Nonlinear Predictive Control of Periodically Forced Chemical Reactors

Fatih Özgülşen, Scott J. Kendra, and Ali Çinar
Chemical Engineering Dept., Illinois Institute of Technology, Chicago, IL 60616

A nonlinear model-predictive control strategy is developed to maintain the superior-to-steady-state performance of a periodically forced chemical reactor. The performance of the predictive controller is investigated in the presence of measurement disturbances and parametric uncertainty. It is also shown that statistically inferred input-output models can be a substitute whenever detailed fundamental models are not available. A nonlinear autoregressive polynomial model based on observed plant data is built and incorporated into the control scheme. The catalytic oxidation of ethylene in a periodically-forced, continuous stirred-tank reactor is considered as the test case.

Introduction

Numerous theoretical and experimental investigations have shown that forced periodic operation of chemical reactors can lead to increased conversion, enhanced yield, reduced parametric sensitivity, reduced hot-spot problems, and stabilized reactor operation in the unstable steady-state region (Bailey, 1977; Cutlip, 1979; Barshad and Gulari, 1985, 1986; Çinar et al., 1987a,b; Rigopoulos et al., 1988; Sterman and Ydstie, 1990a,b; Silveston, 1991; Özgülşen et al., 1992a,b). The control of periodically forced reactors, however, is an important and interesting issue which has not received the attention it deserves. The problem of maintaining improvements that result from forced periodic operation, in the unavoidable presence of model imperfections and disturbances, indeed makes the issue an important one. It is equally important to show that such reactors can be kept under control with the aid of proper strategies, since safety concerns have played a major role in limiting industrial applications of forced periodic operation. On the other hand, the task of controlling a reactor which is subject to intentionally large variations in one or more input variables is rather interesting from a control point of view.

By using the method of vibrational control, Çinar et al. (1987a) investigated the effects of multiple input cycling on the dynamic behavior of a continuous stirred-tank reactor (CSTR). The stabilization effect, due to the formation of a stable limit cycle instead of the unstable steady state of the system with fixed inputs, also enabled them to obtain substantially higher levels of conversion. It should be noted that forced periodic control is an open-loop strategy. This is a valuable approach when measurements are costly and difficult or involve long time delays. However, it can be ineffective in the face of large disturbances or process/model uncertainty. Later, Rigopoulos et al. (1988) used the same CSTR system with forced periodic, proportional-integral (PI) feedback, and nonlinear feedback (push-pull) control laws for stabilized operation. In the case of PI control, the controller settings are tuned such that high-frequency, small-amplitude sustained oscillations in reactor temperature are generated via feedback. As for the nonlinear feedback law, they use a relay with hysteresis action to repeatedly push and pull the process state around the unstable operating point. They conclude that the amplitude of reactor temperature swings can be smaller under forced periodic control with multiple oscillatory inputs than under PI or push-pull control with feed flow rate manipulation. One important advantage of PI and push-pull control is the security of having a closed-loop feedback. Sterman and Ydstie (1990a) used relay feedback controllers with integral and hysteresis action to create small-amplitude, self-sustained oscillations in a polymerization reactor. The desired frequency in the feedback is achieved by reducing the crossover frequency of the system through filters. They report a significant increase in the performance and emphasize that relay controllers are insensitive to model uncertainties and process disturbances. Recently, Sinha et al. (1990) developed an adaptive control scheme for controlling nonlinear systems that exhibit complex periodic behavior. They use the difference between the actual

Correspondence concerning this article should be addressed to A. Çinar.

radius and the radius of the limit cycle to be controlled for regulatory feedback.

The approach that we take here differs from the previous studies in several ways. First, we make no attempt to introduce self-sustained oscillations via feedback. A real-time computer can easily regulate variations in oscillatory inputs, especially if square wave forms are chosen to be implemented. Secondly, we do not focus our attention on the stabilization effect of periodic control; rather we consider the problem of maintaining improved reactor performance under periodic operation by reducing the effects of disturbances and parametric uncertainty. To accomplish this, we utilize a nonlinear model predictive control (NMPC) algorithm. Initially, we assume that a detailed (but not necessarily perfect) reactor model based on fundamental heat and mass balances is available. We then show that statistically inferred input-output models can be a substitute whenever detailed models are missing. In particular, we construct a nonlinear autoregressive model with exogenous inputs (NARX) by using an orthogonal forward-regression estimator and analyze the performance of the NARX-based NMPC algorithm.

Process Description

The system under investigation is a nonadiabatic CSTR in which ethylene oxide (C_2H_4O) is produced by catalytic oxidation of ethylene with air. The production goal is to maximize the C_2H_4O yield and to minimize the CO_2 yield. It is generally accepted that the kinetics can be described by the following triangular scheme:

$$\begin{array}{ccc} C_2H_4 & \xrightarrow{r_1} & C_2H_4O \\ & \searrow^{r_2} \quad \swarrow_{r_3} & \\ & CO_2 + H_2O & \end{array}$$

The rate expressions used in this study are by Alfani and Carberry (1970) and characterize an unmodified, commercial silver catalyst. The dimensionless reactor model consists of the following conservation equations:

$$\frac{dy_1}{dt} = q(1 - y_1 y_4)$$

$$\frac{dy_2}{dt} = q(c - y_2 y_4) - A_1 \exp\left(\frac{\gamma_1}{y_4}\right)(y_2 y_4)^{0.50} - A_2 \exp\left(\frac{\gamma_2}{y_4}\right)(y_2 y_4)^{0.25}$$

$$\frac{dy_3}{dt} = -q y_3 y_4 + A_1 \exp\left(\frac{\gamma_1}{y_4}\right)(y_2 y_4)^{0.50} - A_3 \exp\left(\frac{\gamma_3}{y_4}\right)(y_3 y_4)^{0.50}$$

$$\frac{dy_4}{dt} = \frac{q}{y_1}(1 - y_4) + \frac{B_1}{y_1} \exp\left(\frac{\gamma_1}{y_4}\right)(y_2 y_4)^{0.50} + \frac{B_2}{y_1} \exp\left(\frac{\gamma_2}{y_4}\right)(y_2 y_4)^{0.25} + \frac{B_3}{y_1} \exp\left(\frac{\gamma_3}{y_4}\right)(y_3 y_4)^{0.50} - \frac{B_4}{y_1}(y_4 - u) \quad (1)$$

where t is the time, y_1 is the density of gas mixture, y_2 is the ethylene concentration, y_3 is the ethylene oxide concentration, and y_4 is the reactor temperature. u, the coolant temperature, has a nominal value of 1.0 (= 523 K). q and c represent the volumetric feed flow rate and ethylene feed concentration, respectively. $\gamma_i (i = 1,2,3)$ denotes the activation energy. $A_i (i = 1,2,3)$ and $B_j (j = 1,2,3,4)$ represent dimensionless coefficients. The reader is referred to Özgülşen et al. (1992a) for a detailed description of the dynamic model.

