

APPLICATION OF MULTIVARIATE STATISTICAL PROCESS CONTROL (MSPC) TO THE WEST VALLEY SLURRY-FED CERAMIC MELTER PROCESS

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ABSTRACT

During operation the West Valley Demonstration Project will solidify high level wastes from nuclear fuel reprocessing using the Slurry-Fed Ceramic Melter (SFCM) process. In order to produce a durable waste glass within the specified composition range, West Valley must assure consistent operation of the SFCM process and its feed system. There are a large number of both monitored and controlled variables in this system. However, there is also a great deal of correlation between variables. This makes the West Valley SFCM system a logical place to apply Multivariate Statistical Process Control (MSPC) technology. In contrast to univariate Statistical Process Control (SPC), MSPC takes advantage of the correlation structure in the process data to assure that the system and its sensors are operating normally. The result is a system that can detect individual sensor faults and process upsets. This article summarizes the MSPC technology developed by the Center for Process Analytical Chemistry (CPAC) for application to the West Valley SFCM system. While some information concerning the workings of the methods is presented, the article focuses on the results of the applications. Examples include applications of MSPC to both the SFCM feed system and the melter itself.

INTRODUCTION

Starting in 1992, West Valley Nuclear Services Co., Inc. (WVNS) will solidify the wastes from nuclear fuel reprocessing campaigns performed in the late 1960's and early 70's by Nuclear Fuel Services at a site near Buffalo, N.Y. The Slurry-Fed Ceramic Melter (SFCM) process will be used to vitrify a mixture of the original wastes, contaminated zeolite and glass forming additives. To date, WVNS has performed 12 tests of the vitrification system lasting from a few days to nearly 2 months. These tests employed a non-radioactive feed that was chemically similar to the feed expected during actual operations.

WVNS has contracted with the Center for Process Analytical Chemistry (CPAC) at the University of Washington to develop Multivariate Statistical Process Control (MSPC) tools for monitoring the performance of the SFCM and its feed system. MSPC will be used in addition to other (conventional) control procedures at West Valley.

The goal of MSPC at West Valley is to help assure smooth operation of the melter and its feed system. This goal is addressed in two ways. The first is by providing measures of the system performance that reduce the number of variables to be monitored, thus reducing the information overload experienced when very large data sets must be reviewed. The second is by developing methods to detect any systematic changes to the system or its sensors. Both of these problems are attacked with Principal Components Analysis (PCA), and the related technique of Varimax rotation. These methods are described in the sections that

follow. Examples, taken from the periods of testing at WVNS, are given to illustrate the methods.

THE WEST VALLEY FLOWSHEET

A simplified schematic of the West Valley flowsheet is shown in Fig. 1. The reprocessing wastes on site initially consisted of about 2×10^6 liters of alkaline PUREX wastes and about 3.5×10^4 liters of waste from the reprocessing of an experimental thorium reactor fuel. The PUREX waste had separated into a sludge layer consisting primarily of metal hydroxides, and a supernatant layer of dissolved salts. Radioactive cesium has been removed from the supernatant with zeolite molecular sieves. Prior to radioactive startup the contaminated zeolite will be mixed with the PUREX sludge, along with the Thorex wastes. During operation of the vitrification system this mixed waste will be transferred to a feed makeup tank where the batch will be sampled and a calculated amount of glass forming materials will be added. This mixture will then be concentrated and transferred to the feed tank.

The SFCM is the reference process in the United States for solidifying the liquid wastes produced during the reprocessing of nuclear fuels (1). A simple schematic of the LFCM operated at West Valley is shown in Fig. 2. The slurry consisting of waste and glass formers is fed onto the surface of the molten glass pool which is heated by passing a current between pairs of the three electrodes. Volatiles, consisting primarily of water and acids, are driven off and treated in an off-gas system which is not shown. The dried feed which remains forms a "crust" or "cold cap" which melts continuously into the glass. Glass is poured periodically

