (the true attribute value is "bad"); in other words, confidence controls *the probability of accepting a bad batch* (since a batch is deemed bad and rejected if any attribute of the batch is deemed bad). That is, when all the assumptions of the statistical test are met, the probability of rejecting a borderline batch (one for which the true attribute value is exactly equal to the specified upper limit) is equal to the confidence level, e.g., 95%, and the probability of accepting a bad batch is no greater than 5%. As the true attribute value approaches the specified upper limit, the probability of rejection approaches 95%, implying that good batches with true attribute values near the limit are very likely to be rejected. Increased conservatism, in the form of increased statistical confidence (which implies a larger statistical multiplier and hence a larger UCB), is tantamount to increased probability of rejection of all process batches, regardless of true status (acceptable or unacceptable). That is, higher confidence levels correspond to lower probabilities of accepting bad batches (beneficial) *and to higher probabilities of rejecting good batches* (detrimental). This demonstrates that choosing a confidence level entails compromise. (Indeed, in this sense, the most "conservative" test is to simply reject all process batches!) In addition, smaller uncertainties (as measured by smaller

values of the standard deviation) correspond to smaller UCBs and hence to decreased rates of rejection for good batches. Since the procedure fixes the probability of rejection of a borderline batch at 95% (or whatever confidence level is chosen), the probability of accepting a bad batch still does not exceed 5%. This demonstrates the intuitive concept that a smaller standard deviation corresponds to increased efficiency of the statistical acceptance test.

Confidence and prediction intervals are used to draw inferences about a single (fixed or random) value; tolerance intervals are used to draw inferences about the population from which random values are drawn. Most of the requirements imposed on HWVP pertain to single fixed values (the true attribute value in a single batch), and hence the use of confidence intervals is a reasonable approach to testing most requirements.

The WAPS 1.3 PCT requirements are the only constraints in the current reference constraint set for which confidence intervals may not be appropriate. The wording of WAPS 1.3 is such that an argument could be made for using *either* a confidence interval for each batch *or* a tolerance interval over an entire waste type. The FTA will use a tolerance interval approach to check compliance with the WAPS 1.3 PCT requirements over an entire waste type. In addition, these requirements will also be tested with confidence intervals within each process batch (i.e., as single-batch requirements), so that the FTA will be checking compliance with WAPS 1.3 during production (not just after). This will add conservatism to the FTA and will serve as a check on the multiple-batch tolerance interval approach.

In the multiple-batch tolerance interval approach, complications arise from the need to incorporate model uncertainty and the need to control the WAPS 1.3 properties sequentially, i.e., as the individual batches are being processed. These complications are discussed in Section 5.3.

5.0 TESTING ACCEPTABILITY

Designing the appropriate statistical test for a given requirement entails answering three questions:

- Is this a direct constraint (i.e., a constraint applied directly to a measured quantity or to a known function of measured quantities) or a constraint on a modelled property? If the former, only composition uncertainty enters into calculation of statistical bounds; if the latter, model uncertainty must also be factored in. See Sections 2 and 3.1 for more information.

- Is this a single-batch requirement or a multiple-batch requirement? If the former, the proper estimate of composition uncertainty is within-batch variability; if the latter, the proper estimate of composition uncertainty must also include batch-to-batch variability. See Sections 2 and 3.1 and Bryan et al. (1994b) for more information on this choice.

- Is the quantity for which inference is to be drawn a single fixed value, a single random value, or some proportion of a statistical distribution? This question relates to the type of statistical interval (confidence, prediction, or tolerance, respectively) to be employed. See Section 4 for more information on this choice.

The first two of these questions are dichotomous, while the last is trichotomous. Therefore, there are twelve (= 2·2·3) possible combinations of answers to these questions. For the current reference constraint set, only three of these combinations occur; the proper statistical tests for these combinations are described in Sections 5.1, 5.2, and 5.3. One of these sections is likely to cover any constraints added in the future. In any case, the principles exhibited in these sections should provide sufficient guidance for constructing statistical tests for added constraints.