Forced periodic operation of Eq. 1 is realized through periodic variations in feed flow rate and ethylene feed concentration. Hence, the vector of oscillatory inputs $p = [c,q]^T$. Although there are infinitely many forcing functions to choose from, nonsymmetric rectangular pulses are particularly attractive for such waves and are easy to generate in an experimental system.

$$q(t) = \begin{cases} q_{\min} & \kappa\tau \le t < (\kappa + \beta)\tau \\ q_{\max} & (\kappa + \beta)\tau \le t \le (\kappa + \alpha + \beta)\tau \\ q_{\min} & (\kappa + \alpha + \beta)\tau < t \le (\kappa + 1)\tau \end{cases} \quad (2)$$

$$c(t) = \begin{cases} c_{\min} & \kappa\tau \le t < (\kappa + \beta_c)\tau \\ c_{\max} & (\kappa + \beta_c)\tau \le t \le (\kappa + \alpha + \beta_c)\tau \\ c_{\min} & (\kappa + \alpha + \beta_c)\tau < t \le (\kappa + 1)\tau \end{cases} \quad (3)$$

Here, $\kappa = 0,1,2,..,\alpha$ is the duty fraction, τ is the dimensionless period, and β and β_c are the fractions of period at which switchings from minimum to maximum values occur. Note that both waves have the same duty fraction and frequency. Figure 1 illustrates the variation of input variables with time.

Given the values of time-average feed flow rate (q_{av}) and ethylene feed concentration (c_{av}), and with fixed $R_q (= q_{\max}/q_{\min})$ and $R_c (= c_{\max}/c_{\min})$, specifying three parameters: α, σ and τ is sufficient to describe the pulses. Here, the phase shift is defined as:

$$\sigma = \beta - \beta_c. \quad (4)$$

In an earlier study, Özgülşen et al. (1992a) computed the optimal values of these parameters to maximize the time-average yield of ethylene oxide. They reported as high as 24% yield improvement relative to steady-state operation. According to their results, the optimal steady-state operation corresponds to $q = 0.06$ and $c = 1$ vol. %, resulting in an ethylene oxide yield of 11.495%. However, forced periodic operation of the same system around the optimum steady state with:

$$\begin{aligned} q_{av} &= 0.06 \\ c_{av} &= 1.00 \\ R_c &= 10.00 \\ R_q &= 10.00 \end{aligned}$$

can provide a time-average yield of 12.64%, when the optimal waves described by $\alpha = 0.4057$, $\sigma = -0.3561$ and $\tau = 30.6124$ (≈ 10 min/cycle) are implemented. This translates into an improvement of 10% over the optimal steady-state (unforced) yield. All of these results are based on calculations done by using the reactor model described by Eqs. 1.

To introduce parametric uncertainty into the problem, the dimensionless heat-transfer coefficient (B_4) of the true plant is assumed to have a value of 0.702, while the model B_4 is

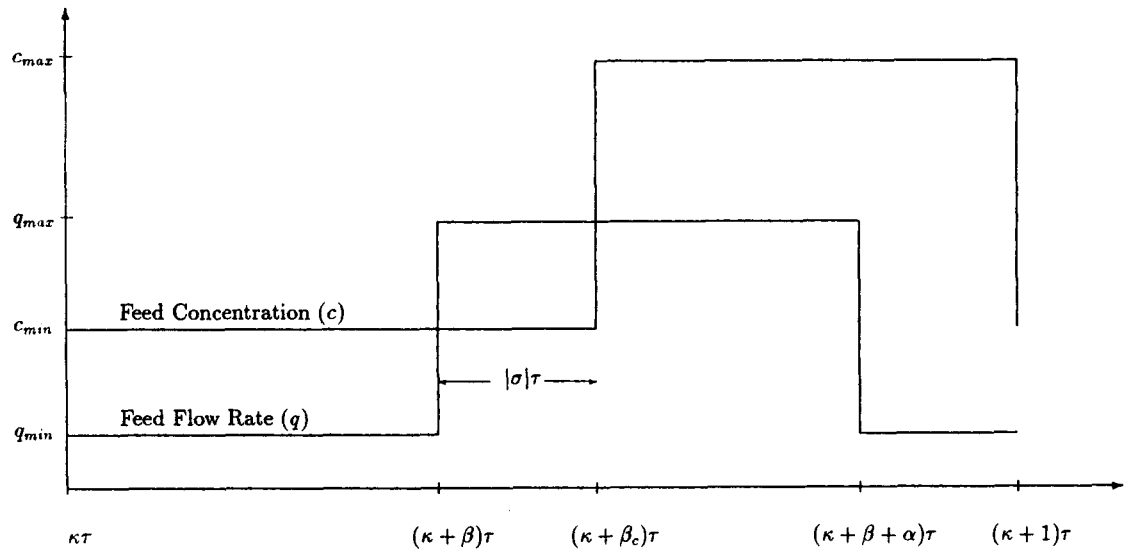

Figure 1. Variation of oscillatory inputs with time.

7.02. Because of this mismatch in B_4, forced periodic operation of the plant (perturbed model) with the optimal forcing parameters results in a 2% deterioration compared to its steady-state yield (10.206%, lower dotted line in Figure 2C). The performances of the model and the plant (perturbed model) under two different modes of operation are listed in Table 1.

In the control studies to follow, setpoint refers to the trajectories of the nominal system and measurement to the perturbed system output. Figure 2, which shows the open-loop behavior of the plant, clearly shows the need for a controller. The temperature of the perturbed (Figure 2A, solid line) has an average that is about 18 K higher than the average of the

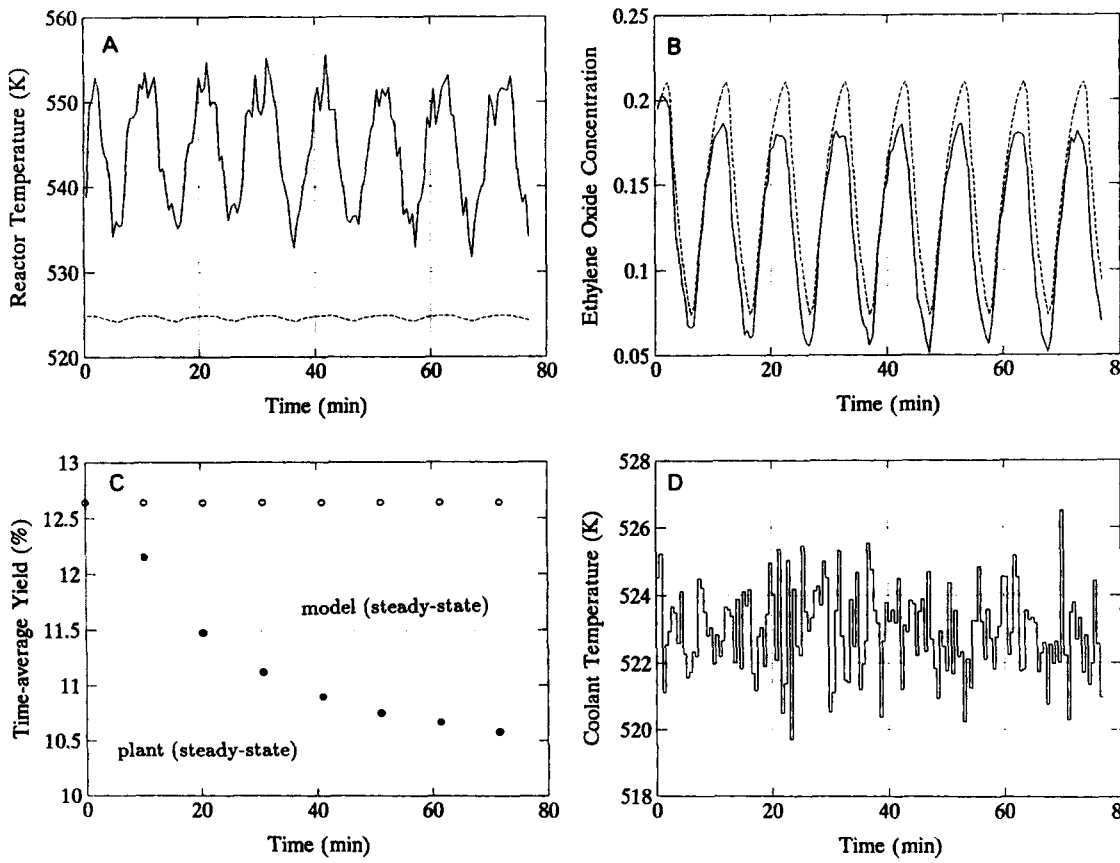

Figure 2. Open-loop simulation of the system with added random disturbances.