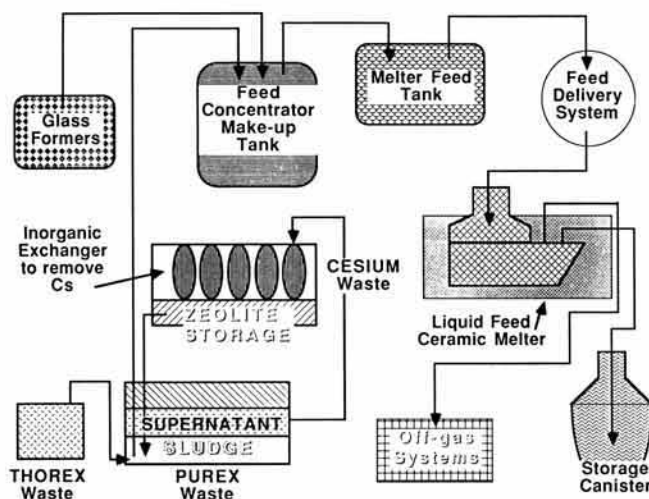


Fig. 1. West valley vitrification system flow sheet.

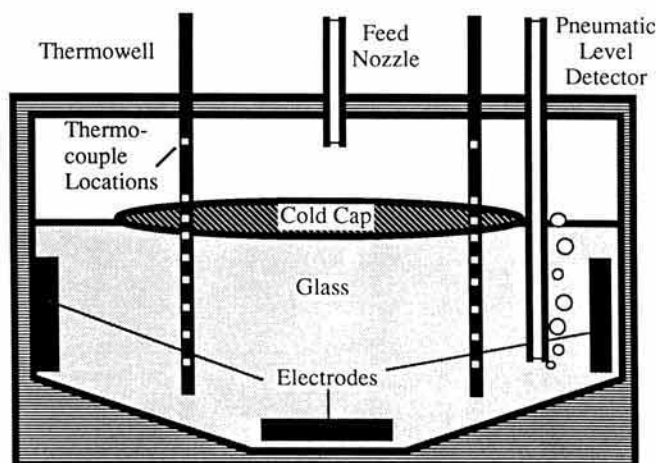


Fig. 2. Schematic drawing of slurry-fed ceramic melter.

from the melter through a riser section which is also not shown. This results in periodic fluctuations of the glass level.

The melter is monitored extensively (2). Temperatures are monitored at 20 locations within the melter (in the molten glass pool and above it in the plenum space), and the resistance and power dissipated between each of the electrode pairs is recorded. Data is also taken on feed flow rate and glass tank level. In all, 29 variables are recorded. As might be expected, many of the variables are highly correlated. Methods such as PCA are more appropriate for data

of this type than methods that assume statistical independence of the variables.

The melter data is recorded at 5 minute intervals, though the process is actually sampled at a much faster rate. Though 5 minutes may seem long for some chemical processes, the SFCM is generally very slow to respond to setpoint changes or disturbances and so the 5 minute sample time is appropriate. The process time constant for temperature changes associated with power setpoint changes is on the order of hours. The glass tank residence time is on the order of days. Process data is stored on the WVNS VAX system and can be transferred to CPAC electronically.

THE PCA AND VARIMAX METHODS

Many references are available on the PCA method, so the mathematical details will be considered only briefly here. The basic idea behind PCA is that the method finds combinations of the original variables that describe large trends in the data. In PCA an m by n data matrix X (where the rows are samples, the columns are variables and each variable has been "mean-centered" to produce variables of zero mean) is decomposed into the sum of the product of n pairs of vectors (3, 4). Each pair consists of a n by 1 vector called the loadings, p_i , and a m by 1 vector referred to as the scores, t_i . Thus X can be written as

$$X = t_1 p_1^T + t_2 p_2^T + \dots + t_n p_n^T \quad \text{Eq. (1)}$$

The matrix of loadings vectors P forms a new orthogonal basis for the space spanned by X and the individual p_i are the eigenvectors of the covariance matrix of mean-centered data matrix X , defined as:

$$\text{covariance}(X) = \frac{1}{m-1} (X^T X) \quad \text{Eq. (2)}$$

Thus

$$\text{covariance}(X) p_i = \lambda_i p_i \quad \text{Eq. (3)}$$

where λ_i is the eigenvalue associated with the eigenvector p_i . If the variables in X have been autoscaled (mean-centered and divided by the standard deviation to produce variables of zero mean and unit variance) the covariance matrix becomes the correlation matrix. The loadings vectors p_i are referred to as principal components because they are linear combinations of the original variables that together explain large fractions of the information in the original matrix. Each of the the scores vectors t_i is simply the projection of X onto the new basis vector p_i :

$$t_i = X p_i \quad \text{Eq. (4)}$$

The value of each λ_i is an indicator of the covariance in the data set in the direction p_i . In fact

$$\% \text{ variance in } p_i = (\lambda_i / \sum \lambda_i) \cdot 100 \quad \text{Eq. (5)}$$

