Statistical Operating Strategies for Charging Batch Reactors

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Two critical decisions must be made in the charging sequence of batch reactors: target setting and alarm generation. A number of statistics-based strategies are proposed in this article to perform these tasks. The off-line and on-line target setting procedures developed in this work can be adopted to increase the profit margin of any given batch process without sacrificing reliability. A synthesis method for building optimal alarm logic is also described in detail. Monitoring systems constructed according to this suggested approach are effective in reducing the probability of undetected faulty batches. Extensive simulation studies show that the proposed strategies are suitable for application of manufacturing high-value-added products, which is a prevailing practice in batch processes.

Introduction

Batch processes have always been employed in the manufacture of small amounts of products with purities and/or other qualities not readily attainable in the continuous ones. In particular, specialty chemicals with high added value are often produced in batch reactors, such as specialty polymers, pharmaceuticals and biochemicals. Since batch processing is time-variant in both operating conditions and system configuration, there is a need to place significant emphasis on effective control in order to ensure operational safety and to achieve repeatable and accurate batches.

The control functions of a batch process can usually be divided into several levels (Severns and Hedrick, 1983). The top of the hierarchy is batch-cycle control. It has a plant-wide scope to accommodate tasks such as scheduling, production planning, and optimization. Although this is an area of active research (Birewar and Grossman, 1989; Tan et al., 1993; Kondili et al., 1993; Shah et al., 1993), the focus of this work is concerned with the next level of control functions, i.e., those needed to automatically sequence and execute all steps within a single batch. Further, for the sake of illustration clarity, the discussion presented later in this article will be limited to the specific case of batch reaction only.

According to Perry et al. (1984), the operating sequence of a batch reactor typically consists of three stages, i.e., charging the reactor, manipulating the operating conditions to meet some processing criterion, and shutting down and emptying the reactor. In principle, the specific control actions needed to perform these works can be synthesized manually based on experience or automatically with one of the algorithms proposed in the literature (Fusillo and Powers, 1987, 1988a,b; Lakshmanan and Stephanopoulos, 1988a,b; 1990; Foulkes et al., 1988; Aelion and Powers, 1991; Crooks and Machietto, 1992). In executing these operating procedures, the targets of control actions must be specified first. For example, the target amounts of the raw materials that must be charged into the reactor should be determined on the basis of production demand and/or some other operational constraints. Also, if an accurate mathematical model of the reaction system is available, the target profiles of the operating conditions can be established with the Pontryagin maximum principle (Cuthrell and Biegler, 1989; Villermaux and Georgakis, 1991).

In realistic operations, uncertainties and unexpected disturbances in the control actions are almost unavoidable. The above target setting policies must thus be adjusted accordingly. As a result, monitoring becomes another critical control function. Most of the related studies in the past are only concerned with the second stage of batch reaction sequence. The task of monitoring is confined to checking that the planned operation steps are followed and that certain measurement variables are following predetermined trajectories *during* reaction. A detailed account of the literature can be found in Nomikos and MacGregor (1994). However, it should be noted that, in addition to the operating conditions during reaction, the outcome of a batch is sensitive to the initial state as well. For example, a runaway reaction or at least a

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drop in yield may result from improper preparation of the raw materials (Haldar and Rao, 1992). In other cases, the unreacted reactants remaining in the product may create undesirable downstream problems, such as separation difficulties, pollution and safety hazards. Thus, it is clear that an accurate charging procedure, which includes a target setting method and a monitoring scheme, is one of the primary factors for successful batch reactor control and the development of statistical operating strategies for charging the batch reactors is exactly the objective of this research.

Although in practice there are various different types of batch chargers, the material transfer methods can be broken into a few general categories (Rosenof and Ghosh, 1987). *Direct charging* simply refers to the process of transferring the raw material from a storage tank into the batch reactor. The charge amount may be measured by level or weight sensors at the storage tank or batch reactor, or by flow between them. This method is simple and economical. On the other hand, *indirect material transfer* system use "preweigh" buffer tanks between source and destination. The additional investment in these tanks is justified by improved charge accuracy and reduced batch time. Whatever the method used, uncertainties are always associated with the material transfer process. The current target-setting practice in the industry is mainly experience-based. In this work, however, a statistics-based approach has been taken. Various simple error models were first formulated according to the characteristics of different types of charging and measurement methods. The techniques for extracting the parameters of these models from historical and/or on-line measurement data were then established. On the basis of these parameters, several target setting procedures were developed to achieve a given level of reliability.

The statistical process control (SPC) methods are probably the most natural choices for monitoring purpose (Badavas, 1993; Al-Salti and Statham, 1994; Nomikos and MacGregor, 1994). However, if SPC techniques such as Shewhart or CUSUM charts are used directly for the present problem, there are drawbacks which require special attention. First of all, since the product value created per batch is quite high, it is not acceptable to detect the out-of-statistical-control batches only *after* several of them have already been completed. In other words, the charging failure should be detected as soon as possible in each batch. Secondly, the alarms may be mistakenly set off due to measurement errors. Again, as a result of the high added value of the product, the financial implications of false alarm and undetected failed batches may be too great to be ignored. Thus, there is also a need to minimize the loss caused by misjudgments in monitoring. A common approach to solve these problems in the industry is to introduce redundancy and diversity in the system. Specifically, several independent sensors are installed to monitor the same material transfer process. Any inconsistency identified in the measurement data obtained from different sensors is usually resolved on the basis of operation experience. In this research, however, a systematic method for synthesizing the optimal alarm generation logic has been developed. Basically, the design techniques of trip systems (Inoue et al., 1982; Kohda et al., 1983) were extended for the development of such a method. In addition, in order to implement the proposed logic synthesis method, the probabilities of false alarms and undetected failures must be estimated in advance. An estimation procedure for these parameters was also developed in this study.

In this article, descriptions of the above strategies are presented in detail. The implementation procedure of the proposed techniques is illustrated with an application example. The feasibility and benefits of the present approach are also clearly demonstrated with extensive simulation studies.

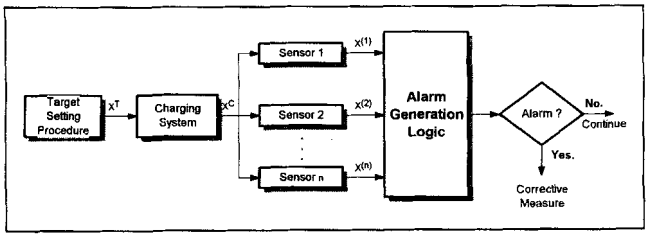

Figure 1. Charging operation of batch reactors.

Charging Operation of Batch Reactors

To avoid confusion, it is best to define the batch reactor charging process conceptually at the beginning of our argument. Figure 1 depicts the procedure of a typical charging operation associated with one reactant. If more than one raw material is involved, it is assumed that they are fed separately according to Figure 1 in sequence.

Before the actual transfer takes place, a target amount X^T, usually determined on the basis of market demand, must be given to the operator or specified in the computer program of a PLC. This value can thus be regarded as a known constant during each batch. Each reactant is delivered from its storage tank either directly to the reactor or temporarily to a measure vessel first. The charge amount is manipulated manually or automatically according to the on-line measurements, such as readings of the level indicator on reactor or measure vessel, or the time elapsed since the inception of pumping operation. It should be noted that the amount of material *actually* charged into the reactor, i.e., X^C, may not be exactly the same as the target. Both bias and random errors are possible in this transfer process. In order to ensure operational reliability and safety, it is a common industrial practice to install several independent and diversified sensors to monitor X^C (Fisher, 1990; Rosenof and Ghosh, 1987; Tsai and Lane, 1976). There are a number of typical approaches available for measuring this amount, such as flow totalization of the raw material through the inlet pipeline and determination of the level change (by DP cell) or weight change (by weight cell) in the reactor or measure tank. Again, due to random and systematic errors, the measurement values, i.e., $X^{(i)}$ $(i = 1, 2, \ldots, n)$, obtained from different sensors are in general not consistent with one another. Nonetheless, one is still required to check the status of the charging process against operational constraints with these data. Usually, detection strategies for unacceptable conditions, i.e., the *alarm generation logic*, are developed on an *ad hoc* basis. For example, an alarm may be set off on the basis of the most reliable sensor or an arbitrarily chosen ℓ-out-of-n ($\ell \leq n$) logic, and so on. If an alarm is generated accordingly, remedial measures can be taken to correct this undesirable condition. Otherwise, the subsequent steps specified in the operation manual can be executed in sequence.

In summary, X^C in the charging process can never be obtained exactly and the data available to us in each batch are the values of X^T and $X^{(i)}$s only. Based on these historical data obtained from previous batches, the target setting procedure and the alarm generation logic are the two essential strategies that must be developed for any batch reactor charging sequence.

Operational Constraints

As mentioned before, the raw materials are charged into the batch reactor in sequential steps. In general, the actual amount transferred in each step is required to satisfy one or more operational constraints. Both inequality and equality constraints can be identified in practical applications.

The inequality constraints are often imposed to address safety concerns. For example, in pesticides-producing processes, toxic chemicals such as phosgene are frequently used in batch reactors. Complete conversion of these chemicals is thus highly desirable in this situation. In some other processes, the possibility of an exothermic runaway reaction may occur if the ratio of two reactants exceeds the design value. Occasionally, it may even be necessary to satisfy multiple constraints in order to ensure operational safety.