For each requirement in the reference constraint set, the FTA will perform the

following series of steps:

- Obtain an estimate of the quantity to be tested. The quantity may be supplied to the FTA (e.g., oxide mass fractions) or may be calculated by the FTA from other inputs.
- Obtain the proper estimate of uncertainty. This entails acquiring or calculating an estimate of variance, then computing the standard deviation (the square root of the variance).
- Choose the appropriate statistical multiplier.
- Use Equations (4) or (5) to produce the required statistical bounds. If a lower limit on the quantity is specified, a lower bound is calculated; if an upper limit is specified, an upper bound is calculated.
- Compare the calculated bound(s) to the corresponding limit(s), and record the results of each comparison. For a suggested list of reported results, see Section 7.2

Sections 5.1, 5.2, and 5.3 concentrate on estimation of uncertainty and the choice of interval type. The choice of statistical multipliers is discussed in Section 6.1.

5.1 CONFIDENCE INTERVALS FOR SINGLE-BATCH DIRECT CONSTRAINTS

The constraint on redox state, the CVS region constraints, the critical component constraints, and the stand-in processability constraints are direct constraints. Therefore, only composition uncertainty enters into testing of these constraints. Since these are single-batch constraints, within-batch uncertainty is the proper estimate of composition uncertainty for these constraints. Finally, in each case, inference is required for a single fixed true value. Thus, confidence intervals provide the proper approach to statistical testing of these constraints.

The first three types of constraints (the constraint on redox state, the CVS region constraints, and the critical component constraints) are applicable directly to measured quantities (redox state and oxide mass fractions). For each of these constraints, the FTA must be supplied with an estimate of composition uncertainty. For example, the estimated variance

for an oxide mass fraction will be extracted from the covariance matrix associated with the estimate of the feed composition. This covariance matrix will be obtained from the existing MEM (see Section 3.3 for a discussion of the MEM).

The remaining direct constraints, the five stand-in processability constraints, are known simple functions of oxide mass fractions. Four of these five are constraints on sums of oxide mass fractions. Computation of the variance of a sum is quite simple: the variance of a sum is the sum of the variances plus twice the relevant covariances. The FTA will extract all the relevant variances and covariances from the input covariance matrix for the feed composition and will use these to construct the variance and the standard deviation of the sum. This standard deviation will be used to construct the confidence bounds.

The remaining stand-in constraint applies to the ratio of two oxide mass fractions. An application of the general method of error propagation given in Section 3.3 shows that the variance of the ratio, σ_r^2, is approximately:

$$\sigma_r^2 \approx \left(\frac{x}{y}\right)^2 \left(\frac{\sigma_x^2}{x^2} + \frac{\sigma_y^2}{y^2}\right) - 2\left(\frac{x}{y}\right)^2 \left(\frac{\sigma_{xy}}{xy}\right)$$

where x and y denote the oxide mass fractions of SiO_2 and Al_2O_3, respectively, and σ_x^2, σ_y^2, and σ_{xy} are the variances and covariance. Once this variance is calculated, it will be used to compute the standard deviation and confidence bounds as above.

5.2 CONFIDENCE INTERVALS FOR SINGLE-BATCH CONSTRAINTS ON MODELLED PROPERTIES

This section covers the testing of a single fixed value for single-batch constraints on modelled properties. Constraints on five modelled properties are covered by this section: viscosity at 1150°C, electrical conductivity at 1150°C, and PCT for Li, Na, and B. (The WAPS 1.3 PCT requirements will also be tested with tolerance intervals for multiple-batch constraints on modelled properties; see Section 5.3.) When liquidus temperature models become available, they will be used as described in this section to test constraints on liquidus

temperature. (It is possible that several liquidus temperature models, corresponding to different primary crystalline phases, may be used. If so, each will be used as described here.)

Since the target of inference for each attribute is a single fixed value (the true attribute value), use of a confidence interval is appropriate. For single-batch constraints, the proper estimate of composition uncertainty is that corresponding to within-batch variability. The required covariance matrix, Σ_x, will be estimated by the MEM and supplied to the FTA.

For constraints on modelled properties, uncertainty must be estimated by propagation of error. This error propagation must account for composition uncertainty and for model uncertainty. The parameters of each property model, β, and the associated uncertainty (expressed as a covariance matrix, Σ_β) must be supplied to the FTA. These quantities will be obtained from the latest CVS results.

From Equation (2), the approximate variance for a modelled property is

$$\sigma_y^2 \approx d_x'\Sigma_x d_x + d_\beta'\Sigma_\beta d_\beta$$

First-order property models are linear in both the oxide mass fractions, x, and the parameters, β, so the simplified Equation (3) applies to properties modelled by first-order equations:

$$\sigma_y^2 \approx \beta'\Sigma_x\beta + x'\Sigma_\beta x$$

This approximate variance will be used to calculate a standard deviation and then the required confidence bounds.