PCA is very closely related to the Singular Value Decomposition (SVD) (5) where a data matrix X is decomposed as

$$X = USV^T \quad \text{Eq. (6)}$$

where V contains the eigenvectors (p_i) and S is a diagonal matrix containing the square roots of the eigenvalues (the singular values) of the covariance matrix of X .

Once the eigenvectors have been determined using PCA or SVD, projections of the data onto the eigenvectors can be made. These projections are commonly referred to as "scores plots" and are often useful for showing the relationships between the samples (rows) in the data set. Plots can be done as the projections of the samples onto a single eigenvector versus sample number (or time) or onto the plane formed by two eigenvectors. A projection of the samples onto the two eigenvectors associated with the largest eigenvalues depicts the largest amount of information about the relationship between the samples that can be shown in two (linear) dimensions. It is for this reason that PCA is often used as a pattern recognition and sample classification technique.

Plots of the coefficients of the eigenvectors themselves, known as "loadings plots", show the relationships between the original variables in the data set. Correlations between variables show up in the loadings plots.

When PCA is done on a data set it is often found (and it is generally the objective) that only the first few eigenvectors are associated with systematic variation in the data and that the remaining eigenvectors are associated with noise. Noise in this case refers to uncontrolled experimental and instrumental variations arising from random processes. PCA models are formed by retaining only the eigenvectors which are descriptive of systematic variation in the data. Determination of the proper number of eigenvectors can be done by cross-validation or other techniques (6, 7). Once the PCA model is formed new data can be viewed as projections onto single eigenvectors (scores plots) or the plane formed by pairs of eigenvectors. The scores can be used to obtain the "PCA estimate" of a given sample, *i.e.*, the projection of the sample into the PCA model. For a reduced-order model P_k (where only the first k of the n total eigenvectors are retained) and a new sample x_i this is obtained from:

$$\hat{x}_i = t_{ki} P_k^T = x_i P_k P_k^T \quad \text{Eq. (7)}$$

where t_{ki} is the (1 by k) vector of scores on the model P_k for sample x_i .

The "goodness" of fit between new data and the model can be monitored by calculating the data residual, *i.e.*, the difference between the actual sample and its PCA estimate. The residual r_i for sample x_i is given by

$$r_i = x_i - \hat{x}_i = x_i(I - P_k P_k^T) \quad \text{Eq. (8)}$$

The magnitude of the residual for any sample x_i is

$$Q = ||r_i|| = r_i^T r_i = x_i^T (I - P_k P_k^T) x_i \quad \text{Eq. (9)}$$

While the PCA model is an optimal model in terms of describing the variance structure in a given data set, it is often difficult to interpret in terms of the physical significance of the individual PC loadings. This is especially true when all the variables are inherently related to each other as is the case for composition variables from batching processes where all components must add up to 100%. In these cases, the PC loadings frequently contain intermediate loadings for many, or most, of the variables. From the interpretation standpoint, it is desirable that each principal component have only a few variables with high (positive or negative) loadings and that most of the other variables have zero, or near-zero, loadings.

The method of Varimax rotation consists of iterative rotations of PC pairs where the rotation operation is selected to maximize the "simplicity" of the PC loading vectors while retaining the maximum amount of information. This results in principal components which are much easier to interpret when applied to the feed composition data. In this report we will present the results of Varimax rotated PCA models when discussing feed monitoring applications and will refer to the Varimax rotated PCs as simply PCs.

SFCM FEED SYSTEM MONITORING

During operation the SFCM feed will be sampled at least twice during makeup. The first sample is taken after a transfer is made from the combined wastes in the PUREX tank. The calculations for the amount and composition of the glass forming additives are based on this sample. After addition of the additives the feed mixture is sampled again to assure that the target compositions have been met. If the batch is sufficiently close to the target composition, no further additions are made and the feed is transferred to the final feed tank. If the feed does not meet specifications, a "shim" is calculated and added to the feed and the tank is re-sampled.