On the other hand, the charge amount may have to be equal to a prescribed value c_1 to meet the production demand

$$X^C = c_1 \tag{1a}$$

Or, for the sake of maximizing yield or selectivity, it is sometimes necessary to maintain the ratio of X^C to the amount of another reactant at a target level

$$X^C = c_2 Z^C \tag{1b}$$

where Z^C denotes the actual amount of one of the reactants which have already been charged into the reactor and c_2 is a constant. Violation of the equality constraints usually results in inefficient operation or degradation in product quality.

Due to various uncertainties involved in the charging process, the above two types of constraints cannot always be satisfied exactly in practice. Thus, the requirements implied by these constraints must be relaxed. In the former case, the expected probability of successful charging operation (in the sense that the constraints are satisfied) should be assigned. For the latter case, the equality constraints must first be changed into suitable forms. For example, Eqs. 1a and 1b can be replaced with

$$c_1^L \le X^C \le c_1^U \tag{1c}$$

and

$$c_2^L Z^C \le X^C \le c_2^U Z^C \tag{1d}$$

where c_j^L and c_j^U represent the lower and upper bounds of c_j respectively and $j = 1, 2$. As a result, the acceptable probability of satisfying these alternative constraints can be specified accordingly.

From the above discussions, one can see that the realistic operational constraints can be written in a generalized form with a *performance function* $\mathcal{G}$

$$\mathcal{G}(X^C; Z_1^C, Z_2^C, \cdots, Z_m^C) \ge 0 \tag{2}$$

where Z_j^C ($j = 1, 2, \ldots, m$) represents the actual amount of the jth raw material which has already been transferred into the reactor. In this article, only single-constraint problems are treated due to space limitation. Issues concerning the multiple constraints will be addressed in a separate article.

Error Models

To understand the nature of our problem at hand, it is necessary to gain a clear picture of the errors associated with the charging and measuring processes. Let us first consider the process of charging. Owing to imperfect control and operation of the equipment, the actual amount of reactant charged into reactor is usually not exactly the same as the target value

$$X^C = X^T + \Delta \tag{3}$$

where Δ is the error due to the charging system and, for convenience, it is assumed to be a normally distributed random variable. Note that, although both X^C and Δ are random variables, the target X^T should be viewed as a deterministic value in the charging process. As a result of variable market demand, it is often necessary to alter X^T, accordingly. Further, in some cases, this target can be adjusted according to on-line measurements of the other previously-charged reactants to optimize a predefined performance index. Thus, there is a need to formulate the error model associated with the charging process and, particularly, develop an expression of Δ as an explicit function of X^T.

The simplest approach to describe the behavior of Δ is to assume that it is independent of the target amount. This assumption is reasonable if the charging process is stopped on the basis of an end-point measurement, such as the output from a level or weight sensor. Throughout this article, these errors will be referred to as the *charging errors of type A*. The second approach adopted in this study is to assume that the charging error is proportional to the targeted amount

$$\Delta = X^T \delta \tag{4}$$

where δ is a random variable and its mean and variance can be considered to be independent of X^T. In other words, the actual amount X^C can be expressed with an alternative form

$$X^C = X^T X^\delta \tag{5}$$

where X^δ is a random variable and

$$X^\delta = 1 + \delta \tag{6}$$

From a practical viewpoint, these charging errors exist when the transferring flow is maintained and terminated according

to flow measurements. Errors that can be described by Eqs. 5 or 6 are called *charging errors of type B* in this study.

Next, let us consider the process of measuring X^C with sensor i ($i=1, 2, \ldots, n$). Bias and random errors may both exist in this process. Thus,

$$X^{(i)} = X^C + \Xi^{(i)} \tag{7}$$

where $\Xi^{(i)}$ is the measurement error associated with sensor i and it is also assumed to be normally distributed. In this study, it is further assumed that the charging errors Δ and these measurement errors $\Xi^{(i)}$ are statistically independent.

As mentioned previously, the amount of a reactant charged into reactor can be determined with level, pressure, weight, or flow sensors. The typical measurement errors of these instruments are in general reported in terms of a percentage of full scale and/or a percentage of reading. Thus, the measurement errors can also be described with two corresponding models in this study. *Measurement errors of type A* refers to errors not affected by X^C. Specifically, the corresponding mean $\mathcal{E}[\Xi^{(i)}]$ and Var $[\Xi^{(i)}]$ should be independent of the actual charge amount X^C. On the other hand, *measurement errors of type B* can be described by

$$\Xi^{(i)} = X^C \epsilon^{(i)} \tag{8}$$

Thus, the random variables $X^{(i)}$s can be expressed alternatively as a product of two random variables

$$X^{(i)} = X^C X_\epsilon^{(i)} \tag{9}$$

where

$$X_\epsilon^{(i)} = 1 + \epsilon^{(i)} \tag{10}$$

In this case, it is assumed that the mean and variance of $\epsilon^{(i)}$ are not affected by the value of X^C.

Parameter Estimation

Since the statistics-based techniques are probably the most appropriate tools for the development of a target setting procedure and to devise the alarm generation logic, the parameters that describe the probability distributions of charging and measurement errors must first be estimated correctly. To be specific, let us assume that the measurement data obtained from K previous batches are available to us. Also, it is assumed that the measurement errors associated with n_A sensors are of type A and those with the other n_B ($n_A + n_B = n$) sensors are of type B. On the other hand, the charging errors of both type A and B are allowed in the following discussions. Our task is thus to determine the sample means and variances associated with various errors from the historical data.

In this study, the maximum likelihood estimation techniques were adopted. In particular, the likelihood function for the parameters given the measurement data from one batch is *assumed* to be

$$L(\boldsymbol{\theta}_\mu, \boldsymbol{\theta}_\Sigma \mid \boldsymbol{x}_k) = \frac{1}{(2\pi)^{n/2} \mid \Sigma_k \mid^{1/2}} \exp\left\{ -\frac{1}{2} (\boldsymbol{x}_k - \vec{\mu}_k)^T \Sigma_k^{-1} (\boldsymbol{x}_k - \vec{\mu}_k) \right\} \qquad k = 1, 2, \cdots, K \tag{11}$$

where the vector $\boldsymbol{x}_k$ is defined as

$$\boldsymbol{x}_k = [x_k^{(1)}, x_k^{(2)}, \cdots, x_k^{(n)}]^T \tag{12}$$

and $x_k^{(i)}$ is the measurement value of sensor i ($i = 1, 2, \cdots, n$) obtained from the kth batch. The vectors $\boldsymbol{\theta}_\mu$ and $\boldsymbol{\theta}_\Sigma$ consist of, respectively, the expected values and the variances of the charging and measurement errors

$$\boldsymbol{\theta}_\mu = \left[\theta_\mu^C, \theta_\mu^{(1)}, \theta_\mu^{(2)}, \cdots, \theta_\mu^{(n)} \right]^T \tag{13}$$

$$\boldsymbol{\theta}_\Sigma = \left[\theta_\Sigma^C, \theta_\Sigma^{(1)}, \theta_\Sigma^{(2)}, \cdots, \theta_\Sigma^{(n)} \right]^T \tag{14}$$

and

$$\theta_\mu^C = \begin{cases} \mathcal{E}[\Delta] & \text{if the charging error is of type A} \\ \mathcal{E}[X^\delta] & \text{if the charging error is of type B} \end{cases} \tag{15}$$

$$\theta_\Sigma^C = \begin{cases} \text{Var}\,[\Sigma] & \text{if the charging error is of type A} \\ \text{Var}\,[X^\delta] & \text{if the charging error is of type B} \end{cases} \tag{16}$$

and

$$\theta_\mu^{(i)} = \begin{cases} \mathcal{E}[\Xi^{(i)}] & \text{if the measurement error of sensor } i \text{ is of type A} \\ \mathcal{E}[X_\epsilon^{(i)}] & \text{if the measurement error of sensor } i \text{ is of type B} \end{cases} \tag{17}$$

$$\theta_\Sigma^{(i)} = \begin{cases} \text{Var}[\Xi^{(i)}] & \text{if the measurement error of sensor } i \text{ is of type A} \\ \text{Var}\,[X_\epsilon^{(i)}] & \text{if the measurement error of sensor } i \text{ is of type B} \end{cases} \qquad i = 1, 2, \cdots, n \tag{18}$$

Finally, the vector $\vec{\mu}_k$ contains the expected values of the online measurements associated with the target X_k^T of batch k and the matrix Σ_k is the corresponding covariance matrix. Notice that the target value X_k^T may vary from batch to batch. It should also be noted that both $\vec{\mu}_k$ and Σ_k are functions of the target X_k^T and the $2n+2$ parameters in $\boldsymbol{\theta}_\mu$ and $\boldsymbol{\theta}_\Sigma$. Thus, the best parameter values can be found by maximizing the product of the likelihood functions associated with all K batches

$$\max_{\theta_\mu, \theta_\Sigma} \prod_{k=1}^{K} L(\theta_\mu, \theta_\Sigma \mid x_k) \tag{19}$$

This is a nonlinear optimization problem which can be solved iteratively with a standard numerical technique. It should be noted that since the measurements $X^{(i)}$s can be nonlinear functions of the charging and measurement errors the assumption of a normally distributed likelihood function, i.e., Eq. 19, may not be theoretically rigorous in every application. However, since these functions can often be well approximated by linearized models and, also, the subsequent decisions involving target setting and alarm generation are relatively insensitive to the accuracy of parameter estimates, the present approach is still taken for the sake of convenience.