—, observed trajectory; ----, desired trajectory; •, observed value of time-average yield, ○, desired value of time-average yield.

Table 1. Time-Average Yield of Ethylene Oxide ($u=1.0$, No Noise)

	Steady-State Operation	Forced Periodic Operation	Improvement
plant ($B_4=0.702$)	10.206%	10.036%	−1.664%
model ($B_4=7.02$)	11.495%	12.640%	9.968%

nominal system (Figure 2A, dashed line) and a lower ethylene concentration (Figure 2B). The higher temperature, which results from the lower heat-transfer coefficient, increases the rate of oxidation of ethylene oxide to CO_2 and H_2O. If the state trajectories of the perturbed system can be made to follow the state trajectories of the nominal system through feedback control, it will be possible to maintain the superior time-averaged yield of ethylene oxide due to forced periodic operation.

Nonlinear Model-Predictive Control

In recent years, several nonlinear control system design methodologies have been developed and successfully implemented in chemical processes. These techniques were reviewed extensively by Bequette (1991). One of these strategies, nonlinear model-predictive control, seems suitable particularly for the task of controlling periodically-forced chemical reactors. The NMPC scheme does not require a convolution model. It can easily handle constraints imposed on the manipulated variable. Due to the moving predictive horizon approach, the NMPC scheme is also capable of handling future setpoint changes. In the case of periodically-forced systems, the period of oscillations is a good choice for the extent of prediction horizon.

The NMPC problem can be formally stated as:

$$\min_{u(k),\ldots,u(k+L-1)}\left\{\sum_{i=k+1}^{k+M}\sum_{j=1}^{n}\mu(i,j)[y_{sp}(i,j)-y_{\text{pred}}(i,j)]^2\right\} \tag{5}$$

subject to:

i. *Constraints in continuous time*

$$\frac{d}{dt}y-f(y,u,p,w)=0 \tag{6}$$

$$p(t)-p(t+\tau)=0 \tag{7}$$

$$y(t_0)=y_0 \tag{8}$$

ii. *Constraints in discrete time*

$$y_{sp}(k,j)=y_{sp}(k+M,j),\quad j=1,\ldots,n \tag{9}$$

$$u_{\min}\le u(i)\le u_{\max},\quad i=k,\ldots,(k+L-1) \tag{10}$$

$$u(i+1)=u(i),\quad i=(k+L-1),\ldots,(k+M-2)\quad \text{for } M>L \tag{11}$$

$$|u(i+1)-u(i)|\le\Delta u_{\max},\quad i=k,\ldots,(k+L-2) \tag{12}$$

In the above, u is the manipulated variable (here, coolant temperature), y_{sp} is the setpoint matrix, y_{pred} is the predicted output matrix, and d is the disturbance vector. Recall that p is the vector of oscillatory inputs and τ is the period of oscillations. n, m, L, and k represent the dimension of state vector, the prediction horizon, the control horizon, and the current time step, respectively. Finally, μ is the $M\times n$ dimensional weighting matrix.

Equation 5 shows that the control objective is to select the series of future control signals which minimize the sum of the squares of the residuals over the prediction horizon of M time steps. Here, all of the state variables are assumed to be measurable. Equation 6 represents the modeling ODEs. Note that $u\notin p$. Initial conditions for the state variables at the beginning of the prediction horizon are given in Eq. 8. The solution of the model equations from the previous time step are used as initial conditions at the current time step. Equation 9 indicates that all setpoint values are τ-periodic and the span of prediction horizon is equal to one cycling period. Equations 10 and 12 represent the absolute and velocity constraints on u, respectively. Equation 11 shows that the control action is kept constant at $u(k+L-1)$ from the end of the control horizon to the end of the prediction horizon. The disturbance vector is updated by using:

$$d(j)|_k=y_{\text{plant}}(j)|_k-y(j)|_k,\quad j=1,\ldots,n \tag{13}$$

where $y_{\text{plant}}|_k$ is the plant output and $y|_k$ is the model output at the current time step. By definition, d contains the measured/unmeasured disturbances as well as the modeling errors. The vector d is used to incorporate feedback into the scheme:

$$y_{\text{pred}}(k+i,.)=y(.)|_{k+i}+d(.)|_k,\quad i=1,\ldots,M \tag{14}$$

During the prediction process, future disturbances $(d|_{k+1},\ldots d|_{k+M})$ are assumed to remain constant at the current level $d|_k$. After solving Eq. 5 for $u(k),\ldots,u(k+L-1)$, only the first control action is implemented. Then, the output measurements are read at the $(k+1)$th time step and the prediction procedure is repeated. In general, either the *sequential* or the *simultaneous* approach is used for computing the future values of the manipulated variable (Patwardhan et al., 1990; Biegler and Rawlings, 1991; Sistu and Bequette, 1991). Due to the presence of oscillatory inputs, we preferred to use the sequential approach which does not require the removal of derivative terms in modeling equations by discretization. The dynamic model equations (Eq. 6) are integrated by using ODESSA (Leis and Kramer, 1988) and the nonlinear programming problem is solved by using OPTIMIZATION TOOLBOX (Grace, 1990).

Figure 3 is a block diagram representation of the problem. Note that w is an additive noise corrupting the control signal and v is a vector of n measurement errors. Here, it is assumed that $w\sim N(0,\sigma_w^2)$ and $v\sim N(\mathbf{0},\sigma_v^2)$. Figure 4 illustrates the closed-loop performance of the reactor system. The tuning parameters are set to $M=20$, $L=3$, and $\Delta u_{\max}=0.05$ (≈ 26 K). The upper ($u_{\max}$) and lower ($u_{\min}$) bounds imposed on the control signal are 1.1 (≈ 575 K) and 0.6 (≈ 314 K), respectively. Deviations from the setpoint are larger for y_1 and y_4; hence, the weights on these state variables are chosen to be bigger than the ones on y_2 and y_3. It takes approximately three cycling periods for the time-average yield to reach its desired value as

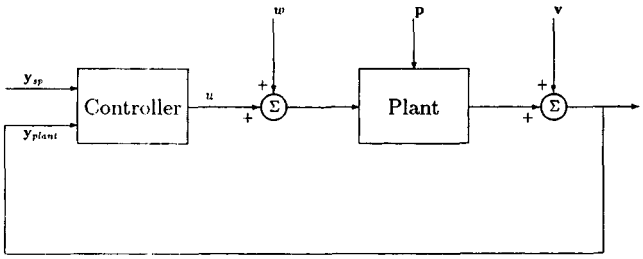

Figure 3. Block diagram of the problem.

the transient response due to model/plant mismatch dies out. Figure 5 shows the dynamic response of the system to a combination of mismatch, unmeasured disturbances and a step increase in q_{av}. A step change of +5% is introduced at the beginning of the fourth cycle period. Figures 4 and 5 indicate that the NMPC scheme can maintain the reactor performance at the optimal level despite all of these disturbances and uncertainty.