Based on this sampling scheme, there are several opportunities to apply MSPC to the feed makeup process. During the experimental phase of the West Valley program the feed makeup stages of the feed train have been simulated. (It has not, of course, been possible to simulate the actual drawing of the wastes from the original PUREX tank.) The MSPC approach has been used to model the natural variation in the feed makeup process and to catch systematic deviations in the feed due to changes in the process or difficulties with the calibration of the instruments used to monitor the feed. Several examples of this follow.

Figure 3 shows the scores on the first principal component from the analysis of the feed composition from the

SF-9 Sample Scores on First Principal Component

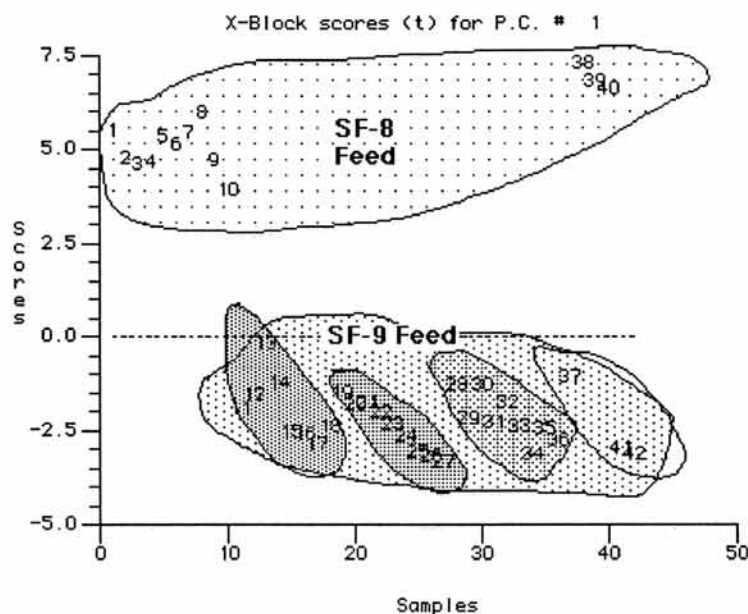


Fig. 3. Scores on first PC of SF-9 feed model showing systematic variation.

melter trial designated SF-9. (Also shown on the plot are some feed scores from the previous SF-8 trial.) Here the score value of each sample is plotted in the order in which the samples were taken. Thus, the x-axis is proportional to time. There is evidence here of a systematic variation in the score value with time. Consideration of the loadings vector associated with this scores vector showed that the variables associated with the observed trends were indicative of changes in the suspended solids content of the feed. Based on this analysis an investigation was made concerning the homogeneity of the mixing in the feed tank from which the samples were taken. It was discovered that when the feed level in the tank dropped below the top agitator blade the mixing suffered and some solids tended to drop out of the feed, resulting in a slightly off-specification composition. This problem was eliminated very simply by changing the tank operating procedure so that the feed level was not allowed to go below the top agitator blade. This trend was absent in all of the following melter trials.

An example of detection of drifting calibration of the analytical instruments used to monitor feed composition is shown in Fig. 4. The figure shows the scores on the third principal component from the SF-9B run. Here it can be seen that the sample scores "drifted" away from the normal region. Based on this analysis, the samples were re-analyzed at a later date. The result of this is shown in Fig. 5, which shows that the re-analyzed samples lie in the normal region. Thus, it was deduced that the cause of the apparent

change in composition was actually due to instrument calibration drift, as opposed to a change in the feed system.

MSPC charts have also been made to compare the feed compositions of the different runs. An example of this is the feed scores from runs SF-10 to SF-12 on the first PC of the SF-12 model, as shown in Fig. 6. From the figure it can be seen that the feed composition from many of the earlier runs was substantially different from the SF-12 feed. This was primarily due to changes in the desired composition which was still being finalized as the melter trials proceeded. As the changes in the desired composition became less significant the scores "home in" on the SF-12 model. This is also illustrated in Fig. 7, which shows a 3-d plot of the feed composition residuals for the earlier runs based on the SF-12 feed model. It is apparent that the major differences in the earlier feeds were primarily changes in some minor constituents of the glass, such as Sr and Ba. Note that this figure represents a change in relative, not absolute, amounts of these constituents.