5.3 TOLERANCE INTERVALS FOR MULTIPLE-BATCH CONSTRAINTS ON MODELLED PROPERTIES

Currently the only multiple-batch constraints in the reference constraint set are the WAPS 1.3 requirements, PCT results for B, Li, and Na. These requirements will be checked both on a single-batch basis (Section 5.2) and on a multiple-batch basis. This section describes the multiple-batch testing procedure.

As discussed in Section 4.4, each WAPS 1.3 requirement will be tested using an upper tolerance bound. Both the nominal confidence and content for this tolerance bound will be set at 95%. This should ensure demonstrable compliance with the WAPS 1.3 product consistency requirement: "One acceptable method of demonstrating that the acceptance criterion is met ... would be to ensure that the mean PCT results *for each waste type* are at least two standard deviations below the mean PCT results of the EA glass" (DOE, 1993; italics added). See Bryan et al. (1994a) for a more detailed discussion of testing multiple-batch requirements, with specific reference to the WAPS 1.3 requirements.

Feed composition will vary somewhat among batches in the same waste type, and therefore the calculated property values will also vary. This variability must be monitored to preclude its growing large enough to weaken the ability to statistically demonstrate compliance over the entire waste type. The FTA will achieve this control by the use of a running tolerance bound for each multiple-batch requirement. A record will be kept of the calculated property value, the estimated model uncertainty (see below), and the feed composition for each process batch in the waste type. The calculated value for the current batch will be temporarily added to the database and used to compute a mean and standard deviation for this property to date; this mean and standard deviation will be used to construct a tolerance bound in the standard fashion. If this bound is within the specified limit, the current batch will be deemed acceptable with respect to this requirement. If the batch is deemed acceptable with respect to all other attributes as well (and hence the batch is sent on to the melter), the calculated property value for this batch will be added permanently to the database for this waste type.

Because these requirements apply to modelled properties, the standard deviation to be used in construction of the tolerance bound must account for both composition uncertainty and model uncertainty. Composition uncertainty for a multiple-batch requirement must take into account batch-to-batch variability. This can be estimated quite simply for each new batch by calculating the variance of the recorded property values so far in this waste type. To this composition uncertainty must be added an estimate of model uncertainty. For the i-th batch, model uncertainty, σ_i^2, can be estimated as in Equation (2)

$$\sigma_i^2 \approx \mathbf{d}_\beta' \Sigma_\beta \mathbf{d}_\beta$$

For each new batch, the preliminary FTA will calculate the mean model uncertainty for the waste type so far; this will be added to the estimate of composition uncertainty (the variance of the recorded property values), and the square root of the sum will be used as the required standard deviation. This method of incorporating model uncertainty for multiple-batch requirements is somewhat *ad hoc*, and its effect on the performance of the statistical test should be investigated during testing of the FTA.

This testing procedure runs a slight risk that a few aberrant batches at the beginning of processing of a waste type might skew the results for the rest of the waste type. If this appears to be a problem in testing of the FTA or during plant operations, there are several possible solutions to the problem. Stricter requirements might be imposed on the first batches in a waste type, process monitoring algorithms might be designed to scrutinize these batches, and/or a prior estimate of uncertainty might be included in the estimation of overall uncertainty in the calculated property value (for example, by forming a weighted sum of this prior estimate and the composition and model uncertainties discussed above; the weight assigned to the prior estimate should decrease as more batches are processed).

6.0 TECHNICAL ISSUES AND POSSIBLE MODIFICATIONS

In this section, certain technical issues involved in the problem of testing acceptability of feed compositions will be raised. Many of these issues remain open, largely due to inadequate knowledge of the HWVP process and the data arising therefrom. For this reason, the FTA described in this document must be considered preliminary.

The FTA will be tested by implementation and incorporation into the Plant Simulation Code (PSC), where it will be used to "test" the results of simulated HWVP runs. This testing will assist in resolving some of the technical issues and in identifying worthwhile modifications to the FTA. More on the role and application of the PSC to testing of PCC algorithms appears in Bryan and Piepel (1993).

