

STATISTICAL PROCESS CONTROL: AN APPROACH TO QUALITY ASSURANCE  
IN THE PRODUCTION OF VITRIFIED NUCLEAR WASTE

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ABSTRACT

Current planning for liquid high-level nuclear wastes existing in the United States includes processing in a liquid-fed ceramic melter to incorporate it into a high-quality glass, and placement in a deep geologic repository. The nuclear waste vitrification process requires assurance of a quality product with little or no final inspection. Statistical process control (SPC) is a quantitative approach to one quality assurance aspect of vitrified nuclear waste. This method for monitoring and controlling a process in the presence of uncertainties provides a statistical basis for decisions concerning product quality improvement. Statistical process control is shown to be a feasible and beneficial tool to help the waste glass producers demonstrate that the vitrification process can be controlled sufficiently to produce an acceptable product. This quantitative aspect of quality assurance could be an effective means of establishing confidence in the claims to a quality product.

INTRODUCTION

The United States Department of Energy (DOE) is managing several programs to dispose of existing liquid high-level nuclear waste by converting them to a high-quality glass in a liquid fed ceramic melter (LFCM) and placing the products in a deep geologic repository. DOE has instituted a waste-acceptance process to specify the documentation and activities required to ensure that waste forms will be acceptable at any of the potential repositories. The Waste Acceptance Committee has been established to implement the waste acceptance process. This committee has drafted a report dealing with waste acceptance preliminary specifications (WAPS). The waste form producers will demonstrate compliance to the WAPS in a waste form qualification report.

During FY 1982, the DOE assigned responsibility for managing civilian nuclear waste treatment programs in the United States to the Nuclear Waste Treatment Program (NWTP) at Pacific Northwest Laboratory (PNL)<sup>(a)</sup>. A principal program objective is to establish relationships between vitrification process control and glass-product quality. Users of the liquid-fed ceramic melter (LFCM) process will need such relationships in order to establish compliance with the waste acceptance specifications to qualify the waste for repository disposal. The NWTP proposed that compliance to specifications

pertaining to the waste form be shown primarily by demonstrating that a certain glass composition is approached; therefore, the uncertainty in the composition and in the process behavior that can affect the composition must be well understood. Understanding, monitoring, and controlling the uncertainties affecting the product composition is important to assuring a quality product.

In the manufacturing industry, product quality is often determined through contact inspection of the completed product. Destructive examination is often required. This method of quality assurance is not reasonable for a process that manufactures canisters of vitrified high-level nuclear waste. A desirable and feasible alternative is to correlate the glass-product quality with information obtained during monitoring and control of the process. This information, coupled with a limited number of glass samples obtained from the pour stream as the canisters are filled, would provide a measure of product quality. Quality assurance not only entails accurate documentation and strict adherence to procedures, but also includes continual monitoring and control of the vitrification process in the presence of uncertainties. Actions taken to improve the product quality must be based on sound decision rules that account for the uncertainties associated with the entire vitrification process.

Statistical process control (SPC) is a method for monitoring and controlling a process in the presence of uncertainties. It can provide the information necessary to determine a quantitative

(a) Operated by the U.S. Department of Energy (DOE) by Battelle Memorial Institute Under Contract DE-AC06-76RLO 1830.

measure of product quality and the reliability of such a measure. It provides a statistical basis for decisions concerning product quality improvement. SPC is a widely applied and accepted industrial manufacturing method of quality control. In this application, it can provide a mechanism for detecting unacceptable process changes affecting glass quality early in the production cycle - early enough that corrective adjustments can be made to avoid the production of substandard glass. SPC can be an important means of demonstrating in a Waste Form Qualification Report that a vitrification process can be controlled sufficiently to produce an acceptable product, and that when produced, the product will be characterized sufficiently to compare it to the WAPS. SPC could be a particularly effective means of establishing confidence in the quality of waste glass outside those close to the technology.