Although, in principle, the maximum likelihood estimates of parameters θ_μ and θ_Σ can be obtained with the procedure described above, the computational load is usually extremely demanding. Thus, there is a need for the development of more efficient alternatives. In this study, it was found that this computation can be simplified if all measurement errors are of the same type. Specifically, it should be noted that, if only measurement errors of type A are involved, the differences between the measurement data from two different sensors does not involve the charging error

$$X^{(i)} - X^{(j)} = \Xi^{(i)} - \Xi^{(j)} \tag{20}$$
$$i \neq j \qquad i, j = 1, 2, \cdots, n$$

Thus, the following relations among the means of the measurement errors can be utilized in producing the maximum likelihood estimates

$$\mathcal{E}[\Xi^{(i)}] = \mathcal{E}[X^{(i)} - X^{(r)}] + \mathcal{E}[\Xi^{(r)}] \tag{21}$$
$$i \neq r \qquad i = 1, 2, \cdots, n$$

where, the superscript (r) denotes an arbitrarily chosen reference sensor. Notice that the first term on the right side of Eq. 21 can be estimated from historical data and thus only the mean of one of the measurement errors, i.e., $\mathcal{E}[\Xi^{(r)}]$, must be estimated iteratively. Further, useful information can be extracted from the variances of these differences. From Eqs. 20, one can obtain

$$\mathrm{Var}[X^{(i)} - X^{(j)}] = \mathrm{Var}[\Xi^{(i)}] + \mathrm{Var}[\Xi^{(j)}] \tag{22a}$$

Notice that there are in total $n(n-1)/2$ such equations. Adding all of them together yields

$$\sum_{j=1}^{n} \sum_{\substack{i=1 \\ i \neq j}}^{j} \mathrm{Var}[X^{(i)} - X^{(j)}] = (n-1) \sum_{k=1}^{n} \mathrm{Var}[\Xi^{(k)}] \tag{22b}$$

Thus, the variances of the measurement errors can be expressed as

$$\mathrm{Var}[\Xi^{(k)}] = \frac{1}{n-1} \sum_{j=1}^{n} \sum_{\substack{i=1 \\ i \neq j}}^{j} \mathrm{Var}[X^{(i)} - X^{(j)}] - \frac{1}{n-2} \sum_{\substack{j=1 \\ j \neq k}}^{n} \sum_{\substack{i=1 \\ i \neq j}}^{j} \mathrm{Var}[X^{(i)} - X^{(j)}] \tag{23}$$

Notice that all terms on the right side of Eq. 23 can be estimated from measurement data and, consequently, the estimates of Var $[\Xi^{(k)}]$s can be determined without iteration.

If, on the other hand, only type B errors exist in the measurement process, a similar approach can also be taken. In this case, the available data must first be transformed into acceptable formats. Specifically, the following equations can be used

$$\ln X^{(i)} - \ln X^{(j)} = \ln X_\epsilon^{(i)} - \ln X_\epsilon^{(j)} \tag{24}$$
$$i \neq j \qquad i, j = 1, 2, \cdots, n$$

Notice that, since the magnitude of measurement error is usually small when compared with that of the actual charge amount, i.e., $\epsilon^{(i)} \ll 1$, the following approximations are in general valid

$$\ln X_\epsilon^{(i)} = \ln(1 + \epsilon^{(i)}) \simeq \epsilon^{(i)} \tag{25}$$
$$i = 1, 2, \cdots, n$$

Thus, relations similar to Eqs. 21 can be developed accordingly

$$\mathcal{E}[\epsilon^{(i)}] \simeq \mathcal{E}[\ln X^{(i)} - \ln X^{(r)}] + \mathcal{E}[\epsilon^{(r)}] \tag{26}$$
$$i \neq r \qquad i = 1, 2, \cdots, n$$

In addition, the variances can be expressed as

$$\mathrm{Var}[\epsilon^{(k)}] \simeq \frac{1}{n-1} \sum_{j=1}^{n} \sum_{\substack{i=1 \\ i \neq j}}^{j} \mathrm{Var}[\ln X^{(i)} - \ln X^{(j)}] - \frac{1}{n-2} \sum_{\substack{j=1 \\ j \neq k}}^{n} \sum_{\substack{i=1 \\ i \neq j}}^{j} \mathrm{Var}[\ln X^{(i)} - \ln X^{(j)}] \tag{27}$$

After obtaining the approximated values of Var $[\Xi^{(k)}]$s or Var$[\epsilon^{(k)}]$s, maximum likelihood estimates of the other parameters can be determined according to Eqs. 19, 21 and 26. Notice that the number of decision variables of this optimization problem is reduced to 3, i.e., θ_μ^C, θ_Σ^C and $\theta_\mu^{(r)}$. Thus, the iterative computation process should converge at a much faster rate. Finally, it should be noted that, if *both* the charging and measurement errors are of type A, infinite number of solutions can be produced with this approach. In such case, additional assumptions must be introduced to produce separate estimates of these parameters.

On-Line Target Setting Procedure

Because of charging errors, the plant engineers usually cannot guarantee the products of all batches in a production campaign to be satisfactory. Thus, the probability of a successful charging operation, i.e., reliability, can be considered as a performance index and should be chosen in advance. In practice, perfect operation cannot be expected, i.e., the achievable reliability should be always less than one.

If there is only one operational constraint involved in the charging process, any given level of performance can be achieved with a proper target setting procedure. Since the performance function $\mathcal{G}$ is a function of random variables X^C and Z_j^Cs, the probability of $\mathcal{G} < 0$, i.e., the *demand probability* P_F, can be expressed as

$$P_F = 1 - \int_0^\infty f_{\mathcal{G}}(\eta)d\eta = 1 - \int_{-\gamma}^\infty \tilde{f}_{\tilde{\mathcal{G}}}(\xi)d\xi \tag{28}$$

where

$$\tilde{\mathcal{G}} = \frac{\mathcal{G} - \mathcal{E}[\mathcal{G}]}{\sqrt{\mathrm{Var}[\mathcal{G}]}} \tag{29}$$

$$\gamma = \frac{\mathcal{E}[\mathcal{G}]}{\sqrt{\mathrm{Var}[\mathcal{G}]}} \tag{30}$$

and $f_{\mathcal{G}}$ and $\tilde{f}_{\tilde{\mathcal{G}}}$ are the probability density functions of $\mathcal{G}$ and $\tilde{\mathcal{G}}$ respectively. The lower bound of the integral in Eq. 28, i.e., γ, will later be referred to as the *safety index.*

Notice that X^C and Z_j^Cs are functions of the charging errors only and, also, these charging errors are assumed to follow Gaussian distribution in this study. If the performance function is a linear function of X^C and Z_j^Cs, then $\mathcal{G}$ must also be normally distributed. Thus, the value of γ should be 1.65 if the probability of failure is required to be 0.05 and 3.1 if a probability of 0.001 is expected. However, if the performance function is nonlinear, then the value of safety index associated with a given level of demand probability must be determined with Monte Carlo simulation. This approach requires extensive computation. Thus, an alternative procedure was taken in this study. In particular, $\mathcal{G}$ was linearized by taking Taylor series expansion around μ_{X^c} and $\vec{\mu}_{Z^c} = [\mu_{Z_1^c}, \mu_{Z_2^c}, \ldots, \mu_{Z_m^c}]^T$. The mean and variance of the resulting function were then used as approximations (Kapur and Lamberson, 1991)

$$\mathcal{E}[\mathcal{G}] \simeq \mathcal{G}(\mu_{X^c}, \vec{\mu}_{Z^c}) + \frac{1}{2}\mathrm{Var}[X^C]\left(\frac{\partial^2 \mathcal{G}}{\partial X^{C2}}\right)_{\mu_{X^c}, \vec{\mu}_{Z^c}} + \frac{1}{2}\sum_{j=1}^{m}\mathrm{Var}\left[Z_j^C\right]\left(\frac{\partial^2 \mathcal{G}}{\partial Z_j^{C2}}\right)_{\mu_{X^c}, \vec{\mu}_{Z^c}} \tag{31}$$

$$\mathrm{Var}[\mathcal{G}] \simeq \mathrm{Var}[X^C]\left(\frac{\partial \mathcal{G}}{\partial X^C}\right)^2_{\mu_{X^c}, \vec{\mu}_{Z^c}} + \sum_{j=1}^{m}\mathrm{Var}\left[Z_j^C\right]\left(\frac{\partial \mathcal{G}}{\partial Z_j^C}\right)_{\mu_{X^c}, \vec{\mu}_{Z^c}} \tag{32}$$

Due to the fact that the value of $\mathcal{E}[\mathcal{G}]$ can be easily controlled by adjusting X^T and Z_j^Ts, it is convenient to set the charge targets according to Eq. 30 so that a required level of demand probability P_F can be achieved. In the industry, this task is usually carried out *off-line*. Specifically, the charge targets are determined in advance and maintained constant for every batch throughout the production campaign. However, it is also clear from Eq. 30 that these targets are affected by the variance of $\mathcal{G}$. Thus, if the off-line approach is taken, the corresponding target setting procedure tends to be conservative. This is due to the fact that variabilities associated with X^C and Z_j^Cs must both be accounted for in estimating $\mathrm{Var}[\mathcal{G}]$. Consequently, the plant productivity may be less than maximum due to excessively large amount of unreacted reactant remaining in each batch.