Nonlinear Identification

Having shown that the NMPC algorithm can successfully be implemented in periodically forced processes, we turn now to look at the *black box* identification problem. Specifically, we construct the NARX model of the forced CSTR which has the following general structure:

$$\hat{y}_j(k) = \hat{f}_j[\hat{y}_1(k-1),\ldots,\hat{y}_1(k-m_y),\ldots,\hat{y}_n(k-1),\ldots,\hat{y}_n(k-m_y),$$
$$u(k-1),\ldots,u(k-m_u),q(k-1),\ldots,q(k-m_q),$$
$$c(k-1),\ldots,c(k-m_c)] + \xi_j(k), \quad j=1,\ldots,n \qquad (15)$$

where $\hat{y}_j(k)$ denotes the estimate of y_j at the kth time step. $\hat{f}_j(.)$ is a nonlinear function which describes the jth subsystem and contains the previous input and output values. $\xi(.)$ is the prediction error, and m_y, m_u, m_q, and m_c represent the maximum lags in $\hat{y}$, u, q, and c, respectively. Obviously, different lag values may be used for each input/output in each subsystem. We disregard this situation in our equations for simplicity in notation. Generally, $\hat{f}$ is approximated by polynomial models:

$$\hat{f}_j(k) = \theta^j{}_0 + \theta^j{}_1\varphi(k,1) + \ldots + \theta^j{}_m\varphi(k,m)$$
$$+ \theta^j{}_{m+1}\varphi(k,1)\varphi(k,1) + \ldots$$
$$+ \theta^j{}_{\left[m+\frac{(m+1)!}{2!(m-1)!}\right]}\varphi(k,m)\varphi(k,m) + \text{h.o.t.} \qquad (16)$$

where

$$m_{ny} = n \times m_y$$
$$m = m_{ny} + m_u + m_q + m_c$$

and

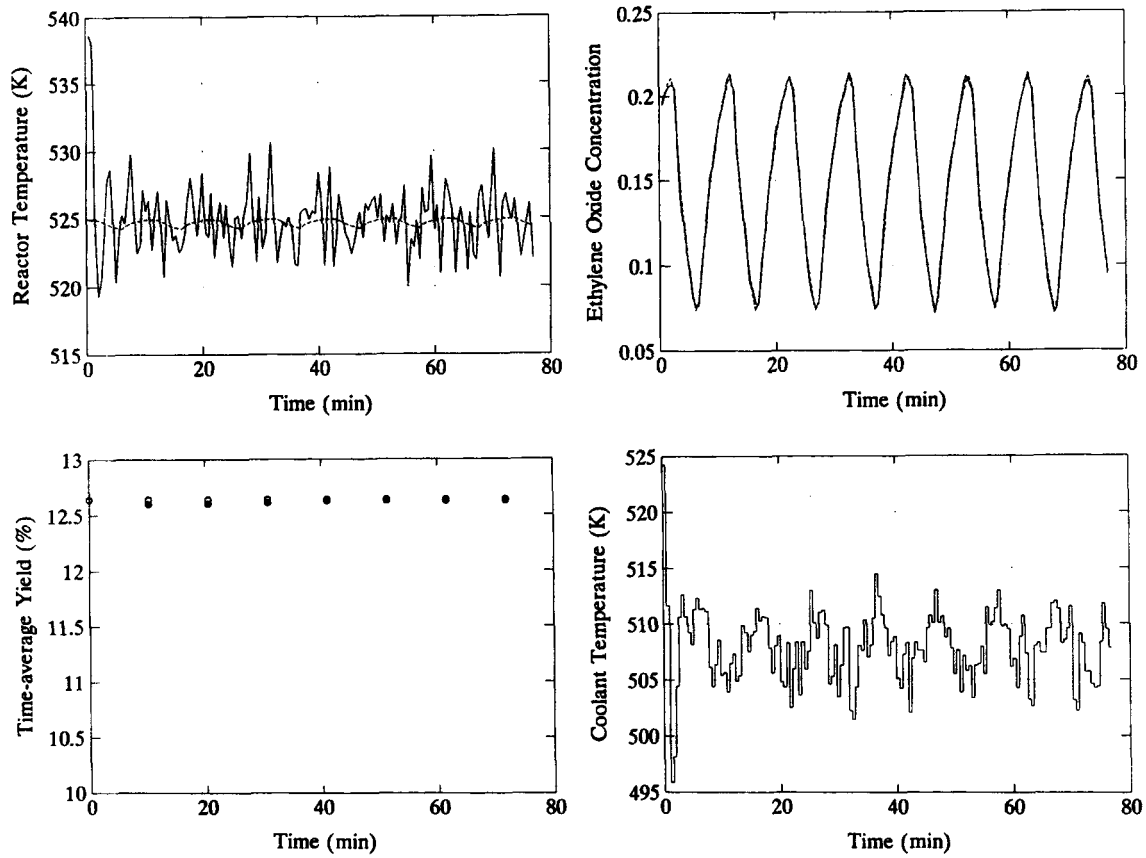

Figure 4. Closed-loop simulation of the system with added random disturbances and model/plant mismatch.

—, measurement; ----, setpoint; •, observed value of time-average yield; o, desired value of time-average yield.