#### Fundamentals of Statistical Process Control

Basic to SPC is the concept that measurements or characterizations of any manufactured product (or manufacturing process) exhibit inherent variation because of unintentional process changes and random mechanisms. Even when an attempt is made to control the process, ongoing measurements will reveal this inevitable process variation even though all components are operating properly. Usually there are many sources of variation which affect a process. "Common cause variations" refer to the many sources of variation within a process that is in statistical control. All combined common-cause variations form a pattern that can be characterized by some distribution. "Special-cause variations" are not part of the process and appear in unpredictable ways. In the LFCM process, for example, a common-cause variation might be the inherent lack of precision associated with the mechanism used to measure the level of the material in a tank; a special-cause variation may be due to a plugged pipe.

Only when a process is in statistical control (no special-cause variations present) can the level and variability of the process parameters be predicted; statistical control is the key to predicting waste glass quality characteristics. SPC aids in eliminating the special causes of variation, thereby bringing the process into statistical control. This refinement provides predictability to waste glass characteristics. Once the process is in control, SPC aids in monitoring the process for any unacceptable changes.

Many of the existing SPC tools and techniques could be applied to the LFCM process. Only one of these tools, the Shewhart control chart, will be presented in this demonstration of SPC applied to the LFCM process (Ref. 1). Control charts provide a means for monitoring a process parameter and determining whether it is in statistical control or fluctuating beyond its expected inherent variation. The statistical theory supporting control charts is based on the central limit theorem and normal distribution theory. The central limit theorem states that the form of the probability distribution of sample means approaches the form of a normal probability distribution as the size of the sample is increased, regardless of the

distribution describing the individual samples used to calculate the sample means. For a normal distribution, 68% of randomly selected values would be expected to fall within  $\pm 1$  standard deviation of the mean, about 95% within  $\pm 2$  standard deviations, and 99.7% within  $\pm 3$  standard deviations. The Shewhart control chart establishes control limits at  $\pm 3$  standard deviations from the mean of a process variable being monitored. Figure 1 depicts the relationship between the normal distribution and a control chart. When measured values fall outside these limits, it is taken as evidence that those process fluctuations are not part of the random fluctuations inherent in the process; the process has been altered and is determined to be out of statistical control.

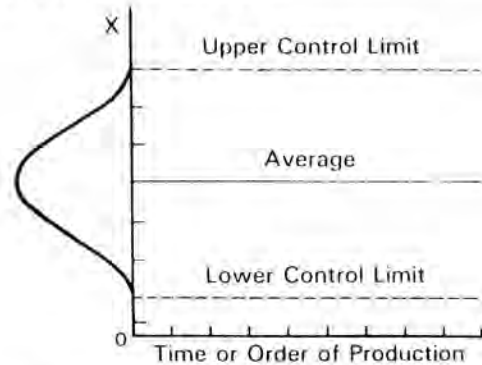
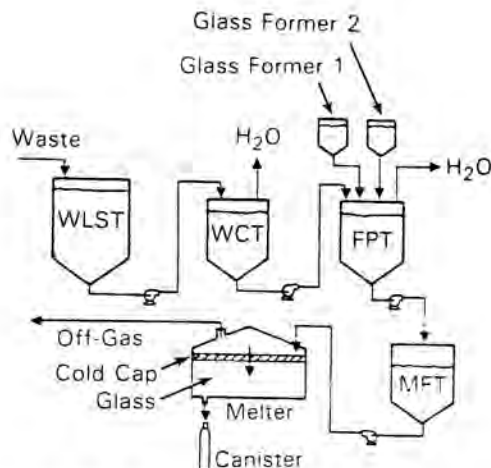


Fig. 1. Illustration of the Theoretical for a Control Chart

The control chart is a method for determining whether process fluctuations are 1) due to inherent process variability and require no special action by the operator or management, or 2) result from special-cause variabilities that should be examined, isolated, and corrected to bring the process back into control. Isolating and eliminating special-cause variability is vital to quality assurance in the LFCM process. Waste glass characteristics (product quality) can be reliably predicted from operational data only when the process is in statistical control. If the LFCM users can demonstrate that their process is in statistical control, the quality of the product will be more easily demonstrable.