In this study, several *on-line* strategies have been developed for setting the charge target X^T. Notice that, in addition to the statistical data of the previous batches, the measurements of the previously-charged reactants in the *same* batch are also available. Since the purpose for installing sensors is to ensure operational reliability and safety, it is reasonable to believe that the magnitudes of the means and variances of the measurement errors are smaller than those of the charging errors. If one can produce accurate estimates of Z_j^Cs with these on-line data of the current batch, an alternative performance function can be used to replace the original one

$$\hat{\mathcal{G}} = \mathcal{G}\left(\bar{X}^T + \Delta_X; \hat{Z}_1^C - d_1, \hat{Z}_2^C - d_2, \cdots, \hat{Z}_m^C - d_m\right) \tag{33}$$

where $\bar{X}^T$ represents the target of X^C in the current batch and Δ_X denotes the corresponding charging error and $\hat{Z}_j^C$ $(j = 1, 2, \ldots, m)$ is the estimated amount of the jth raw material which has already been transferred into the reactor. Notice that the estimates $\hat{Z}_j^C$s should have been computed before setting $\bar{X}^T$, but their deviations from Z_j^Cs, i.e., the d_js, are still uncertain. If the variances of these estimates can be made smaller than those of Z_j^Cs

$$\mathrm{Var}[d_j] < \mathrm{Var}[\Delta_{Z_j}] \qquad j = 1, 2, \cdots, m \tag{34}$$

then a more aggressive target setting policy can be developed accordingly. Notice that, given the value of target Z_j^T, the variance of Z_j^C is essentially the same as that of the corresponding charging error Δ_{Z_j}.

One of the approaches used in this study for determining $\hat{Z}_j^C$s is to produce least-square estimates on the basis of on-line measurements of the current batch

$$\min_{\hat{Z}_j^C} \sum_{i=1}^{n_j} \frac{\left(\bar{Z}_j^{(i)} - \hat{Z}_j^C - \mathcal{E}\left[\Xi_{Z_j}^{(i)}\right]\right)^2}{\mathrm{Var}\left[\Xi_{Z_j}^{(i)}\right]} \qquad j = 1, 2, \cdots, m \tag{35}$$

or

$$\hat{Z}_j^C = \left(\sum_{i=1}^{n_j} \frac{\bar{Z}_j^{(i)} - \mathcal{E}\left[\Xi_{Z_j}^{(i)}\right]}{\mathrm{Var}\left[\Xi_{Z_j}^{(i)}\right]} \right) \Bigg/ \left(\sum_{i=1}^{n_j} \frac{1}{\mathrm{Var}\left[\Xi_{Z_j}^{(i)}\right]} \right) \quad (36)$$

where n_j is the number of sensors used for Z_j^C and $\bar{Z}_j^{(i)}$ denotes the current measurement value of the ith sensors. The deviations is this case can be expressed by

$$d_j = \left(\sum_{i=1}^{n_j} \frac{\Xi_{Z_j}^{(i)} - \mathcal{E}\left[\Xi_{Z_j}^{(i)}\right]}{\mathrm{Var}\left[\Xi_{Z_j}^{(i)}\right]} \right) \Bigg/ \left(\sum_{i=1}^{n_j} \frac{1}{\mathrm{Var}\left[\Xi_{Z_j}^{(i)}\right]} \right) \quad (37)$$

Also, the means and variances of these deviations can be expressed with the following equations

$$\mathcal{E}[d_j] = 0 \quad (38)$$

$$\mathrm{Var}[d_j] = \left(\sum_{i=1}^{n_j} \frac{1}{\mathrm{Var}\left[\Xi_{Z_j}^{(i)}\right]} \right)^{-1} \quad (39)$$

From Eq. 39, one can clearly see that the on-line strategy is indeed more aggressive if the variability of measurement error is on average smaller than the charging error. Thus, this approach is recommended for increasing the profit margin of batch processes.

Optimal Alarm Generation Logic

Since the expected values and variances of the charging and measurement errors must be estimated with an iterative numerical procedure, the accuracy of parameter estimates cannot always be guaranteed. As a result, the desired level of demand probability P_F may be unachievable even with the proposed target setting strategy. Also, there are sometimes economical incentives to set the targets according to a safety index which is associated with a higher-than-acceptable level of P_F. In these situations, the eventual probability of failed operation can be reduced with an on-line alarm system to detect undesirable batch state after completion of the charging process.

As mentioned before, multiple sensors are used to monitor the charging sequence. On-line measurement data can be substituted into the performance function to determine the status of the current batch. Let us assume that S distinct sets of sensors are chosen for this purpose. For illustration convenience, these sets are collected in a sensor set $\mathfrak{M}$

$$\mathfrak{M} = \{\boldsymbol{m}_s \mid \boldsymbol{m}_s = (i, j_1, \cdots, j_m) \text{ and } s = 1, 2, \cdots, S\} \quad (40)$$

where $i, j_1, \ldots, j_m$ are the labels of the sensors for X^C, Z_1^C, $\ldots$, Z_m^C, respectively. Corresponding to each $\boldsymbol{m}_s \in \mathfrak{M}$, the value of a binary indicator variable y_s can be determined

$$y_s = \begin{cases} 1 & \text{if } \mathcal{G}^{(s)} < 0 \\ 0 & \text{otherwise} \end{cases} \quad (41)$$

$$s = 1, 2, \cdots, S$$

where $\mathcal{G}^{(s)}$ is an *indicator function* whose value can be determined by substituting the sth set of measurement values into the performance function

$$\mathcal{G}^{(s)} = \mathcal{G}(X^{(i)}; Z_1^{(j_1)}, \cdots, Z_m^{(j_m)}) \quad (42)$$

$$i, j_1, \cdots, j_m \in \boldsymbol{m}_s$$

and $X^{(i)}$, $Z_1^{(j_1)}$, $\cdots$, $Z_m^{(j_m)}$ are the on-line measurements of X^C, Z_1^C, $\cdots$, Z_m^C. The system alarm can then be generated on the basis of these indicators. The logic for setting off the alarm can be explicitly expressed with an alarm function $f(\boldsymbol{y})$

$$f(\boldsymbol{y}) = \begin{cases} 1 & \text{if the system is generating an alarm} \\ 0 & \text{otherwise} \end{cases} \quad (43)$$

where $\boldsymbol{y} = [y_1, y_2, \cdots, y_s]^T$.

Obviously, the values of the indicator variables $y_s s$ may not be consistent with the true batch state after charging. Specifically, let us consider the true value of the performance function

$$\mathcal{G}^C = \mathcal{G}(X^C; Z_1^C, Z_2^C, \cdots, Z_m^C) \quad (44)$$

There are two kinds of mistakes that can be identified accordingly, i.e., y_s is set to be 1 when $\mathcal{G}^C \geq 0$ (type I mistake) or y_s is set to be 0 when $\mathcal{G}^C < 0$ (type II mistake). Similarly, the mistakes committed in generating the *system alarm* can also be classified into types I and II. The conditional probabilities associated with these two mistakes, i.e., P_a and P_b, can be expressed as

$$P_a = Pr\{f(\boldsymbol{y}) = 1 \mid \mathcal{G}^C \geq 0\} \quad (45)$$

$$P_b = Pr\{f(\boldsymbol{y}) = 0 \mid \mathcal{G}^C < 0\} \quad (46)$$

Since both types of mistakes result in financial losses, there are incentives for developing an optimal alarm generation logic which minimizes the expected loss $\mathcal{L}$

$$\min_{f(\boldsymbol{y})} \mathcal{L} \quad (47)$$

where

$$\mathcal{L} = C_a(1 - P_F)P_a + C_b P_F P_b \quad (48)$$

where C_a and C_b respectively denote the losses caused by types I and II mistakes in alarm generation. Notice that the conditional probabilites P_a and P_b can also be written as

$$P_a = \mathcal{E}[f(\boldsymbol{y}) \mid \mathcal{G}^C \geq 0] = \sum_{\boldsymbol{y}} f(\boldsymbol{y}) Pr\{\boldsymbol{y} \mid \mathcal{G}^C \geq 0\} \quad (49)$$

$$P_b = \mathcal{E}[1 - f(\boldsymbol{y}) \mid \mathcal{G}^C < 0] = \sum_{\boldsymbol{y}} [1 - f(\boldsymbol{y})] Pr\{\boldsymbol{y} \mid \mathcal{G}^C < 0\} \quad (50)$$

Thus, the expected loss becomes

$$\mathfrak{L} = C_b P_F - \sum_y f(y)h(y) \tag{51}$$

where

$$h(y) = C_b P_F Pr\{y \mid \mathcal{G}^C < 0\} - C_a(1 - P_F) Pr\{y \mid \mathcal{G}^C \geq 0\} \tag{52}$$

Notice that the first term on the righthand side of Eq. 51 represents the expected loss of no-sensor system. Thus, it is apparent that the expected loss is minimized if the alarm function is chosen such that

$$f(y) = \begin{cases} 1 & \text{if } h(y) > 0 \\ 0 & \text{if } h(y) \leq 0 \end{cases} \tag{53}$$

After obtaining the values of $f(y)$ for all possible y, its functional form can be constructed accordingly. With the functional form given, the logic associated with $f(y)$ can be implemented as a hard-wired circuit or as a computer program.

It should be noted that in order to compute $h(y)$ and then construct $f(y)$ the estimates of conditional probabilities $Pr\{y \mid \mathcal{G}^C < 0\}$ and $Pr\{y \mid \mathcal{G}^C \geq 0\}$ must be obtained first. For convenience, it is assumed in this study that the S sets of measurements in $\mathfrak{M}$ are statistically independent. Thus

$$Pr\{y \mid \mathcal{G}^C \geq 0\} = \prod_{s=1}^{S} a_s^{y_s} (1 - a_s)^{1 - y_s} \tag{54}$$

$$Pr\{y \mid \mathcal{G}^C < 0\} = \prod_{s=1}^{S} (1 - b_s)^{y_s} b_s^{1 - y_s} \tag{55}$$

where

$$a_s = Pr\{y_s = 1 \mid \mathcal{G}^C \geq 0\} \tag{56}$$

$$b_s = Pr\{y_s = 0 \mid \mathcal{G}^C < 0\} \tag{57}$$

An estimation procedure has been developed in this study for computing the conditional probabilities a_ss and b_ss. This procedure is explained in detail in the Appendix.