As a final example of the application of MSPC to feed monitoring, the mean of each of the feed batches from the separate runs was subtracted out and the results compared to the SF-12 model. This was done to eliminate the influence of the largely intentional changes in the feed composition from run to run so that the variation within runs could be more clearly observed. The scores of these "adjusted" feed samples are shown on the first PC of the SF-12 model in Fig. 8. Note how the scores from each of the runs become

Scores on First Principal Component of Model

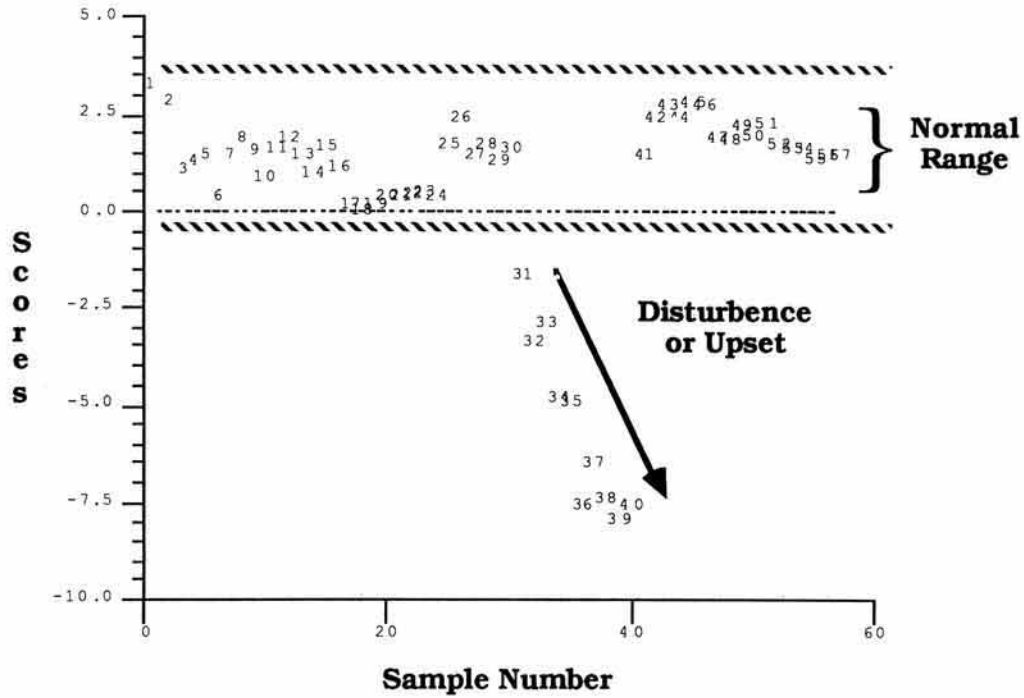


Fig. 4. Scores on first PC of SF-9B feed model showing calibration drift.