#### Monitoring the Composition of the Material Fed into the Melter

To demonstrate the use of SPC on the LFCM system, the LFCM process is defined and simulated. Figure 2 presents a hypothetical LFCM system. The flow of material through the system is as follows. Dilute waste slurry is periodically batched to the waste lag storage tank (WLST). It is transferred to the waste concentration tank (WCT), where the slurry is concentrated by evaporation to less than one-fourth the original volume. Once the slurry has been adequately concentrated, it is transferred to the feed preparation tank (FPT), where glass formers are added and additional evaporation occurs. After the slurry has been adequately concentrated in the FPT, it is transferred to the melter feed tank (MFT). From there the slurry is continuously fed into the melter.



WLST: Waste Lag Storage Tank  
 WCT: Waste Concentration Tank  
 FPT: Feed Preparation Tank  
 MFT: Melter Feed Tank

Fig. 2. Flow Diagram of a Hypothetical LFCM Vitrification System

To determine the quantity of glass formers to add to the waste constituents in the FPT, the waste composition must be determined. Each WLST batch is sampled and analyzed. Samples will also be obtained from the combined waste and glass-former feed slurry in the FPT to characterize the feed composition. The accuracy and precision of composition estimates of the slurry in the WLST and the FPT depends on how many samples are taken and how many analytical determinations are made on each sample.

If demonstration of product quality depends partly upon demonstrating a stable process, one obvious place to implement SPC would be on the composition of the material being transferred to the MFT. Assume that durability tests of glasses with various compositions have resulted in a target feed composition. If the LFCM operators can demonstrate that the feed composition is in statistical control centered at the target composition with acceptable variation, demonstration of product quality will be greatly enhanced.

Three major sources of common-cause variability affect the apparent FPT average composition. Sample-to-sample differences provide one source. Some variability exists among replicate samples taken from the same FPT batch. This source may be reduced by adequate mixing in the tank, but variabilities associated with the preparation of analytical samples will still exist. The second source of variation is analytical variability. Differences will exist between replicate analyses of the same sample. The third source is inherent process variability, caused by variations in such factors as tank-level measurements, frit additions, and the measured WLST composition (which drives the frit addition). Under normal operations, the magnitude of these variations should remain constant (in control). If changes beyond this expected normal variation occur, the process could be deemed out of control, requiring a corrective action.

The combined effect of all sources of variability contributing to the overall process variation is difficult to quantify. Process variability may best be estimated by running the process or by simulating a run of the process when it is in statistical control. An attempt has been made to simulate the LFCM process and include all major sources of variation that contribute to the overall process variation (Ref. 2). Estimates of sampling and analytical variation were obtained from laboratory data of samples taken from a previous run of a pilot-scale LFCM and used in our simulation. SPC is demonstrated in this paper using the simulated data resulting from this work.

For illustrative purposes, the simulated data of one 4000 hour melter run was used to construct a control chart for the boron content of the feed composition (see Fig. 3). None of the simulated observations (with one possible exception) fall outside the control limits because the simulation was set up to be a process in statistical control. The sampling and analytical standard deviations were each assumed to be 4% of the target boron concentration, consistent with previous experience with similar LFCM tests. Pulsipher and Kuhn<sup>(a)</sup> present a more detailed treatment of the statistical properties and formulas relating to the application of SPC to the LFCM process.

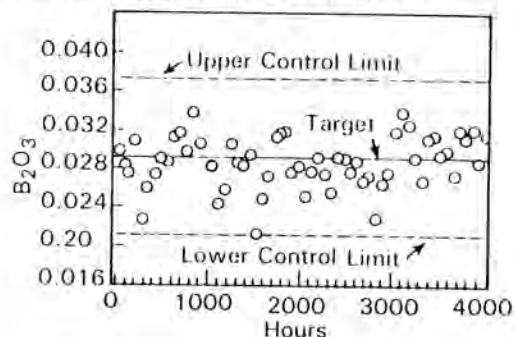


Fig. 3. Control Chart of Simulated Boron Feed Composition

By implementing SPC, a LFCM operator would have the tools to demonstrate a stable, predictable process. If the control chart indicated statistical control as shown in Fig. 3, predictions of product quality using the information about the estimated mean and variance of the feed samples would be appropriate. However, out-of-control batches suggest an unstable process; neither the variability nor the composition is well characterized. The cause for the out-of-control batches must be determined, and the feed composition must be corrected (or abandoned by sending it back to the waste tank) in order to bring the system back into statistical control. A statistically controlled system would help defend the reliability of predictions of product quality.