Finally, it should be pointed out that the sensor set $\mathfrak{M}$ may not be unique. It is possible to identify different combinations of s-independent sets $\boldsymbol{m}_s$ from all available sensors. Thus, the alarm logic is also affected by the sensors selected for determining the indicator variables y_ss and the optimal system design should be obtained by

$$\min_{\mathfrak{M}_\kappa} \min_{f(y)} \mathfrak{L} \tag{58}$$

where $\mathfrak{M}_\kappa$ (κ = I, II, III,...) denotes the κth sensor set.

Application Example

The example presented here was designed to illustrate the implementation procedure of the proposed techniques. Since the actual amount of each reactant charged to the reactor is always unknown in practice and, also, there is an obvious need to quantify the benefits of adopting the present approach, a fictitious system is adopted in this article for the purpose of producing simulated data.

Problem statement

Let us consider a batch process in which two reactions take place

$$\alpha_R A + \beta_R B \xrightarrow{k_1} R \tag{59a}$$

$$\alpha_S A + \beta_S B \xrightarrow{k_2} S \tag{59b}$$

where R is the batch product; S is an unwanted byproduct; A and B are the reactants; α_R, α_S, β_R and β_S are stoichiometric coefficients of reactants A and B in the above two reactions respectively. The rate equations of these two reactions are

$$r_1 = \frac{dC_R}{dt} = k_1 \left(\frac{1}{\alpha_R}\right) C_A C_B^2 \tag{60a}$$

$$r_2 = \frac{dC_S}{dt} = k_2 \left(\frac{1}{\alpha_S}\right) C_A C_B \tag{60b}$$

Let us further assume that reactant B is hazardous. Thus, the amount of B should be kept to a minimum after the batch reaction is terminated. It can be shown from Eqs. 59 and 60 that

$$C_{Af} - \left(\frac{\alpha_R}{\beta_R}\right) C_{Bf} = C_{A0} - \left(\frac{\alpha_R}{\beta_R}\right) C_{B0} - \left(\frac{\alpha_R}{\beta_R} - \frac{\alpha_S}{\beta_s}\right) \Re \ln \frac{\Re}{C_{B0} + \Re} \tag{61}$$

where

$$\Re = \left(\frac{\alpha_R}{\beta_R}\right)\left(\frac{\beta_S}{\alpha_S}\right)\left(\frac{k_2}{k_1}\right)$$

Also, C_{A0} and C_{B0} denote the initial concentrations of reactants A and B, respectively; C_{Af} and C_{Bf} represent the final concentrations of the two reactants. From Eqs. 60a and 60b, one can see that the reaction rate should be very slow at the end of batch if both C_{Af} and C_{Bf} approach zero. However, from a practical standpoint, it is necessary to complete the batch within a reasonable amount of time. One of the methods that can be adopted to achieve this purpose is to keep the ratio C_{A0}/C_{B0} sufficiently high during the charging process so that

$$C_{Af} - \left(\frac{\alpha_R}{\beta_R}\right) C_{Bf} \geq c \tag{62}$$

where c is a chosen constant. On the other hand, it is apparent that the excess of reactant A initially charged to the reactor should not be too great. This is due to the need to avoid an unnecessary loss of raw material A and a reduction in the yield of product R.

Operational Constraint

By assuming reactant A is the first one charged, we can formulate a performance function according to Eqs. 61 and 62

$$\mathcal{G}(X_B^C; Z_A^C) = \left(\frac{\alpha_R}{\beta_R} - \frac{\alpha_S}{\beta_S}\right) \Re \ln\left[\frac{\rho_A \rho_B X_B^C}{\Re M_B(\rho_B Z_A^C + \rho_A X_B^C)} + 1\right] + \left(\frac{\rho_A \rho_B}{M_A M_B}\right) \frac{M_B Z_A^C - (\alpha_R/\beta_R) M_A X_B^C}{\rho_B Z_A^C + \rho_A X_B^C} - c \quad (63)$$

where Z_A^C represents the weight (kg) of A which has already charged to the reactor; X_B^C denotes the weight (kg) of B fed to the reactor in the present charging sequence; ρ_A and ρ_B are the densities of A and B, respectively; M_A and M_B are the molecular weights of the two reactants. In this example, the following parameter values were adopted

$\alpha_R = 1$ $\quad$ $\beta_R = 2$

$\alpha_S = 3$ $\quad$ $\beta_S = 2$

$M_A = 190.66\ (\text{g}\cdot\text{mol}^{-1})$ $\quad$ $M_B = 139.12\ (\text{g}\cdot\text{mol}^{-1})$

$\rho_A = 1.30\ (\text{g}\cdot\text{cm}^{-3})$ $\quad$ $\rho_B = 1.46\ (\text{g}\cdot\text{cm}^{-3})$

$c = 1.5\times10^{-5}\ (\text{mol}\cdot\text{cm}^{-3})$

$k_1 = 4.7\times10^{3}\ (\text{mol}^{-2}\cdot\text{cm}^{6}\cdot\text{s}^{-1})$

$k_2 = 9.6\times10^{-2}\ (\text{mol}^{-1}\ \text{cm}^{3}\cdot\text{s}^{-1})$

Error models

The charging errors associated with both A and B in this case are assumed to be of type B

$$Z_A^C = Z_A^T Z_A^\delta \quad (64a)$$

$$X_B^C = X_B^T X_B^\delta \quad (64b)$$

where Z_A^T and X_B^T are the targets of reactants A and B, respectively and Z_A^δ and X_B^δ denotes the corresponding charging errors. It is further assumed that three independent sensors are used for each reactant and the measurement errors are all of type A

$$Z_A^{(i)} = Z_A^C + \Xi_{Z_A}^{(i)} \quad i = 1,2,3 \quad (65a)$$

$$X_B^{(j)} = X_B^C + \Xi_{X_B}^{(j)} \quad j = 1,2,3 \quad (65b)$$

where, $\Xi_{Z_A}^{(i)}$ and $\Xi_{X_B}^{(j)}$ are the measurement errors associated with the ith sensor of reactant A and jth sensor of reactant B, respectively.

In order to simulate the batch charging process with a random number generator, the means and variances of the random variables in the error models, i.e., Z_A^δ, X_B^δ, $\Xi_{Z_A}^{(i)}$s, and $\Xi_{X_B}^{(j)}$, must be specified. These data are in Tables 1a and 1b.

Parameter estimation

In realistic operation, the statistics of the charging and measurement errors must be obtained from historical data. Using subroutine DRNNOA in IMSL (Kinderman and Ramage, 1976), these data have been created according to the targets specified for each batch and the parameters listed in Tables 1a and 1b. A total of 320 "previous" batches have been simulated in this example.

By making use of Eqs. 23, the variances of all measurement errors can be estimated first. Then, maximum likelihood estimates of the rest of the parameters associated with the charging and measurement errors can be determined on the basis of Eqs. 19 and 21. The results of the iterative calculation are presented in Tables 2a and 2b.

Target setting procedures

Having obtained the estimated parameters, one can then compute the charging targets for "future" batches. First, it should be noted that Eq. 30 is the foundation of all targeting methods and thus the mean and variance of the performance function must be estimated in advance. Also, since this function in the present example (Eq. 63) is nonlinear, the linearization techniques suggested in Eqs. 31 and 32 have been adopted in both the off-line and on-line target-setting processes.

In order to quantitatively demonstrate the advantages and disadvantages of various approaches developed in the article, additional simulation studies have been carried out. The detailed simulation procedure and the corresponding results are presented as follows:

Off-Line Strategy. Without evaluating the actual charge amount of A in every batch, Eq. 30 can be solved in advance to determine a constant X_B^T for use throughout the production campaign according to a set of given values of Z_A^T and γ. In our studies, 1,024 batches have been simulated with the same target for reactant A, i.e., $Z_A^T = 18{,}638$ kg.

If the consequences of violating the operational constraint are extremely serious, then one may wish to choose a safety

Table 1a. Means and Variances of Charging and Measurement Errors Associated with Reactant *A*

Parameters	Z_A^δ	$\Xi_{Z_A}^{(1)}$	$\Xi_{Z_A}^{(2)}$	$\Xi_{Z_A}^{(3)}$
Mean	1.1030	-2.4232×10^{2}	5.8252×10^{3}	-4.9648×10^{3}
Variance	2.6293×10^{-4}	6.0225×10^{4}	3.9931×10^{4}	2.4572×10^{4}

Table 1b. Means and Variances of Charging and Measurement Errors Associated with Reactant *B*

Parameters	X_B^δ	$\Xi_{X_B}^{(1)}$	$\Xi_{X_B}^{(2)}$	$\Xi_{X_B}^{(3)}$
Mean	1.1310	-4.3240×10^{2}	7.5857×10^{3}	-8.2824×10^{3}
Variance	1.2212×10^{-4}	4.3096×10^{4}	5.0022×10^{4}	4.4159×10^{4}

Table 2a. Estimated Means and Variances of Charging and Measurement Errors Associated with Reactant A

Parameters	Z_A^δ	$\Xi_{Z_A}^{(1)}$	$\Xi_{Z_A}^{(2)}$	$\Xi_{Z_A}^{(3)}$
Est. mean	1.1030	-2.4727×10^{2}	5.8436×10^{3}	-4.9686×10^{3}
Est. variance	2.6128×10^{-4}	6.0826×10^{4}	3.7412×10^{4}	3.1001×10^{4}

Table 2b. Estimated Means and Variances of Charging and Measurement Errors Associated with Reactant B

Parameters	X_B^δ	$\Xi_{X_B}^{(1)}$	$\Xi_{X_B}^{(2)}$	$\Xi_{X_B}^{(3)}$
Est. mean	1.1268	-3.4201×10^{2}	8.4896×10^{3}	-7.3784×10^{3}
Est. variance	1.3129×10^{-4}	4.0878×10^{4}	4.6118×10^{4}	4.5346×10^{4}

index γ so that the demand probability P_F is kept under a very low level. For example, the value of X_B^T corresponding to $P_F = 10^{-6}$ can be determined to be 23,956 kg. On the other hand, if the profit of producing more product per batch is quite high and the loss due to improper charging operation is not overwhelming, there may be incentives to accept a higher demand probability. As another example, the target for reactant B can be raised to 25,326 kg if $P_F = 0.05$. This change represents a 5.7% increase in productivity.