Sample Scores on First Principal Component

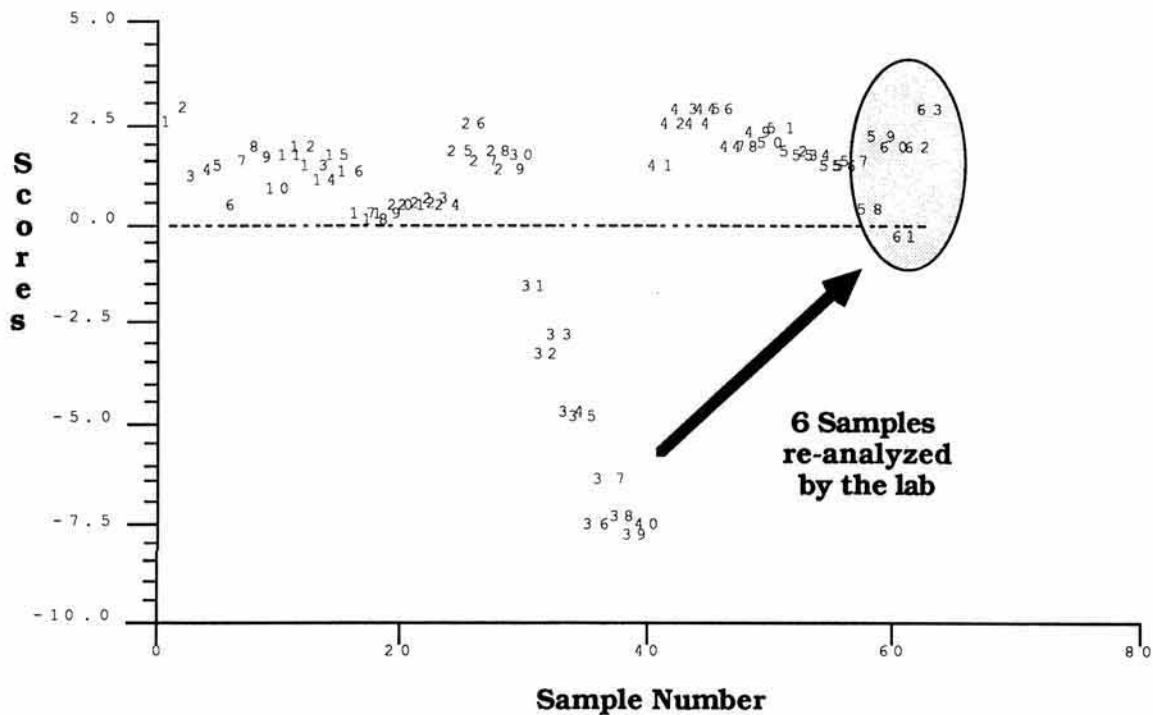


Fig.5. Re-analyzed feed samples on first PC of SF-9B model.

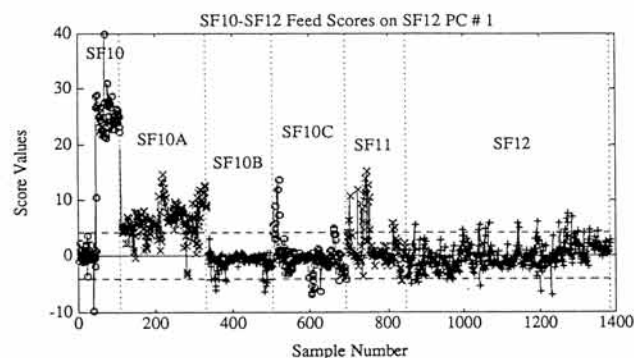


Fig. 6. Scores on first PC of SF-12 feed model comparing previous runs.

largely indistinguishable. This is an indication that the variation in the feed composition for each run has been approximately the same. In this sense, the feed composition could be considered to be well "in control."

MELTER MONITORING

PCA has shown great utility as an MSPC tool for situations like feed composition monitoring where, under normal circumstances, the samples can be considered independent. The utility of the method for monitoring dynamic processes has also been shown (8-10). Until re-

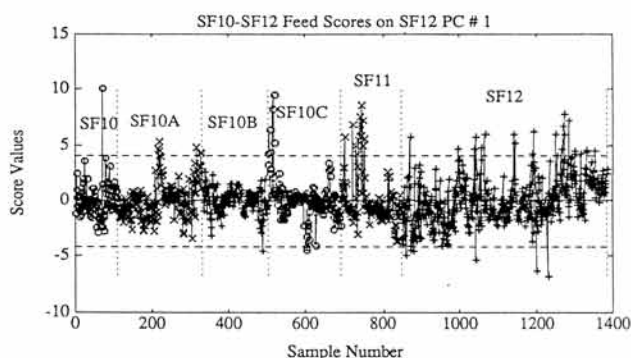


Fig. 8. Mean adjusted feed scores on first PC of SF-12 model.

cently, however, a firm theoretical basis for its use with dynamic process data has been lacking. This basis was been provided by the CPAC authors of this article (11). This theoretical development is reviewed briefly here and applications of the theory to melter monitoring at West Valley are given.

In order demonstrate the effect of PCA on dynamic process data, the state-space model format is introduced here. The state-space form is convenient for modelling linear, time-invariant (LTI) processes. The discrete form of the state-space process model (applicable to sampled data systems) has the form:

$$\mathbf{x}(k+1) = \Phi\mathbf{x}(k) + \Gamma\mathbf{u}(k) + \mathbf{v}(k) \quad \text{Eq. (10)}$$

$$\mathbf{y}(k) = \mathbf{C}\mathbf{x}(k) + \mathbf{D}\mathbf{u}(k) + \mathbf{e}(k) \quad \text{Eq. (11)}$$