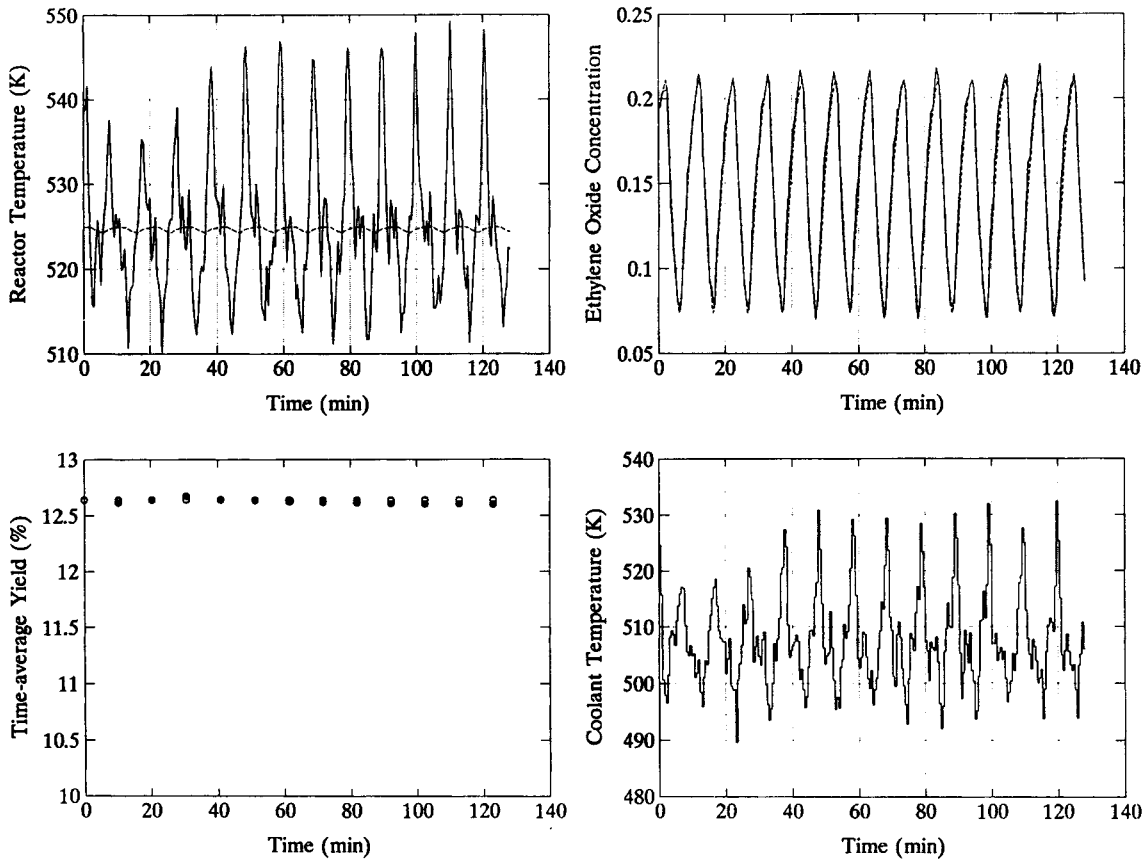

Figure 5. Closed-loop simulation of the system with added random disturbances, model/plant mismatch and a step increase in q_{av}.

—, measurement; ----, setpoint, •, observed value of time-average yield; o, desired value of time-average yield.

$$\varphi(k,1)=z_1(k-1),\ \ldots,\ \varphi(k,m_y)=z_1(k-m_y),$$

$$\vdots \qquad \cdots \qquad \vdots$$

$$\varphi[k,(n-1)\times m_y+1]=z_n(k-1),\ \ldots,\ \varphi(k,m_{ny})=z_n(k-m_y),$$

$$\varphi(k,m_{ny}+1)=u(k-1),\ \ldots,$$

$$\varphi(k,m_{ny}+m_u)=u(k-m_u),$$

$$\varphi(k,m_{ny}+m_u+1)=q(k-1),\ \ldots,$$

$$\varphi(k,m_{ny}+m_u+m_q)=q(k-m_q),$$

$$\varphi(k,m_{ny}+m_u+m_q+1)=c(k-1),\ \ldots,\varphi(k,m)=c(k-m_c).$$

Assuming that $N+m_y$, $N+m_u$, $N+m_q$, and $N+m_c$ data points collected from the plant with identical sampling rates are available, one can determine the unknown coefficients of such a polynomial model. Before we proceed any further, an important remark is in order. Note that y is replaced with z in the above equation. $z_j(k-1)$ denotes the observed value for the jth output at the $(k-1)$th time step during data collection from the open-loop system. Since the open-loop system is stable, input-output data can be collected from the reactor off-line. Because of the constant term θ_0, the first column of the regression matrix is set to **1**. The rest of the columns include all possible combinations of φ up to degree ℓ. Thus,

$$\Phi=[\mathbf{1}\ \Phi_2\ \ldots\ \Phi_{K+1}] \tag{17}$$

where K is the total number of unordered arrangements with repetition:

$$K=C\binom{m}{1}+C\binom{m+1}{2}+\ldots+C\binom{m+\ell-1}{\ell}.$$

1st-order terms 2nd-order terms ℓth-order terms

The number of rows of Φ is equal to the data length (N). Having assembled the regression matrix, the identification problem for each subsystem can now be written in matrix notation as:

$$z_j=\Phi\Theta_j+\Xi_j \tag{18}$$

where Ξ_j is the $N\times 1$ vector of modeling errors and $\Theta_j=[\theta^j_0\ \theta^j_1\ \ldots\ \theta^j_K]^T$ is the vector of unknown coefficients of the jth subsystem. Note that the problem is *linear in the parameters*. The size of Φ can be extremely large, especially if the process has several inputs and outputs to consider and little is known about the structure of the system. For the problem under consideration, with $m_y=m_u=m_q=m_c=9$ and $\ell=3$, Φ consists of 45,760 column vectors. The size of the regression matrix is only one of the reasons for not solving:

$$\Phi^T\Phi\Theta_j=\Phi^T z_j \tag{19}$$

for Θ_j directly by using Gaussian elimination or Cholesky decomposition. It is also very likely that the regression matrix is ill-conditioned (Chen et al., 1989). Hence, these methods could yield erroneous and/or extremely complex I/O descriptions. A simple and efficient method for computing Θ_j is the classical Gram-Schmidt (CGS) algorithm. This method attacks Φ one column at a time; therefore, the entire regression matrix does not need to be stored in computer memory. The CGS algorithm has been discussed extensively by Billings and coworkers (Korenberg et al., 1988; Billings et al., 1989; Chen et al., 1989).

Consider the following factorization of the regression matrix:

$$\Phi = WA \tag{20}$$

where A is an upper triangular matrix with $a_{ii}=1.0$ for $i=1,\ldots,(K+1)$, and $W=[w_1 \ \ldots \ w_{K+1}]$ is the $N\times(K+1)$ orthogonal matrix of auxiliary regressors. W satisfies $W^TW=D$, where D is a diagonal matrix. To select the first term in the polynomial model for subsystem j, we compute:

$$\begin{aligned} w_1^{(i)} &= \Phi_i \\ g_1^{(i)} &= \frac{\langle w_1^{(i)}, z_j\rangle}{\langle w_1^{(i)}, w_1^{(i)}\rangle} \qquad i=1,\ldots,(K+1) \\ e_1^{(i)} &= \frac{(g_1^{(i)})^2\langle w_1^{(i)}, w_1^{(i)}\rangle}{\langle z_j, z_j\rangle} \end{aligned} \tag{21}$$

where $\langle .,. \rangle$ denotes the inner (dot) product, g is known as the auxiliary parameter, and e is the error reduction ratio. Suppose $e_1^{(l)}$ has the largest error reduction ratio among $\{e_1^{(i)}, 1\le i\le (K+1)\}$, then we set:

$$\begin{aligned} \hat{w}_1 = w_1^{(l)} \quad & \hat{g}_1 = g_1^{(l)} \\ = \Phi_l \quad & \hat{e}_1 = e_1^{(l)} \\ \hat{a}_{11} = 1.0. & \end{aligned}$$

The second term of the polynomial model is selected by using the same procedure. But this time, since Φ_l has already been chosen, it is no more considered as a candidate:

$$\begin{aligned} a_{12}^{(i)} &= \frac{\langle \hat{w}_1, \Phi_i\rangle}{\langle \hat{w}_1, \hat{w}_1\rangle} \\ w_2^{(i)} &= \Phi_i - a_{12}^{(i)}\hat{w}_1 \\ & \qquad i=1,\ldots,(K+1),\ i\neq l \\ g_2^{(i)} &= \frac{\langle w_2^{(i)}, z_j\rangle}{\langle w_2^{(i)}, w_2^{(i)}\rangle} \\ e_2^{(i)} &= \frac{(g_2^{(i)})^2\langle w_2^{(i)}, w_2^{(i)}\rangle}{\langle z_j, z_j\rangle} \end{aligned} \tag{22}$$