The correctness of the proposed target setting procedure has been verified with simulation studies. The simulation data of reactants A and B were both obtained with a standard random number generator. The percentage of failed batches, i.e., those in which the outcome of charging operation violates the operation constraint, in the latter example was found to be around 4.5%. This is very close to the target value of P_F. A sample (first 100 batches) of the simulation results is shown in Figure 2a. Note that the data points below the horizontal dashed line, i.e., $\mathcal{G}^C < 0$, represent failed batches.

On-Line Strategy. The simulation data of reactant A used in this case were essentially the same as those adopted for testing the off-line strategy. Having obtained the simulated on-line measurements of A, the value of $\hat{Z}_A^C$ was estimated in every batch according to Eq. 36. Using a demand probability of 0.05, the target of reactant B for every batch was then determined by solving Eq. 30. Finally, the simulated data of reactant B were produced with a random number generator. The percentage of failed batches was again found to be very close to the target demand probability. Its value is approximately 4.2% this time. A sample of the simulation results for this case is provided in Figure 3a. Notice that, when compared with Figure 2a, the average value of $\mathcal{G}^C$ in this figure is closer to zero and the corresponding data variability is significantly smaller. This phenomenon is precisely the result of adopting the on-line estimates $\hat{Z}_A^C$ for calculating X_B^T. Using Eq. 39 the variance of its deviation from the actual value was estimated to be 1.3258×10^4. This value is much smaller than the estimated variance of the charging error (9.1358×10^4), which can be determined on the basis of the following equation

$$\mathrm{Var}[\Delta_{Z_A}] = (Z_A^T)^2 \mathrm{Var}[Z_A^\delta] \tag{66}$$

It should also be noted that the average value of X_B^T now can be raised to 25,562 kg. This result shows that, when compared with the off-line strategy, the profit margin of the batch process can be increased with the on-line approach while still maintaining the same target demand probability. The targets of reactant B used in the first 100 batches are plotted in Figure 4.

Alarm generation logic

As mentioned before, the outcome of reactant-charging operation can be monitored with on-line sensors. All possible sensor sets used in the present example are listed in Table 3. Also, notice that the task of constructing alarm generation logic is essentially equivalent to that of determining the form of the alarm function $f(y)$. To achieve this purpose, one can conclude from Eqs. 52 to 57 that the conditional probabilities

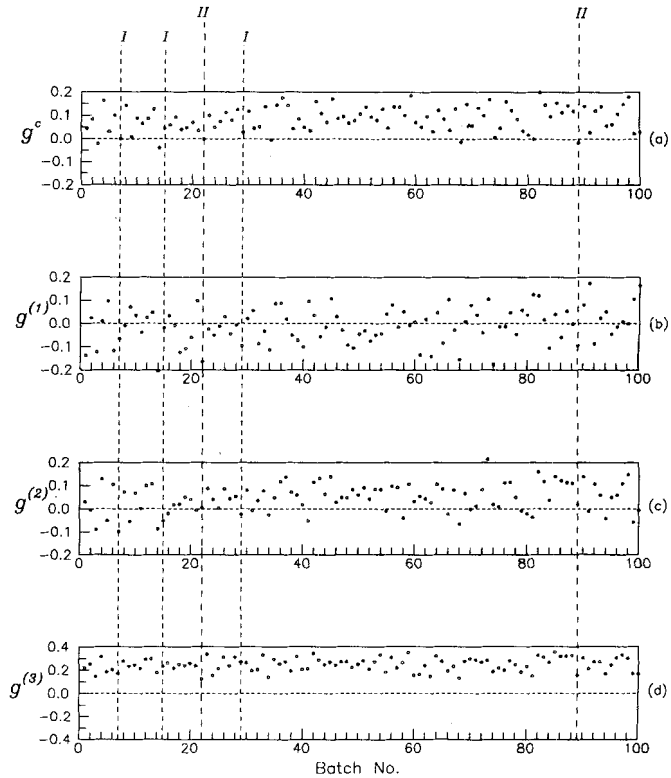

Figure 2. Sample of the simulation results obtained with the off-line target setting strategy ($P_F = 0.05$) and stationary alarm logic.

(a) Actual values of the performance function $\mathcal{G}^C$; (b) values of the indicator function $\mathcal{G}^{(1)}$ corresponding to the set m_1; (c) values of the indicator function $\mathcal{G}^{(2)}$ corresponding to the set m_2; (d) values of the indicator function $\mathcal{G}^{(3)}$ corresponding to the set m_3.

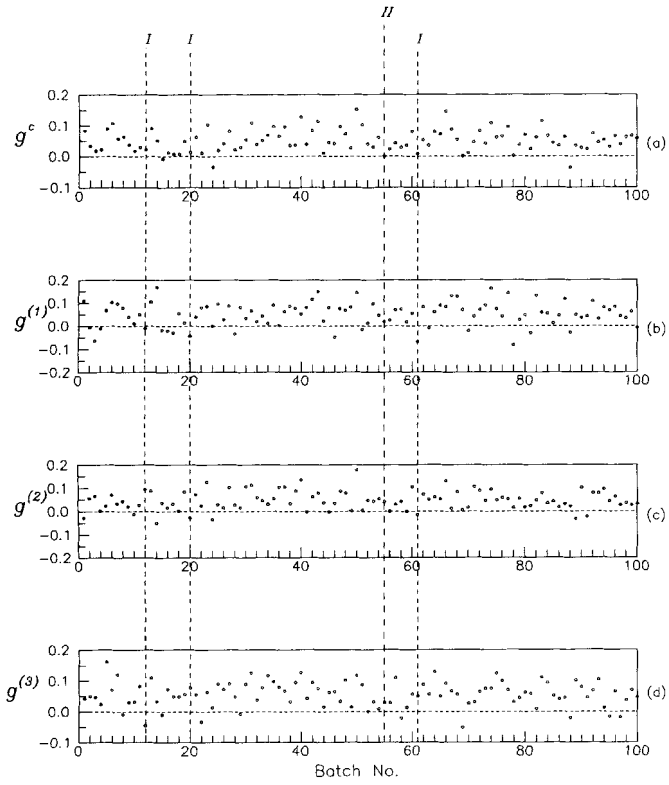

Figure 3. Sample of the simulation results obtained with the on-line target setting strategy ($P_F = 0.05$) and dynamic alarm logic.

(a) Actual values of the performance function $\mathcal{G}^C$; (b) values of the indicator function $\mathcal{G}^{(2)}$ corresponding to the set m_1; (c) values of the indicator function $\mathcal{G}^{(2)}$ corresponding to the set m_2; (d) values of the indicator function $\mathcal{G}^{(3)}$ corresponding to the set m_3.

associated with type I and type II mistakes must be evaluated first. In other words, if all measurements are statistically independent, the following quantities must be computed in advance

$$\bar{a}_{ij} = Pr\{g_{ij} < 0 \mid \mathcal{G}^C \geq 0\} \tag{67a}$$

$$\bar{b}_{ij} = Pr\{g_{ij} \geq 0 \mid \mathcal{G}^C < 0\} \tag{67b}$$

$$i, j = 1, 2, 3$$

where

$$g_{ij} = \mathcal{G}(X_B^{(i)}; Z_A^{(j)}) \tag{68}$$

It should be noted that within a given sensor set $\mathfrak{M}_\kappa$, the conditional probabilities defined in Eqs. 56 and 57 can be related to these quantities by

$$a_s = \bar{a}_{i'j'} \tag{69a}$$

$$b_s = \bar{b}_{i'j'} \tag{69b}$$

and

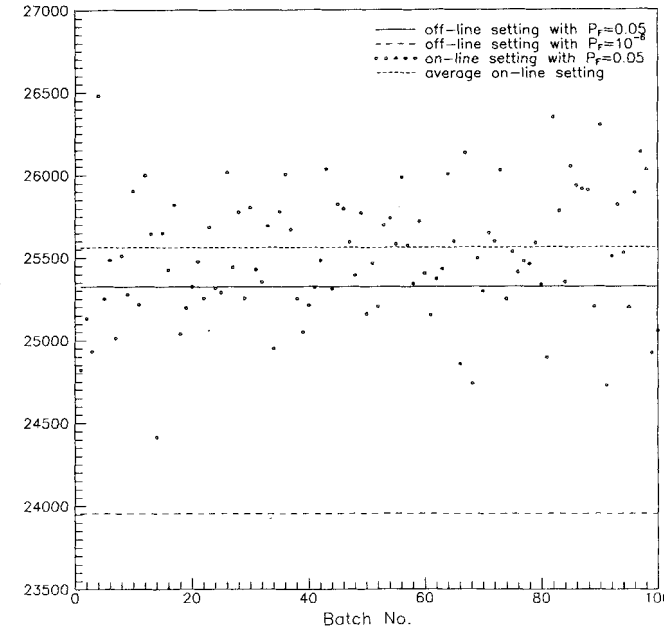

Figure 4. Comparison of target settings determined with different strategies.

$$m_s = (i', j') \in \mathfrak{M}_\kappa \tag{69c}$$

These quantities can certainly be computed with the equations provided in the Appendix. Also, from Eqs. A5 to A8 in the Appendix, one can see that the conditional probabilities a_s and b_s are really functions of the targets for reactant A and B. Thus, if the off-line target setting procedure is adopted in operation, estimates of these probabilites should be unchanged throughout the production compaign. However, if the on-line strategy is implemented, the values of a_s and b_s should vary from batch to batch and the alarm generation logic may change accordingly. Thus, the former alarm configuration is referred to as the *stationary scheme* and the latter as the *dynamic scheme* in this example.