Two topics that require further consideration are (1) the capability of the control charts to detect shifts or changes in the process that may affect the quality of the waste glass and (2) the use of SPC in decisions concerning adjustments to the feed composition. Each of these topics is discussed in detail below.

(a) Pulsipher, B.A., and W.L. Kuhn. 1987 Statistical Process Control Applied to the Liquid-Fed Ceramic Melter Process. PNL Letter Report (in process), Pacific Northwest Laboratory, Richland, Washington.

Average Run Lengths

Comp.	WLST		$\pm$ (a)	Avg. Run Lengths (d)			
	Samp.	Anal.		NS (b)=1			
				NA (c)=1	NA=3	NA=1	NA=3
B <sub>2</sub> O <sub>3</sub>	1	1	10	>20	>20	>20	>20
B <sub>2</sub> O <sub>3</sub>	1	1	15	>20	14	7	7
B <sub>2</sub> O <sub>3</sub>	1	1	25	3	2	2	2
B <sub>2</sub> O <sub>3</sub>	5	5	10	>20	>20	>20	11
B <sub>2</sub> O <sub>3</sub>	5	5	15	19	7	3	3
B <sub>2</sub> O <sub>3</sub>	5	5	25	1	1	1	1
SiO <sub>2</sub>	1	1	10	>20	>20	>20	>20
SiO <sub>2</sub>	1	1	15	>20	>20	>20	18
SiO <sub>2</sub>	1	1	25	8	6	4	5
SiO <sub>2</sub>	5	5	10	>20	>20	>20	>20
SiO <sub>2</sub>	5	5	15	>20	15	9	6
SiO <sub>2</sub>	5	5	25	2	2	2	1
Na <sub>2</sub> O	1	1	10	>20	>20	>20	>20
Na <sub>2</sub> O	1	1	15	>20	>20	>20	>20
Na <sub>2</sub> O	1	1	25	8	6	4	4
Na <sub>2</sub> O	5	5	10	>20	>20	>20	>20
Na <sub>2</sub> O	5	5	15	>20	13	8	7
Na <sub>2</sub> O	5	5	25	2	2	2	2

- (a) Percent shift is represented as a percent of the target value.
- (b) NS is the number of samples obtained from each FPT batch.
- (c) NA is the number of analyses performed per FPT sample.
- (d) Large ARL values are not very accurate because the simulation contained only 1000 FPT batches.

The quality control capability of control charts refers to their ability to detect shifts in the system. If the feed composition has shifted because of some failure or change in upstream processing, that shift should be detected quickly if it is large enough to significantly affect the glass quality. The control capability is affected by several factors: 1) the number of samples obtained from the WLST and the FPT, 2) the number of analyses per sample from the WLST and FPT, 3) the magnitude of the process variation including sampling and analysis variability, and 4) the magnitude of the process shift.

A measure of control chart capability is the average run length (ARL). The ARL is the average number of observations that are plotted on a control chart after a process shift before an out-of-control point is detected. A small ARL suggests that the control chart is capable of detecting a shift in the process quickly.

For demonstration purposes, results obtained from the simulated process mentioned above, coupled with previous estimates of sampling and analytical variations, were used to examine the average run length of control charts for monitoring some of the major components in the feed. The ARL was examined for three shift sizes, four combinations of the number of samples and analyses per sample from the FPT, and two sampling schemes from the WLST. A summary of the ARLs for the control chart on FPT batch averages is presented in Table I. As an example, if the assumed variabilities and process models were correct, and five samples were obtained from the WLST with five analyses conducted on each sample, a 25% shift in the boron concentration would be detected (on the average) on the first FPT batch after the shift. If five samples were obtained from the WLST with five analyses per sample and three samples were obtained from the FPT with only one analysis per sample, it would take three FPT batches on the average before detecting a 15% shift in boron concentration.