6.1 CHOOSING STATISTICAL MULTIPLIERS

Construction of each of the types of statistical intervals discussed in Section 4 requires a statistical multiplier. Several considerations enter into the choice of a statistical multiplier. These multipliers are essentially percentiles of certain statistical distributions. The proper distribution from which the multiplier should be drawn is affected by the assumed statistical distribution of the data and the type of interval being constructed. The statistical intervals to be used in the FTA assume that the underlying data follow a normal (Gaussian) distribution. The role of normality is discussed in Section 6.2. Given normally-distributed data, multipliers for confidence intervals are usually drawn from the family of central t distributions, while multipliers for (one-sided) tolerance intervals are usually drawn from the family of noncentral t distributions. The particular t distributions depend on the *degrees of freedom* available for estimation of uncertainty. The degrees of freedom associated with uncertainty estimates are essentially measures of the strength of information about the uncertainties. Larger degrees of freedom correspond to more information, smaller multipliers, and shorter intervals. In simple cases (e.g., estimating variance from replicate measurements of the same quantity), the number of degrees of freedom is related to the number of observations used to construct the uncertainty estimate. The situation is more complicated if several sources of uncertainty are

combined into a single overall estimate. The number of degrees of freedom associated with the combined uncertainty estimate is affected by the relative sizes and degrees of freedom associated with each of the constituent uncertainties. The FTA will use Satterthwaite's approximation to derive the degrees of freedom associated with a combined uncertainty estimate. Bryan et al. (1994b) discuss this approximation in more detail.

Central t distributions corresponding to large degrees of freedom look very much like the standard normal distribution (a normal distribution with mean zero and variance one). (In fact, as degrees of freedom increase, the central t distributions converge to the standard normal distribution.) For this reason, if the number of degrees of freedom associated with the uncertainty estimate is large, the normal distribution provides an adequate approximation to the central t distribution, and hence confidence intervals may be constructed with multipliers drawn from the standard normal distribution (which is easier to compute than are the central t distributions). Similarly, if a confidence interval is to be constructed from a prior estimate of uncertainty, and the prior information is quite good, multipliers drawn from the standard normal distribution will suffice.

As mentioned above, type of interval and degrees of freedom affect the choice of statistical distribution from which a multiplier is drawn, and the multiplier itself is simply a percentile of this distribution. The percentile is related to the confidence level to be associated with the interval. Larger confidence levels correspond to larger percentiles and hence to wider intervals. As discussed in Section 4.4, for a fixed sampling effort, higher confidence levels correspond to increased probability of rejecting acceptable batches. Thus, a compromise must be reached between confidence level, the probability of rejecting acceptable batches, and the cost of sampling and analysis. Confidence levels should not automatically be set to 95%, 99%, or any other level. Consideration should be given to scaling the required confidence level to the risk or importance of individual attributes.

Another consideration in the choice of statistical multiplier is the nature of the required bounds -- one-sided vs. two-sided. Consider first the case for confidence intervals,

i.e., for testing the behavior of a single fixed (but imperfectly known) true value. Clearly a one-sided interval is appropriate for testing an attribute subject to only one limit (upper or lower), but it may be somewhat surprising that one-sided intervals are also appropriate for testing an attribute subject to *both* a lower limit and an upper limit. This feature stems from the fact that it is impossible for a single fixed value (the true value of the attribute) to simultaneously violate a lower limit and an upper limit (provided of course that the limits themselves are consistent, i.e., that the lower limit is indeed less than the upper limit). Thus, in acceptance testing using confidence intervals, only one-sided intervals are required. This is not the case for tolerance intervals; however, the current reference constraint set contains only upper limits on attributes to be tested with tolerance intervals (the WAPS 1.3 PCT requirements for B, Li, and Na), for which one-sided intervals are appropriate. Thus, for the current reference constraint set, the FTA will employ only one-sided intervals.

6.2 THE ROLE OF NORMALITY

Implicit in the foregoing discussion of statistical intervals is the assumption of normality, i.e., that the data or the random quantities being bounded follow a normal (Gaussian) distribution. This assumption should be examined.

The individual components of a composition cannot be normally distributed. The major reason is that each component of a composition is constrained to fall in the interval [0,1], while all normal distributions put positive probability outside this interval. However, if the standard deviation of the true distribution of a single component is small relative to the mean value, it is quite possible that the normal distribution may provide a good approximation to the true distribution. If estimates of composition are based on several measurements for each batch, the Central Limit Theorem (Lindgren, 1976, pp. 157-159) guarantees that the means of the individual measurements will more closely follow the normal distribution than will the individual observations. It is certainly possible, therefore, that statistical techniques based on the normal distribution will perform reasonably well. Once adequate knowledge is gained of the statistical behavior of the types of compositional data arising from the HWVP process, the performance of normal-based techniques can be more

thoroughly investigated.