Stationary Scheme. Corresponding to a demand probability of 0.05, the targets of reactant A and B have already been determined in this example, i.e., $Z_A^T = 18{,}638$ kg and $X_B^T = 25{,}326$ kg. The conditional probabilities defined in Eqs. 67a and 67b can be computed according to these targets and the parameters listed in Tables 2a and 2b. The results are

$$\bar{A} = (\bar{a}_{ij})_{3\times 3} = \begin{bmatrix} 6.7835\times10^{-2} & 6.8400\times10^{-1} & 1.2293\times10^{-2} \\ 6.6607\times10^{-6} & 5.9823\times10^{-2} & 5.2357\times10^{-8} \\ 1.7174\times10^{-1} & 8.6689\times10^{-1} & 4.1092\times10^{-2} \end{bmatrix}$$

and

Table 3. Sensor Sets

	κ					
s	I	II	III	IV	V	VI
1	(1, 1)	(1, 1)	(1, 2)	(1, 2)	(1, 3)	(1, 3)
2	(2, 2)	(2, 3)	(2, 1)	(3, 1)	(2, 2)	(3, 2)
3	(3, 3)	(3, 2)	(3, 3)	(2, 3)	(3, 1)	(2, 1)

Table 4. Minimum Expected Losses

κ	I	II	III	IV	V	VI
$\mathcal{L}/C_a$	0.1601	0.2929	0.2787	0.1564	0.1683	0.5031

$$\bar{B} = (\bar{b}_{ij})_{3\times 3} = \begin{bmatrix} 3.1899\times 10^{-1} & 5.7517\times 10^{-4} & 6.6944\times 10^{-1} \\ 9.8911\times 10^{-1} & 2.6150\times 10^{-1} & 9.9903\times 10^{-1} \\ 5.9645\times 10^{-2} & 2.4817\times 10^{-7} & 3.4222\times 10^{-1} \end{bmatrix}$$

In this example, the ratio of the loss caused by type II mistakes to that due to type I mistakes, i.e., C_b/C_a, is assumed to be 15. On the basis of this assumption and the conditional probabilities determined with Eqs. 69a to 69c, the minimum expected losses associated with all sensor sets listed in Table 3 can be calculated with Eqs. 49 to 58 (Table 4). The best sensor set among the six possible candidates can then be selected accordingly. From Table 4, it is apparent that $\mathfrak{M}_{IV}$ should be our choice. The corresponding alarm function $f(\mathbf{y})$ can be determined by computing function $h(\mathbf{y})$ with Eqs. 52 to 55.

$$\begin{aligned} h(0,0,0) &= -2.4873\times 10^{-1} \Rightarrow f(0,0,0) = 0 \\ h(1,0,0) &= -4.9415\times 10^{-8} \Rightarrow f(1,0,0) = 0 \\ h(0,1,0) &= -5.1179\times 10^{-2} \Rightarrow f(0,1,0) = 0 \\ h(0,0,1) &= \ \ 1.1769\times 10^{-7} \Rightarrow f(0,0,1) = 1 \\ h(1,0,1) &= \ \ 5.8592\times 10^{-1} \Rightarrow f(1,1,0) = 1 \\ h(1,0,1) &= \ \ 4.3053\times 10^{-5} \Rightarrow f(1,0,1) = 1 \\ h(0,1,1) &= \ \ 3.8818\times 10^{-1} \Rightarrow f(0,1,1) = 1 \\ h(1,1,1) &= \ \ 6.7920\times 10^{-4} \Rightarrow f(1,1,1) = 1 \end{aligned}$$

Thus, the alarm function should be of the form

$$f(y_1, y_2, y_3) = 1 - (1 - y_3)(1 - y_1 y_2) \qquad (70)$$

which is a standard OR-AND system.

Extensive numerical simulation studies have also been carried out to test this alarm logic. The data previously used for evaluating the performance of off-line targeting procedures were again adopted in the present case. A sample of the results is presented in Figures 2b to 2d. It was found in our simulation that the percentage of type I mistakes is about 16.70% and that of the type II is 0.49%. Note that the batches corresponding to types I and II mistakes are marked with vertical dashed lines. For example, the first mistakes in these figures is of type I. One can see from Figure 2a that the corresponding batch operation is successful. However, from Figures 2b to 2d, one can also determine the values of the indicator variables according to $\mathcal{G}^{(s)}$ $(s = 1,2,3)$, i.e., $y_1 = y_2 = 1$ and $y_3 = 0$. Obviously, the alarm must be mistakenly set off on the basis of Eq. 70 in this case. As another example, the last mistake is of type II. The values of corresponding indicator variables are 1, 0 and 0 respectively and, thus, there should be no alarm. However, as indicated in Figure 2a, the corresponding batch failed to satisfy the operation constraint.

It should be noted that, without implementing the alarm logic, only type II mistakes are possible in the charging operation. The corresponding probability is essentially the demand probability P_F. Thus, one can clearly observe that the alarm system can affect the batch reaction process in two different ways. Certainly, the major advantage of the alarm is that the probability of type II mistakes can be further reduced. On the other hand, it also causes an additional loss due to type I mistakes. The relatively high percentage of type I mistakes can be attributed to the fact that a C_b to C_a ratio of 15 is used in this example. Since type II mistakes cost much more, it is not suprising to find that the optimal logic tends to place more emphasis on suppressing type II mistakes and less on type I.

Dynamic Scheme. If the on-line strategy is used for setting targets, the corresponding alarm generation logic may have to be changed from batch to batch. Basically, the computation steps described in the previous section have to be repeated each time a new X_B^T is determined.

Again, extensive simulation studies have been carried out. The data adopted here are essentially the same as those previously used for testing the on-line targeting procedures. A sample of the simulation results is provided in Figures 3b to 3d. It was found in our simulation that the sensor set $\mathfrak{M}_I$ is the best choice for almost every batch and the corresponding optimal alarm logic is virtually unchanged. The averaged expected loss was computed to be 0.1641. The alarm function adopted in most of the batches was

$$f(y_1, y_2, y_3) = (1 - y_1) y_2 y_3 + y_1 (1 - y_2) y_3 + y_1 y_2 (1 - y_3) + y_1 y_2 y_3 \qquad (71)$$

This alarm configuration is simply a typical two-out-of-three logic. From the results obtained in simulation, the failed batches caused by type I and type II mistakes can be easily identified. The percentages of these two types of failures were found to be 3.31% and 2.25% respectively. Thus, when compared with the stationary scheme, the present alarm system is superior in the sense that type I mistakes can be reduced considerably. However, the chance of committing type II mistakes in alarm generation may at the same time become higher. This can be attributed to the fact that 2-out-of-3 strategy is a more "conservative" than the OR-AND logic in the sense that the former places more emphasis on suppressing type I mistakes but less on type II mistakes. For example, when $y_1 = y_2 = 0$ and $y_3 = 1$, an alarm will be generated with the latter logic but not the former one.

Economic assessment

From the above discussions, one can see that the selection of the most appropriate target setting procedure and the corresponding alarm generation logic has to be made on the basis of three quantitative economic criteria, i.e., the profit due to productivity increase and the losses due to type I and type II mistakes. Three alternative were considered. The off-line target setting strategy was used in the first two systems. The demand probability of the first was set at 10^{-6}. Due to the extremely low probability of violating the operational constraint, it was decided not to implement any alarm logic in this situation. The demand probability adopted in the second system was 0.05. The previously described stationary scheme was assumed to be used for detecting the charging failures

Table 5. Economic Parameters

Case No.	P_R (\$/kg)	C_a ($\times 10^3$ \$/Batch)	C_b ($\times 10^3$ \$/batch)
A	4.00	4.2	63.0
B	0.55	4.2	63.0
C	0.20	4.2	63.0

for the second candidate. In the third system, the on-line target setting procedure was applied with a demand probability of 0.05. Consequently, the dynamic alarm generation scheme must be used for this candidate system.

In order to evaluate the profit of productivity increase, production rate of the batch process must be determined first. Corresponding to the given target(s) for reactant A and a specific target setting procedure for reactant B, the total amount of product R produced in a production campaign X_R^{TOT} can be computed from the corresponding simulation results. In the present example, three different values, 4.6342×10^7 kg, 4.9014×10^7 kg, and 4.9535×10^7 kg, were found for the three candidates, respectively. The net profit created after completing a campaign (P^N) can be calculated with the following equation

$$P^N = P_R X_R^{\mathrm{TOT}} - C_a N_a - C_b N_b \tag{72}$$

where P_R denotes the profit of producing one kilogram of R and N_a and N_b represent the numbers of failed batches corresponding to type I and type II mistakes respectively. From Eq. 72, it is obvious that the net profit is dependent upon the values of three parameters P_R, C_a, and C_b. Three cases were studied in this example (see Table 5). The net profits corresponding to the three candidate systems were computed accordingly and also presented in Table 6.