The quality of the waste glass is affected by changes in the composition. The relationship between compositional changes and product quality can be used to identify unacceptable changes. Short ARLs for unacceptable changes are desirable. Because the ARL is dependent upon the WLST and FPT sampling schemes, various sampling schemes can be studied to determine the best sampling scheme for detecting significant shifts. If the ARL is unacceptable for every economically feasible sampling scheme, effort should be focused on eliminating or decreasing some of the major sources of process, sampling, or analytical variation. Through simulation, a sensitivity analysis can be conducted to determine the major contributors to the process uncertainty.

Without an understanding of the control chart capability through an examination of the ARLs, statistical quality control through control charting may be entirely useless in detecting compositional shifts that affect waste glass quality. If the sample sizes are inadequate because of the magnitude of the sampling and analytical variations or because the variability of the process is too large, the process may appear to be in statistical control even when the waste glass quality is poor. Thus, control chart capability is an important factor to consider in demonstrating adequate quality assurance of the waste glass.

SPC's Role in Quality Improvement Through Feed Composition Adjustments

Both special and common cause variations can enter the LFCM system in many ways. The process might consistently produce an acceptable product if the inherent variation is sufficiently small. As discussed above, one method for improving the capability is to decrease the major sources of variation. This may prove difficult or expensive. In this section, another quality improvement method is discussed. This method involves adjusting the feed based on the results of the initial FPT sample analyses.

As shown earlier, there are three major sources of common-cause variation affecting the apparent feed composition. The first is the process variation, a measure of the spread of the true FPT composition about the target composition. The other two sources are sampling and analytical variations. A plausible method for decreasing the difference between the true feed composition and the target feed composition (reducing the process variation) is to allow for a feed composition adjustment after the initial addition of glass formers. The feed in the FPT could be held until the results of the analysis of FPT samples from that batch are available. If the analysis shows a significant departure from the target composition, adjustments to the feed composition are made before sending that batch to the MFT. Adjusting the feed composition can eliminate or significantly decrease the process variation by adjusting the true FPT composition to be nearer the target composition.

Assume that the adjusting procedure is exact. If there were no uncertainty associated with the

determinations of the composition of each FPT batch, one would always adjust unless the composition was already on target. However, the decision of whether to adjust the FPT composition is clouded because there is uncertainty in the estimate of the composition, due to the sampling and analytical variations. Any one of three decision rules for adjustment could be imposed: 1) ignore the sampling and analytical variation and always adjust, 2) account for the sampling and analytical variation and sometimes adjust, and 3) never adjust.

The sampling and analytical uncertainty can be accounted for if the decision rule is based on SPC techniques. Adjusting should be done only when there is a high probability that the true composition is different from the target composition. A control chart centered at the target composition could be established to detect differences between the target composition and the apparent composition that are greater than would be expected given the sampling and analytical uncertainties. An apparent FPT composition outside the control limits would indicate that the true composition is most likely different from the target composition and that adjustment is necessary. If the apparent composition were to fall within the limits, one could not be certain whether or not the true FPT composition is different from the target composition, and no adjustment should be made.

Always adjusting may not only prove costly, but in some cases this rule would actually increase the process variation rather than decrease it. An extreme example magnifies the problem. Suppose that there is no process variability; the true FPT composition is always on target. Because of sampling and analytical variation, the apparent FPT composition would nearly always be different than the target composition. The FPT composition would be unnecessarily adjusted, thereby increasing the process variation because the adjustment moved the true composition away from the target composition. Although this example is exaggerated, a similar increase in the process variation would result whenever the initial process variance is small relative to the combined sampling and analytical variance. This fact is further demonstrated below.