Arguments based on generalizations of the basic Central Limit Theorem suggest that property values calculated from the types of models being developed by CVS may also mimic the normal distribution and, again, that statistical techniques based on the normal distribution will perform reasonably well. This expectation is borne out by preliminary simulation results. More such studies should be performed with updated CVS models and as more is learned of the covariance structure associated with the HWVP feed compositions.

6.3 INDIVIDUAL AND SIMULTANEOUS STATISTICAL CONFIDENCE

As discussed in Section 4.0, the confidence associated with a statistical interval controls the probability that the interval contains the true value[f]. When multiple statistical intervals are constructed, two types of statistical confidence may be considered. For each individual interval, there is a probability (confidence) that this interval contains the corresponding true value. This type of confidence will be referred to as the *individual confidence level.* In addition, there is a probability (confidence) that *all* the intervals contain the corresponding true values simultaneously; this will be referred to as the *simultaneous confidence level.* The simultaneous confidence level associated with a group of intervals cannot be greater than the smallest individual confidence level, and, in fact, the simultaneous confidence level may be much lower than any of the individual confidence levels. For example, if 30 confidence intervals are produced, each with an individual confidence level of 95%, *and each of the attributes under examination is independent of all other attributes*, then the simultaneous confidence level, i.e., the probability that all intervals trap the corresponding true values, is $0.95^{30} = 0.21$ (21%). In other words, if the set of 30 confidence intervals is produced a large number of times (from separate data each time), then in only 21% of the sets of intervals will all the intervals within the set successfully trap the true values. In 79% of the sets of intervals, at least one interval fails to trap the true value. This can be a serious

(f) Again, confidence actually controls the probability that the procedure produces an interval containing the true value, *not* the probability that any particular interval traps the true value. This distinction is not important to the present discussion, so the more concise wording will be used.

problem if the failure of one or more intervals in each set is of concern.

Since statistical intervals will be used for acceptance testing by the FTA, the problem of simultaneous confidence must be addressed. At least two types of simultaneous confidence may be considered in the context of acceptance testing for a series of batches: simultaneous confidence over requirements for a single batch, and simultaneous confidence over batches for a single requirement.

In order to understand the simultaneous confidence associated with testing all requirements for a single batch, it is critical to understand the nature of each individual test. Consider, for example, the test that viscosity at 1150°C is less than 10 Pa·s. As discussed in Section 4.1, using a 95% upper confidence bound to test this attribute means that, when viscosity is greater than 10 Pa·s (i.e., when viscosity is "bad"), the probability of falsely concluding that viscosity is less than 10 Pa·s (i.e., viscosity is "good") is no greater than 5%. Thus, for this method of acceptance testing, the individual confidence level of a given test controls the probability of concluding that an attribute is good when in fact it is bad. In other words, the individual confidence level controls the probability of accepting a bad attribute.

For testing all requirements in a single batch, simultaneous confidence should control the probability of accepting a bad batch (a batch with one or more bad attributes). The FTA will reject (or at least call attention to) a batch if even a single one of the various attributes is rejected. Given this approach, the probability of accepting a bad batch is no greater than the probability of accepting a single bad attribute, i.e., *the simultaneous confidence level is at least as large as the smallest of the individual confidence levels.* (In fact, correlation among the attributes may significantly raise the simultaneous confidence level, but that is beyond the scope of this discussion.) Thus, the FTA does not need to make special provisions for controlling simultaneous confidence over all requirements for a single batch.

Simultaneous confidence over batches for a single requirement requires only replacement of the statistical multipliers mentioned in Section 6.1 by multipliers designed to

guarantee simultaneous confidence. In many cases, multipliers that guarantee simultaneous confidence are much larger than those required for individual confidence levels. This implies that the chance of accepting a bad batch is reduced, but with the side-effect of an increased rate of rejection of acceptable batches (see the discussion in Section 4.4). Therefore, the preliminary FTA will not implement simultaneous confidence over batches; the need for simultaneous control can be investigated as part of testing of the FTA.