At this stage, the monomial with the largest e_2 is selected as the second term of the jth subsystem. The procedure is repeated until the Akaike information criterion (AIC) is minimized upon selection of the n_{sj}th term:

$$\min(\mathrm{AIC}_j) = N\ln(\sigma^2_{sj}) + 4n_{sj}. \tag{23}$$

In the above, σ^2_{sj} denotes the variance of the residuals of subsystem j after the inclusion of the n_{sj}th term into the polynomial model. Bayesian information criterion and F-tests are some of the other statistical criteria that can be used to terminate the selection procedure. Coefficients of the chosen monomials ($\hat{\Theta}_j$) are computed by solving:

$$\begin{bmatrix} 1 & \hat{a}_{12} & \hat{a}_{13} & \ldots & \hat{a}_{1n_{sj}} \\ 0 & 1 & \hat{a}_{23} & \ldots & \hat{a}_{2n_{sj}} \\ 0 & 0 & 1 & \ldots & \hat{a}_{3n_{sj}} \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & 0 & \ldots & 1 \end{bmatrix} \begin{bmatrix} \hat{\theta}^j_1 \\ \vdots \\ \hat{\theta}^j_{n_{sj}} \end{bmatrix} = \begin{bmatrix} \hat{g}_1 \\ \vdots \\ \hat{g}_{n_{sj}} \end{bmatrix}. \tag{24}$$

Using the CGS algorithm, the following NARX model based on observed plant data is constructed:

$$\begin{aligned} \hat{y}_1(k) = {} & 1.0306\hat{y}_1(k-1) - 0.0978\hat{y}_3(k-9)\hat{y}_3(k-2)\hat{y}_2(k-1) \\ & + 0.1363c(k-4)q(k-3)\hat{y}_3(k-8) \\ & - 0.0007c(k-6)c(k-6)q(k-1) \\ & + 0.4177q(k-1)\hat{y}_3(k-7)\hat{y}_3(k-7) \\ & - 0.0454q(k-1)\hat{y}_4(k-3)\hat{y}_2(k-8) \\ & + 0.9620q(k-1)\hat{y}_4(k-5)\hat{y}_2(k-3) \\ & - 0.9150q(k-1)\hat{y}_4(k-1)\hat{y}_2(k-3) \\ & - 0.0541q(k-3)\hat{y}_3(k-7)\hat{y}_2(k-3) \\ & + 0.0007c(k-1)\hat{y}_3(k-8)\hat{y}_2(k-5) \\ & - 0.0126u(k-2)u(k-1)\hat{y}_1(k-1) \\ & + 0.0025c(k-3)q(k-3)\hat{y}_2(k-4) \\ & - 0.0289u(k-5)\hat{y}_1(k-3)\hat{y}_1(k-2) \\ & - 0.0034c(k-3)q(k-8)\hat{y}_3(k-6) \\ & + 0.0095u(k-5)u(k-4)u(k-2) \\ & + 0.0055q(k-6)\hat{y}_2(k-6)\hat{y}_2(k-6) + \xi_1(k) \end{aligned} \tag{25}$$

$$\begin{aligned} \hat{y}_2(k) = {} & 1.5936\hat{y}_4(k-9)\hat{y}_4(k-9)\hat{y}_2(k-1) \\ & + 250.1008q(k-8)q(k-7)q(k-1) \\ & + 0.2492c(k-3)q(k-8)\hat{y}_2(k-7) \\ & + 7.3842q(k-7)q(k-2)\hat{y}_2(k-3) \\ & - 9.5646q(k-8)\hat{y}_3(k-5)\hat{y}_2(k-9) \\ & + 0.2676q(k-3)q(k-2)u(k-8) \\ & + 0.0532c(k-2)\hat{y}_3(k-9)\hat{y}_2(k-5) \\ & + 1.8162q(k-4)q(k-3)\hat{y}_2(k-3) \\ & + 0.0806c(k-4)\hat{y}_3(k-9)\hat{y}_3(k-7) \\ & - 0.6244c(k-6)q(k-4)\hat{y}_3(k-9) \\ & - 0.0295c(k-3)\hat{y}_3(k-9)\hat{y}_3(k-1) \\ & - 0.0971\hat{y}_4(k-8)\hat{y}_2(k-1)\hat{y}_1(k-1) \\ & - 0.3015u(k-9)\hat{y}_4(k-1)\hat{y}_2(k-1) \end{aligned}$$

$$
\begin{aligned}
&-3.0296\hat{y}_4(k-9)\hat{y}_2(k-1)\hat{y}_1(k-7)\\
&+0.2676c(k-6)q(k-5)\hat{y}_3(k-4)\\
&+0.1991u(k-7)\hat{y}_3(k-9)\hat{y}_2(k-1)\\
&+2.9347\hat{y}_2(k-1)\hat{y}_1(k-8)\\
&+0.3044\hat{y}_3(k-9)\hat{y}_3(k-1)\hat{y}_2(k-3)\\
&-0.2325u(k-1)\hat{y}_4(k-2)\hat{y}_2(k-1)\\
&+0.0205c(k-5)q(k-1)\hat{y}_4(k-6)+\xi_2(k) \qquad (26)
\end{aligned}
$$

$$
\begin{aligned}
\hat{y}_3(k) = &-1.2388\hat{y}_3(k-1)\hat{y}_1(k-1)\hat{y}_1(k-1)\\
&+0.8733c(k-5)q(k-4)\hat{y}_2(k-1)\\
&-1.5827q(k-4)\hat{y}_3(k-6)\hat{y}_3(k-1)\\
&+0.1851c(k-1)q(k-4)q(k-1)\\
&+0.0125c(k-4)\hat{y}_4(k-5)\hat{y}_3(k-6)\\
&-0.6332c(k-6)q(k-4)q(k-1)\\
&+0.0876q(k-3)\hat{y}_2(k-5)\hat{y}_2(k-2)\\
&-0.1800c(k-9)q(k-5)\hat{y}_3(k-4)\\
&-0.3568q(k-4)\hat{y}_3(k-3)\hat{y}_2(k-7)\\
&+0.3161q(k-8)q(k-7)q(k-2)\\
&-0.0354\hat{y}_3(k-9)\hat{y}_2(k-8)\hat{y}_2(k-7)\\
&+2.1145\hat{y}_3(k-1)\hat{y}_1(k-1)\\
&+0.0484c(k-6)q(k-3)\hat{y}_3(k-8)\\
&-0.00003c(k-5)c(k-2)\hat{y}_2(k-4)\\
&-0.0969\hat{y}_4(k-9)\hat{y}_4(k-1)\hat{y}_3(k-1)\\
&-0.0203c(k-9)q(k-9)q(k-2)\\
&+0.0656u(k-7)u(k-4)\hat{y}_3(k-1)+\xi_3(k) \qquad (27)
\end{aligned}
$$

$$
\begin{aligned}
\hat{y}_4(k) = &1.6516\hat{y}_4(k-1)\hat{y}_1(k-1)\\
&-0.0136c(k-6)q(k-1)\hat{y}_2(k-9)\\
&+0.0231c(k-2)q(k-2)\hat{y}_2(k-2)\\
&-0.0398c(k-7)\hat{y}_3(k-9)\hat{y}_3(k-9)\\
&-0.4141u(k-1)\hat{y}_4(k-1)\hat{y}_1(k-1)\\
&+0.1317c(k-9)q(k-8)\hat{y}_3(k-8)\\
&-0.2057u(k-4)\hat{y}_4(k-1)\hat{y}_1(k-1)+\xi_4(k). \qquad (28)
\end{aligned}
$$

The response of this model to variations in q and c (periodic), and u (gaussian sequence) is plotted in Figure 6. It is clear that the polynomial NARX model closely approximates the dynamics of the true plant. The predictive controller performs satisfactorily with the NARX model incorporated into the control scheme. Since polynomial models can exhibit explosive