Equation (10) is commonly referred to as the state equation, while equation (11) is the measurement equation. Assuming that the process has n states, r inputs and p measurements, then in the state equation $\mathbf{x}(k)$ is the $(n \text{ by } 1)$ vector of state variables at time k , $\mathbf{u}(k)$ is the $(r \text{ by } 1)$ vector of process inputs at time k , Φ is the $(n \text{ by } n)$ state transition matrix which determines the effect of the states at time k on the states at time $k + 1$, Γ is the $(n \text{ by } r)$ input matrix which determines the effect of the inputs at time k on the states at time $k + 1$ and $\mathbf{v}(k)$ is the $(n \text{ by } 1)$ vector of state disturbances at time k . In the measurement equation, $\mathbf{y}(k)$ is the $(p \text{ by } 1)$ vector of process measurements at time k , \mathbf{C} is the $(p \text{ by } n)$ measurement matrix which describes how the states relate to the measured outputs, \mathbf{D} is the $(p \text{ by } r)$ feed through matrix, which describes the direct effect of the process inputs on the measured outputs and $\mathbf{e}(k)$ is the vector of measurement noise at time (k) . For most processes \mathbf{D} is zero; process inputs rarely have an instantaneous effect on

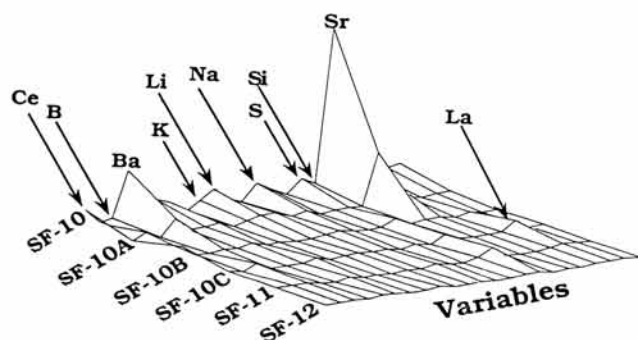


Fig. 7. Residuals from previous run feeds on SF-12 model.

the process outputs. The dimension of the state space can be greater than, less than or equal to the dimension of the measurement space. Many references concerning the state-space formalism are available (12, 13).

We have shown previously that, for processes with more measurements than states, proper application of PCA can facilitate the process monitoring and fault detection problem. This is done by identifying a PCA model of the outputs $y(k)$. Under most circumstances, the PCA model will span the same space as the C matrix in the corresponding state-space representation of the process. When the PCA model is an accurate estimate of C , variations in the process states $x(k)$ appear primarily as variations in the PCA scores, while noise $e(k)$ mainly affects the residuals. This allows one to consider only the noise properties when deriving error-detection limits for the PCA residuals. In particular, the process dynamics need not be considered explicitly. This implies that conventional statistical methods that rely on independence of the samples (such as t - and F -tests and the multivariate T^2) can be applied to the PCA residuals.

The temperature measurements in the West Valley SFCM can be treated as a system with more measurements than states. In all, 20 temperatures are recorded within the melter, but analysis shows that the dynamics of the melter are dominated by ~ 4 states. Thus, a 4 PC model can be used to monitor the SFCM for changes in sensor behavior or process upsets. In practice, application of PCA to the SFCM does not produce completely independent residuals (14), but this is easily compensated for by adjusting the calculated limits on the residuals to account for the autocorrelation remaining. Based on this, it is possible to estimate the size of a sensor bias or the amount of additional sensor noise which will result in the indication of an abnormal event.

These sensor error detection limits are shown for the each of the SFCM thermocouples in Fig. 9. The size of a detectable bias error is shown along with the standard deviation of a detectable added noise error. Note that the magnitude of the detectable bias or noise is different for each of the sensors. The detection limits for the thermocouples in the bulk glass (variables 1-6 and 11-16) are quite small because these variables are very correlated with each other and do not normally have large variances. Variables in the cold cap region of the melter (7-9 and 17-19) show larger limits primarily because these temperatures vary over a larger range under normal circumstances. The plenum temperatures (10 and 20) have the largest detection limits because these two variables are primarily correlated only with each other and can show wide variation under normal circumstances.