6.4 MODEL UNCERTAINTY AND THE QUALIFIED COMPOSITION REGION

For testing constraints on modelled properties, the preliminary FTA will combine composition and model uncertainties into an overall uncertainty estimate and then use this overall uncertainty estimate to set confidence bounds, which will then be compared to limits on property values. In this approach, model and composition uncertainties are combined and associated with the confidence bounds for the estimated property value. This is not the only possible way to deal with model uncertainty. Another approach would be to treat model and composition uncertainties separately, and to associate composition uncertainty with the estimated property value and model uncertainty with the limits on the property value. To illustrate the difference, assume that we wish to use an upper confidence bound (UCB) to compare the estimated property value with an upper limit, U. In the preliminary FTA, this comparison will be done by constructing the UCB using a combined estimate of uncertainty, as in Equation (4):

$$UCB_c = \hat{y} + (m_c \cdot s_c)$$

where, from Equation (2)

$$s_c^2 = \mathbf{d}_x'\Sigma_x\mathbf{d}_x + \mathbf{d}_\beta'\Sigma_\beta\mathbf{d}_\beta$$

Here, UCB_c denotes the upper confidence bound based on the combined estimate of uncertainty, s_c denotes the standard deviation derived from the combined uncertainty estimate, and m_c is the statistical multiplier appropriate for this uncertainty estimate (the other symbols are defined in Section 3.3). This UCB_c will then be compared directly to the upper limit, U, as discussed in Section 4.4. The alternate approach discussed here would compute UCB_x and U_m:

$$UCB_x = \hat{y} + (m_x \cdot s_x)$$

$$U_m = U - (m_m \cdot s_m)$$

where

$$s_x^2 = \mathbf{d}_x' \Sigma_x \mathbf{d}_x$$

$$s_m^2 = \mathbf{d}_\beta' \Sigma_\beta \mathbf{d}_\beta$$

Here, UCB_x is the confidence bound taking into account only composition uncertainty (propagated through the property model and expressed in property units as s_x), U_m is the limit on the property value, modified to incorporate model uncertainty (estimated by s_m), and m_x and m_m are the statistical multipliers appropriate for the separate estimates of uncertainty. UCB_x would then be compared with U_m, just as UCB_c is to be compared with U.

The approach based on separate estimates of composition and model uncertainties has certain advantages. Since two statistical multipliers are used, the confidence level associated with composition uncertainty need not be the same as that associated with model uncertainty. In fact, it would be possible to use a simultaneous confidence approach for one type of uncertainty and an individual confidence approach for the other. In addition, this approach makes clear the relationship between the FTA and the Qualified Composition Region (QCR), which is being developed as part of the CVS work (see Hrma, Piepel, et al., 1992, for more discussion of the QCR). The QCR is intended to include all compositions that are acceptable *given model uncertainty*. Briefly, there exists some multidimensional space of compositions that are acceptable, i.e., for which all true attribute values conform to the requirements imposed on HWVP material. However, this region is not perfectly known, owing to uncertainties in the models used to estimate property values. Thus, the space of acceptable compositions must be "shrunken" to eliminate compositions for which one or more requirements might be violated, due to imperfections in the models. Thus, the QCR consists of all compositions for which all estimated property values, $\hat{y}_k$, fall within the limits defined by the various $U_{m,k}$, where k indexes the requirements imposed on HWVP. The approach to testing acceptability of compositions based on separate estimates of composition and model uncertainty can be thought of as testing whether, given composition uncertainty, it is

reasonable to conclude that the true composition lies within the QCR. (Actually, the QCR may be constructed with simultaneous intervals, while the preliminary FTA will use only individual intervals, but this simply means that different statistical multipliers will be employed; the analogy holds).

The approach based on separate estimates of composition and model uncertainty is likely to be more conservative (i.e., higher probability of rejection of acceptable batches) than that based on a single combined estimate. The preliminary FTA will employ the latter. Once the performance of the preliminary FTA has been investigated, the need for the conservatism of the alternate approach can be evaluated.

6.5 THE BAYESIAN APPROACH

One annoying feature of the standard (frequentist) statistical tests discussed above is the cumbersome interpretation of confidence, the frequentist measure of degree of belief. As discussed in Section 4.0, it is possible to misinterpret the statement "with 95% confidence, this batch is acceptable" as "the probability that this batch is acceptable is 95%." Bayesian statistical methods are designed to produce statements of the second type in a rigorous manner. In addition, Bayesian methods can be used when frequentist methods suffer from lack of data (which may well be the case for HWVP; see Bryan et al., 1994b), and Bayesian methods can be used to smooth data and update existing estimates (e.g., of uncertainty). Of course, there is a price to pay for these seemingly more easily interpretable and effective results -- in order to perform Bayesian analyses, the user is required to provide rigorous information on prior knowledge (indeed, Bayesian analysis with data lacking essentially consists of substituting prior knowledge for data). Still, if reasonable statements of prior knowledge can be obtained, there may be a place for Bayesian methods in the HWVP PCC. This may be especially true in the early stages of operation, when data from actual operation will be scarce.