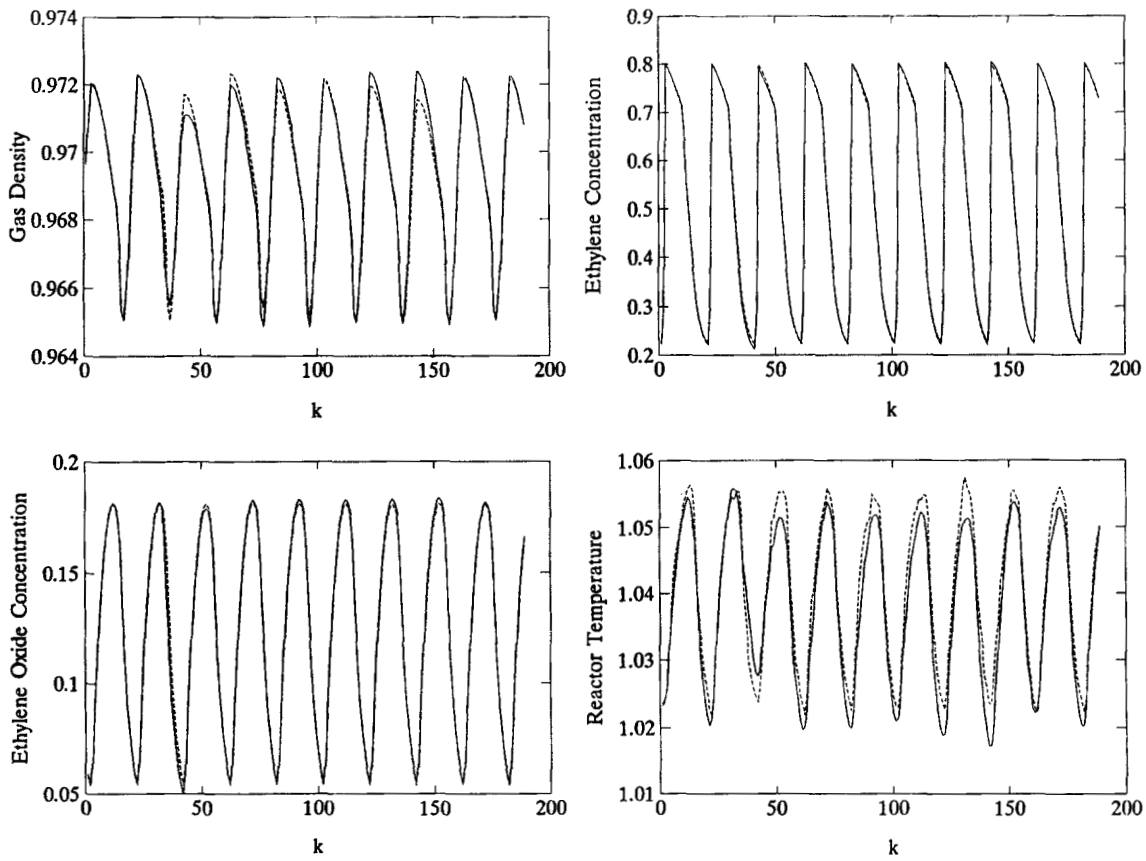

Figure 6. Response of the NARX model to variations in u, q, and c.

u is a sequence of normal distribution, and q and c are periodic. —, estimated output; ----, true output.

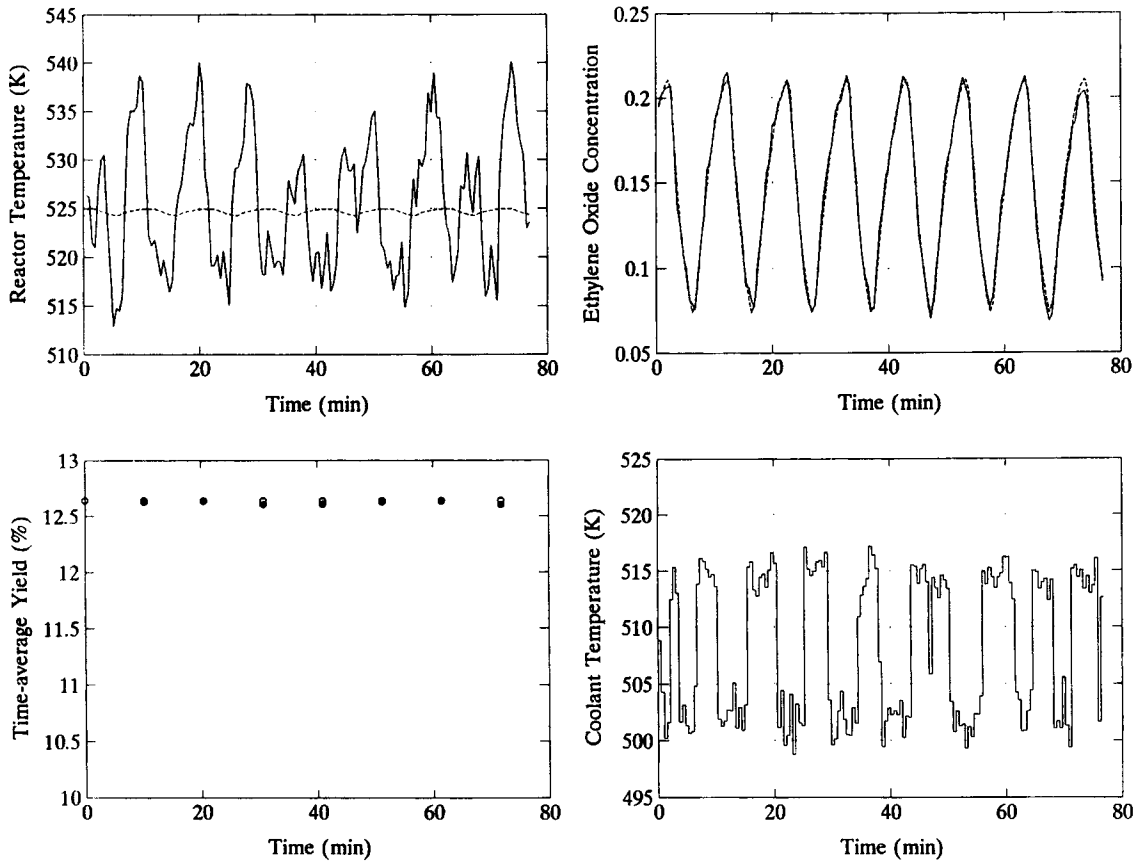

Figure 7. Closed-loop performance of the NARX-based controller with added random disturbances.

—, measurement; ----, setpoint; •, observed value of time-average yield; o, desired value of time-average yield.

behavior, a shorter prediction horizon ($M=3$) is used in this case. The dynamic simulation of the NARX-based controller with w and v acting on the system is displayed in Figure 7.

Conclusion

In this work we have demonstrated the effectiveness of the NMPC strategy for controlling periodically-forced processes. Our results for the CSTR with the ethylene oxidation reactions show that the improvement in reactor performance which results from forced periodic operation can be maintained despite the presence of parametric uncertainty and unmeasured disturbances. The effect of deadtime on controller performance is not considered in this article. However, we wish to stress the fact that nonminimum phase systems can easily be handled in the framework of NMPC. Since operation at an unstable limit cycle can sometimes be beneficial, the problem of controlling open-loop unstable forced systems should also be investigated. There is enough evidence in the literature (although none involve periodically-forced systems) to believe that the NMPC algorithm in conjunction with a state and parameter estimator can perform satisfactorily in the case of open-loop unstable forced systems subject to uncertainties and disturbances (Sistu and Bequette, 1991).