CONCLUSIONS

This article has demonstrated how the MSPC approach to process monitoring is being applied at WVNS. The examples given here demonstrate that the methods are quite effective for assessing the state of the SFCM and its feed system. Unusual trends in the feed or process data are made readily apparent through MSPC. This approach should be especially fruitful during radioactive operation when wastes are being withdrawn from the tank farm. MSPC will provide West Valley a convenient method for tracking trends in the composition of the waste so that the composition of future batches may be anticipated. Unusual batches will also be easily detected. MSPC for melter monitoring will help keep the system operating smoothly through early detection of failing sensors.

REFERENCES

1. Burkholder, H. C., and J. H. Jarrett, compilers. *Nuclear Waste Treatment Program Annual Report for FY 1985*. PNL-5787, Pacific Northwest Laboratory, Richland, WA. 1986.
2. Barnes, S. M., J. H. Westsik, Jr. and B. M. Wise, "Instrumentation Concepts for Nuclear Waste Glass Melters," *Waste Management '85 Proceedings*, Tucson, AZ 1985.
3. Veltkamp, D. J., B. R. Kowalski, N. L. Ricker and B. M. Wise, "Multivariate Statistical Process Control Using Principal Component Analysis", Submitted to *J. Chemometrics*, Feb. 1990.
4. Jackson, J. E., "Principal Components and Factor Analysis: Part 1," *J. Qual. Tech.*, 13(1), 1981.

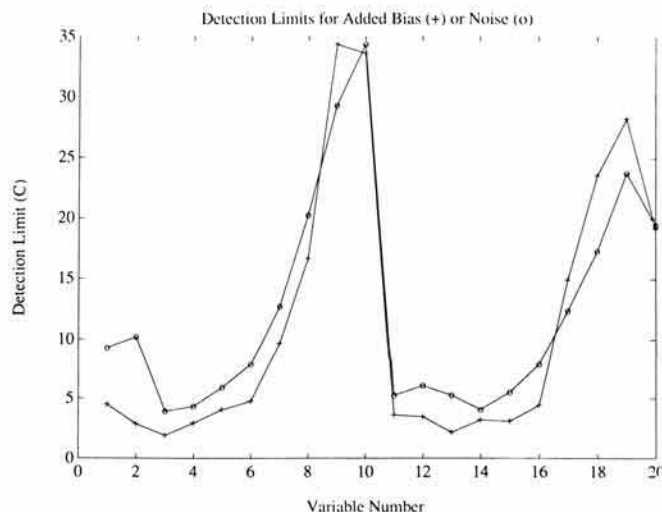


Fig. 9. Detection Limits for Bias and Noise Errors of SFCM Thermocouples.

5. Strang, G., *Linear Algebra and Its Applications*, Academic Press N. Y., 1980.
6. Malinowski, E. R., "Determination of the Number of Factors and the Experimental Error in a Data Matrix," *Anal. Chem.*, **49**, 612 (1977).
7. Malinowski, E. R., "Theory of the Distribution of Error Eigenvalues Resulting From Principal Component Analysis with Applications to Spectroscopic Data," *J. Chemometrics* **1**, 33 (1987).
8. Wise, B. M. and N. L. Ricker, "Feedback Strategies in Multiple Sensor Systems" in *AIChE Symposium Series*, **85**(267) 1989.
9. Wise, B. M., D. J. Veltkamp, B. Davis, N. L. Ricker and B. R. Kowalski, "Principal Components Analysis for Monitoring the West Valley Liquid Fed Ceramic Melter," *Waste Management '88 Proceedings*, Tucson AZ 1988.
10. Kresta, J., J. F. MacGregor and T. E. Marlin, *Multivariate Statistical Monitoring of Process Operating Performance*, Sub. to *Can. J. Chem. Eng.*, Feb. 1990.
11. Wise, B. M., N. L. Ricker, D. J. Veltkamp and B. R. Kowalski, "A Theoretical Basis for the Use of Principal Components for Monitoring Multivariate Processes," *Process Control and Quality*, **1**(1), 1990.
12. Kwakernaak, H. and R. Sivan, *Linear Optimal Control Systems*, Wiley-Interscience, New York 1972.
13. Sage, A. P. and C. C. White III, *Optimum Systems Control*, Second Edition, Prentice-Hall, New Jersey, 1977.
14. Wise, B. M., *Adapting Multivariate Analysis to the Monitoring and Modelling of Dynamic Systems*, Ph.D. Dissertation, University of Washington, Seattle, 1991.