6.6 MISCELLANEOUS ISSUES

As mentioned in Section 2, the framework for testing acceptability of process batches that has been established in this document should be flexible enough to accommodate any additions to or changes in requirements imposed on the HWVP process. Modification of models and updating of model covariance information will be as simple as modifying input files or FORTRAN DATA statements. Incorporation of new constraints requires only the classification of the new constraint into one of the categories described in Section 5.0 and modification of the corresponding input files. If theoretical models for liquidus temperature are developed by CVS, some additional work may be required.

As mentioned in Section 6.1, varying confidence levels can be used to weight constraints according to their relative importance to process/product performance. Methods other than statistical tests with varying confidence levels (e.g., penalty functions rather than strict limits for non-critical attributes) may be more appropriate in some cases.

7.0 FTA INPUTS, OUTPUTS, AND SUPPORTING ALGORITHMS

The lists provided here of inputs, outputs, and supporting algorithms for the FTA are preliminary -- they will be modified during implementation of the FTA in the PSC and as the implementation of the FTA evolves to reflect accumulation of knowledge about the HWVP process.

7.1 INPUTS TO THE FTA

Among the items certain to be required by the FTA are:

- estimated feed composition and relevant covariance matrix (or components);
- the desired confidence coefficient for each attribute;
- the desired content for each tolerance interval;
- the vector of estimated parameters, the covariance matrix, mean squared error, and

associated degrees of freedom for each model obtained from CVS; and

- the vectors of calculated property values, compositions, and model uncertainties for previous batches in the current waste type (one set of vectors for each multiple-batch requirement).

If oxide mass fractions are estimated from more than one sample per batch or more than one analysis per sample, the numbers of samples and analyses should be made available to the FTA. If models nonlinear in either the parameters or the data are to be used for some properties, derivatives of these models (or an algorithm to obtain these derivatives) must be available to the FTA.

7.2 OUTPUTS FROM THE FTA

The possible outputs from the FTA are numerous; the actual set of outputs will be determined from the requirements of the PSC and the testing to be performed with this software. At a minimum, the FTA must return a flag for whether the current batch is deemed acceptable. Possible additional outputs include:

- a list of the current reference constraint set, relevant statistical bounds for each attribute, and a score vector flagging any violated constraints;
- for violated constraints, the distance of the statistical bound from the limit, in absolute terms and in terms of standard deviation units; and
- for an acceptable batch, the attribute (or attributes) coming closest to violating constraints.

7.3 SUPPORTING ALGORITHMS

The FTA will require routines for various matrix and vector manipulations. Several of these already exist in the PSC. As mentioned in Section 7.1, for testing multiple-batch requirements, access to the calculated property values, compositions, and model uncertainties for previous batches in the current waste type will be required, as will the capability to update these databases. Also, if models nonlinear in either the parameters or the data are to be used

for some properties, derivatives of these models (or an algorithm to obtain these derivatives) must be available to the FTA.

Several statistical multipliers will be required by the FTA. These multipliers might be provided as inputs (in the form of tables to be consulted by the FTA), or algorithms to calculate the multipliers as needed might be provided (either from code in the PSC or from a standard mathematical/statistical subroutine library). Among the statistical distributions from which multipliers (i.e., percentiles) might be required are:

- the normal (or Gaussian) distribution, for use in constructing confidence intervals when the relevant standard deviations are well known or the sample sizes are quite high;
- the central t distribution, for use in constructing confidence intervals when the relevant standard deviations are not well known or the sample sizes are relatively low; and
- the noncentral t distribution, for calculation of one-sided tolerance intervals.

When various estimated sources of uncertainty are combined into a single pooled estimate of uncertainty, the FTA must be able to compute an estimate of the degrees of freedom associated with this pooled estimate. A general implementation of the Satterthwaite approximation is desirable, since this requirement will surface at several places in the PSC as more statistical algorithms are added.

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