From the results presented in Table 6, it can be concluded that the best choice of batch reactor charging system is really process-dependent. If the profit of productivity increase dominates, the combination of on-line target setting procedure and dynamic alarm generation scheme should be the most favorable operating strategy. If, on the other hand, this profit is very low when compared with the losses due to operational failures, a conservative off-line target setting policy should be adopted without alarm. Between the above two extremes, the second strategy, i.e., using an aggressive off-line target in conjunction with a stationary alarm scheme, may become more desirable.

Table 6. Net Profits ($\times 10^6$ \$/Campaign)

Case No.	System No.	Net Profit
A	1	185.37
	2	195.07
	3	196.17
B	1	25.49
	2	25.97
	3	25.27
C	1	9.27
	2	8.81
	3	7.66

Conclusions

Several statistical operating strategies for charging the batch reactors are presented in this article. Based on measurement data, either an off-line or on-line target setting procedure can be implemented to achieve a given level of reliability. In addition, the optimal alarm generation system can be installed to reduce the probability of undetected charge failures. The results of implementing the suggested strategies to the application example show that the approach taken in this study is feasible and effective. Further, when compared with the current practice in the industry, these strategies are superior under the condition that a higher-value-added product is produced in the process.

Notation

a_s = conditional probability of type I sensor mistakes corresponding to the set $\boldsymbol{m}_s$
b_s = conditional probability of type II sensor mistakes corresponding to the set $\boldsymbol{m}_s$
C_a = financial loss resulted from type I mistakes
C_b = financial loss resulted from type II mistakes
C_A, C_B = molar concentrations of components A and B, respectively
C_R, C_S = molar concentrations of components R and S, respectively
d_j = deviation of estimate $\hat{Z}_j^C$ from its true value Z_j^C
$\mathcal{E}[\cdot]$ = mean operator
$f(\cdot)$ = alarm function
$\mathcal{G}(\cdot)$ = performance function
$\hat{\mathcal{G}}(\cdot)$ = performance function form adopted for implementing the on-line target setting strategy
$\tilde{\mathcal{G}}$ = random variable obtained by normalizing $\mathcal{G}$
$\mathcal{G}^{(s)}$ = indicator function for the sensors in set $\boldsymbol{m}_s$
$\mathcal{G}^C$ = true value of the performance function
k_1, k_2 = rate constants
$L(\cdot)$ = maximum likelihood function
$\mathcal{L}$ = expected loss due to failed operation
m = number of reactants which have already been charged in the current batch
M_A, M_B = molecular weights of reactants A and B, respectively
n = total number of sensors used for monitoring the same reactant
N_a = number of failed batches resulted from type I mistakes
N_b = number of failed batches resulted from type II mistakes
P_a = conditional probability associated with type I system mistakes
P_b = conditional probability associated with type II system mistakes
P^N = net profit of a production campaign
P_R = profit per kg of product R
Var[·] = variance operator
$x_k^{(i)}$ = measurement value of sensor i obtained from the kth batch
X^C = amount of a reactant actually charged into the reactor
$X^{(i)}$ = measurement of X^C from the ith sensor
X^T = target value of X^C
$\bar{X}^T$ = target value of X^C determined with the on-line strategy
X_R^{TOT} = total amount of product R produced in a campaign
y_s = binary indicator variable corresponding to the sensors in set $\boldsymbol{m}_s$.
Z_j^C = actual amount of the jth reactant which has already been charged into the reactor
$\hat{Z}_j^C$ = estimate of Z_j^C
$Z_j^{(i)}$ = measurement of Z_j^C obtained from the ith sensor
$\bar{Z}_j^{(i)}$ = measurement values of Z_j^C from the ith sensor in the current batch
Z_j^T = target of Z_j^C

Greek letters

α_R, α_S = stoichiometric coefficients of reactant A, primary and side reaction, respectively
β_R, β_S = stoichiometric coefficients of reactant B, primary and side reaction, respectively
γ = safety index
δ = charging error of type B
Δ = charging error of type A
$\epsilon^{(i)}$ = measurement error of type B associated with sensor i
$\boldsymbol{\theta}_\mu$ = vector of the expected values for the charging and measurement errors
$\boldsymbol{\theta}_\Sigma$ = vector of the variances for the charging and measurement errors
$\vec{\mu}_k$ = mean vector for the kth batch run
$\Xi^{(i)}$ = measurement error of type A associated with sensor i
ρ_A, ρ_B = densities of reactants A and B, respectively
Σ_k = covariance matrix for the kth batch run

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Appendix

Let us first assume that the performance function $\mathcal{G}^C$ is linear or can be approximated by a linear function. Alternative formulations of the function can be written accordingly

$$\mathcal{G}^C = \mathcal{G}^T + \mathcal{G}_\Delta \tag{A1}$$

or

$$\mathcal{G}^C = \mathcal{G}^{(s)} - \mathcal{G}_\Xi^{(s)} \tag{A2}$$
$$s = 1, 2, \cdots, S$$

where

$$\mathcal{G}^T = \mathcal{G}(X^T; Z_1^T, Z_2^T, \cdots, Z_m^T)$$
$$\mathcal{G}_\Delta = \mathcal{G}(\Delta_X; \Delta_{Z_1}, \Delta_{Z_2}, \cdots, \Delta_{Z_m})$$
$$\mathcal{G}^{(s)} = \mathcal{G}(X^{(i)}; Z_1^{(j_1)}, Z_2^{(j_2)}, \cdots, Z_m^{(j_m)})$$
$$\mathcal{G}_\Xi^{(s)} = \mathcal{G}\left(\Xi_X^{(i)}; \Xi_{Z_1}^{(j_1)}, \Xi_{Z_2}^{(j_2)}, \cdots, \Xi_{Z_m}^{(j_m)}\right)$$
$$i, j_1, j_2, \cdots, j_m \in i_s$$

Notice that the above formulations are independent of the error types associated with the charging and measurement processes.

Let us further assume that the charging and measurement errors are normally distributed. Thus, the probability density functions of $\mathcal{G}_\Delta$ and $\mathcal{G}_\Xi^{(s)}$ can be written as

$$p(\mathcal{G}_\Delta) = \frac{1}{\sqrt{2\pi}\,\sigma_{\mathcal{G}_\Delta}} \exp\left\{-\frac{(\mathcal{G}_\Delta - \mu_{\mathcal{G}_\Delta})^2}{2\sigma_{\mathcal{G}_\Delta}^2}\right\} \tag{A3}$$

$$p(\mathcal{G}_\Xi^{(s)}) = \frac{1}{\sqrt{2\pi}\,\sigma_{\mathcal{G}_\Xi^{(s)}}} \exp\left\{-\frac{\left(\mathcal{G}_\Xi^{(s)} - \mu_{\mathcal{G}_\Xi^{(s)}}\right)^2}{2\sigma_{\mathcal{G}_\Xi^{(s)}}^2}\right\} \tag{A4}$$

From Eqs. 59 and 60, one can express the conditional probabilities a_s and b_s with

$$a_s = \frac{Pr\{\mathcal{G}^{(s)} < 0,\ \mathcal{G}^C \geq 0\}}{Pr\{\mathcal{G}^C \geq 0\}} \tag{A5}$$

$$b_s = \frac{Pr\{\mathcal{G}^{(s)} \geq 0,\ \mathcal{G}^C < 0\}}{Pr\{\mathcal{G}^C < 0\}} \tag{A6}$$

In order to evaluate the denominators in Eqs. A5 and A6, one must integrate the probability density function $p(\mathcal{G}^C = v)$. From Eqs. A1 and A3, this function can be expressed as

$$p(\mathcal{G}^C = v) = p(\mathcal{G}_\Delta = v - \mathcal{G}^T)$$

$$= \frac{1}{\sqrt{2\pi}\,\sigma_{\mathcal{G}_\Delta}} \exp\left\{ - \frac{\left[(v - \mathcal{G}^T) - \mu_{\mathcal{G}_\Delta}\right]^2}{2\sigma^2_{\mathcal{G}_\Delta}} \right\} \tag{A7}$$

In order to evaluate the numerators in Eqs. A5 and A6, we can first derive the integral form of conditional probability $Pr\{\mathcal{G}^{(s)} < u \mid \mathcal{G}^C = v\}$, using Eqs. A2 and A4. This form is then differentiated to obtain the corresponding probability density function. Finally, from the *multiplication rule* of conditional probability, an explicit formula of the joint probability function can be derived

$$p(\mathcal{G}^{(s)} = u,\ \mathcal{G}^C = v) = \frac{1}{2\pi\sigma_{\mathcal{G}_\Delta}\sigma_{\mathcal{G}^{(s)}_\Xi}}$$

$$\times \exp\left\{ - \frac{1}{2}\left[\frac{(u - v - \mu_{\mathcal{G}^{(s)}_\Xi})^2}{\sigma^2_{\mathcal{G}^{(s)}_\Xi}} + \frac{\left(v - \mathcal{G}^T - \mu_{\mathcal{G}_\Delta}\right)^2}{\sigma^2_{\mathcal{G}_\Delta}} \right] \right\} \tag{A8}$$

Therefore, if the means and variances of the charging and measurement errors can be estimated correctly, one can then determine a_s and b_s easily by integrating Eqs. A7 and A8 with a standard numerical technique.

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