It has been shown in this study that the off-line identification methods applied to periodically-forced systems can produce reliable nonlinear input-output models. The NARX model built for the test case captures the essential dynamics of the process and generates a response that is very close to that of the original system. When this polynomial model is incorporated into the control scheme, no significant degradation of performance is noted.

Acknowledgment

Financial support by the Department of Energy under Grant DE-FG02-87ER13696 and by the donors of the Petroleum Research Fund, administered by the American Chemical Society, is gratefully acknowledged. IIT Research Institute Fellowship to S. J. Kendra is also gratefully acknowledged.

Notation

A = upper triangular matrix defined in Eq. 20
A_1, A_2, A_3 = dimensionless terms in mass balance equations
B_1, B_2, B_3, B_4 = dimensionless terms in heat balance equation
c = dimensionless ethylene feed concentration
d = vector of disturbances
e = error reduction ratio
g = auxiliary parameter
k = current time step
K = total number of monomials
L = control horizon
m_c, m_q, m_u, m_y = maximum lags in c, q, u, and $\hat{y}$
M = prediction horizon
n = number of outputs
N = data length in off-line identification
p = vector of oscillatory inputs
q = dimensionless volumetric feed flow rate
R_c = maximum to minimum ethylene feed concentration ratio

R_q = maximum to minimum feed flow rate ratio
t = dimensionless time
u = coolant temperature, manipulated variable
v = vector of measurement errors
w = noise corrupting the control signal
W = matrix of auxiliary regressors
y_{plant} = measured output value
y_{pred} = predicted output defined by Eq. 14
y_{sp} = setpoint value
y = output vector (detailed model)
$\hat{y}$ = output vector (NARX model)

Greek letters

α = duty fraction of pulse
β, β_c = fractions of time at which switchings occur
γ_1, γ_2, γ_3 = dimensionless activation energies
$\boldsymbol{\theta}$ = vector of unknown coefficients
μ = weighting matrix
$\boldsymbol{\Xi}$ = vector of modeling errors
σ = phase shift
σ_{sj}^2, σ_v^2, σ_w^2 = variances
τ = dimensionless cycle period
$\boldsymbol{\Phi}$ = regression matrix

Subscripts

av = time-average value
max = maximum value
min = minimum value

Superscripts

T = transpose
$\hat{}$ = estimated value

Abbreviations

AIC = Akaike information criterion
CGS = classical Gram-Schmidt
CSTR = continuous stirred tank reactor
h.o.t. = higher-order terms
NARX = nonlinear autoregressive model with exogeneous inputs
NMPC = nonlinear model-predictive control
ODE = ordinary differential equation

Literature Cited

Alfani, F., and J. J. Carberry, "An Exploratory Kinetic Study of Ethylene Oxidation over an Unmoderated Supported Silver Catalyst," *La Chimica e L'Industria*, **52**, 1192 (1970).

Bailey, J. E., "Periodic Phenomena," *Chemical Reactor Theory: A Review*, p. 758, L. Lapidus and N. R. Amundson, eds., Prentice-Hall, Englewood Cliffs, NJ (1977).

Barshad, Y., and E. Gulari, "A Dynamic Study of CO Oxidation on Supported Platinum," *AIChE J.*, **31**, 649 (1985).

Barshad, Y., and E. Gulari, "Modification of Product Distribution through Periodic Operation: Fischer-Tropsch Synthesis over Ru/Al_2O_3," *Chem. Eng. Comm.*, **43**, 39 (1986).

Bequette, B. W., "Nonlinear Control of Chemical Processes: A Review," *Ind. Eng. Chem. Res.*, **30**, 1391 (1991).

Biegler, L. T., and J. B. Rawlings, "Optimization Approaches to Nonlinear Model Predictive Control," *Chemical Process Control—CPC IV*, Y. Arkun and W. H. Ray, eds., CACHE/AIChE, New York (1991).

Billings, S. A., S. A. Chen, and M. J. Korenberg, "Identification of MIMO Non-linear Systems Using a Forward-regression Orthogonal Estimator," *Int. J. Control*, **49**, 2157 (1989).

Chen, S., S. A. Billings, and W. Luo, "Orthogonal Least Squares Methods and Their Application to Non-linear System Identification," *Int. J. Control*, **50**, 1873 (1989).

Çinar, A., J. Deng, S. M. Meerkov, and X. Shu, "Vibrational Control of an Exothermic Reaction in a CSTR: Theory and Experiments," *AIChE J.*, **33**, 353 (1987a).

Çinar, A., K. Rigopoulos, X. Shu, and S. M. Meerkov, "Vibrational Control of Chemical Reactors: Stabilization and Conversion Improvement in an Exothermic CSTR," *Chem. Eng. Comm.*, **59**, 299 (1987b).

Cutlip, M. B., "Concentration Forcing of Catalytic Surface Rate Processes," *AIChE J.*, **25**, 502 (1979).

Grace, A., *OPTIMIZATION TOOLBOX User's Guide*, The MathWorks, Inc., Natick, MA (1990).

Korenberg, M., S. A. Billings, Y. P. Liu, and P. J. McIlroy, "Orthogonal Parameter Estimation Algorithm for Non-linear Stochastic Systems," *Int. J. Control.*, **48**, 193 (1988).

Leis, J. R., and M. A. Kramer, "The Simultaneous Solution and Sensitivity Analysis of Systems Described by Ordinary Differential Equations," *ACM Trans. Math. Software*, **14**, 45 (1988).

Özgülşen, F., R. A. Adomaitis, and A. Çinar, "A Numerical Method for Determining Optimal Parameter Values in Forced Periodic Operation," *Chem. Eng. Sci.*, **47**, 605 (1992a).

Özgülşen, F., K. R. Rigopoulos, and A. Çinar, "A Comparative Study of Tools for Assessing the Effects of Forced Periodic Operation of Catalytic Reactors," *Chem. Eng. Comm.*, **112**, 85 (1992b).

Patwardhan, A. A., J. B. Rawlings, and T. F. Edgar, "Nonlinear Model Predictive Control," *Chem. Eng. Comm.*, **87**, 123 (1990).

Rigopoulos, K., X. Shu, and A. Çinar, "Forced Periodic Control of an Exothermic CSTR with Multiple Input Oscillations," *AIChE J.*, **34**, 2041 (1988).

Silveston, P. L., "Catalytic Oxidation of Carbon Monoxide under Periodic Operation," *Can. J. Chem. Eng.*, **69**, 1106 (1991).

Sinha, S., R. Ramaswamy, and J. Subba Rao, "Adaptive Control in Nonlinear Dynamics," *Physica D*, **43**, 118 (1990).

Sistu, P. B., and B. W. Bequette, "Nonlinear Predictive Control of Uncertain Processes: Application to a CSTR," *AIChE J.*, **37**, 1711 (1991).

Sterman, L. E., and B. E. Ydstie, "The Steady-state Process with Periodic Perturbations," *Chem. Eng. Sci.*, **45**, 721 (1990a).

Sterman, L. E., and B. E. Ydstie, "Unsteady-state Multivariable Analysis of Periodically Perturbed Systems," *Chem. Eng. Sci.*, **45**, 737 (1990b).

Manuscript received Mar. 20, 1992, and revision received Sept. 24, 1992.