The performance of the three decision rules were compared by simulating boron data with varying sampling and analytical strategies. Several values for the initial process variance were examined. The results of these simulations are shown in Fig. 4. The process variation is represented as a standard deviation relative to the target boron concentration. The vertical axis represents the true process relative standard deviation after the adjustment rule has been applied (RSDA). The horizontal axis is the true process relative standard deviation before adjusting (RSDB). When there is no adjustment, the RSDB and RSDA are the same (producing a straight line on the plots). When the composition is always adjusted, the process variation is increased rather than decreased if the RSDB is less than the combined sampling and analysis relative standard deviation; otherwise, the process variation is decreased. The decision rule based on SPC protects against adjusting when it would only inflate the process variance. Furthermore, the rule protects against not adjusting when adjustment actually is needed. This attribute is illustrated by 1) the similarity of the RSDA values for the never-adjust and SPC-based rules when the RSDB is low and 2) by the similarity of the RSDA for the

always-adjust and SPC-adjust rules when the RSDB is high.

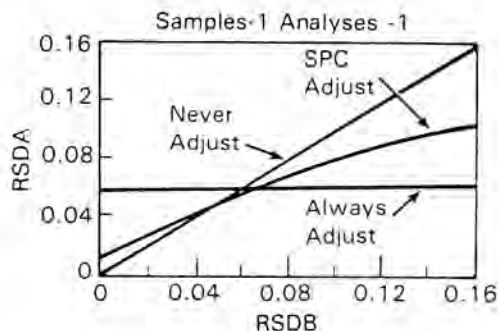


Fig. 4. Comparison of Decision Rules for Adjusting Feed Composition

When the decision is based on the Shewart control chart method as shown in Fig. 4, the adjustment effect is partially governed by the probability level chosen (i.e., at 99.7% the probability is 0.003 that an adjustment will be made when the composition is actually on target). If a larger risk of unnecessary adjustments is tolerable (say 0.05 or 0.10), then the SPC-adjust curve would be closer to the always-adjust line when the RSDB is greater than the combined sampling and analytical relative standard deviation. The SPC-adjust curve would be farther from the never-adjust line when the RSDB is less than the combined sampling and analytical relative standard deviation.

Figure 4, indicates that SPC is a valuable tool for improving glass quality through feed adjustments. Without SPC, the process variation could be increased, thereby adversely affecting the quality of the product. The assumption of an exact adjustment was used to produce Fig. 4. If the adjustment is an operation with some uncertainty involved, the always-adjust criterion could increase the variability to a greater extent than shown in Fig. 4. The SPC-based rule is somewhat flexible; sample sizes and probability levels can be manipulated to form a desirable adjustment decision criterion. With this decision criterion in place, the quality of the product will be more demonstrable.

#### Conclusions and Recommendations

The LFCM operator should have a quantitative method for demonstrating the production of a quality product. If the LFCM process is shown to be stable (in statistical control), characterizations of the glass product from feed samples will be more reliable. Although glass samples may be obtained as the glass is poured into the canisters or from the top of an open canister, the number of samples will be limited. The information obtained from feed sampling and during process monitoring will help substantiate the claims for a quality product.

Statistical process control is a quantitative method for monitoring and improving the LFCM process. SPC provides a mechanism for obtaining and demonstrating a stable process by detecting unacceptable process changes affecting glass quality early enough in the production cycle that corrective adjustments can be made. A statistical basis for decisions (accounting for process

uncertainties) concerning actions towards improving the quality of the product is also provided.

This paper focuses on applying SPC to the feed composition. However, SPC could be applied upstream to other aspects of the LFCM process, e.g., analytical standards, process instrumentation, and tank level measurements. Trouble spots in the process can then be pinpointed before it becomes too late or too costly to make the appropriate adjustments. Although some LFCM operators may elect to provide a final adjustment to the feed composition, the adjusting procedure may only increase the process variation if the adjustments are not sufficiently exact or may require substantial effort and time. In such cases, eliminating the sources of initial feed composition errors would provide a significant savings.

Statistical process control techniques could help the LFCM operators support their claims that a vitrification process can be controlled sufficiently to produce an acceptable product. This quantitative aspect of quality assurance, coupled with accurate documentation and adherence to quality related procedures, would provide strong evidence of product quality.

#### REFERENCES

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