JOURNAL OF APPLIED ELECTROCHEMISTRY **27** (1997) 774–783

Multiple model-based controller design applied to an electrochemical batch reactor

A. KRISHNAN, K. A. KOSANOVICH*

Department of Chemical Engineering, University of South Carolina, Columbia, SC 29208, USA

Received 4 March 1996; revised 17 September 1996

This study demonstrates the control of an electrochemical batch reactor, that produces the desired product in a competing chemical/electrochemical reaction network, using a multiple model-based controller design. Since this type of control framework requires a process model to provide predictions of the controlled variables, and because batch reactors are highly nonlinear and nonstationary in nature, a bank of linear, dynamic, state-space models rather than a single, linear state-space or convolution model is developed to represent the nonstationary behaviour of the batch process. It is shown that the performance of this model/controller design can provide good reactor performance in the face of known disturbances.

Nomenclature

a	electrode surface area/unit volume (cm^{-1})
A	linear dynamic matrix, Jacobian matrix
B	Jacobian matrix
c_i	bulk conc. of component i ($mol\,cm^{-3}$)
c_{is}	surface conc. of component i ($mol\,cm^{-3}$)
d	measured disturbance
D	sensitivity matrix
e	error vector
E	electrode potential (V)
h	impulse response coefficients
$\mathscr{F}$	F/RT, V^{-1}
F	Faraday constant ($96\,487\,C\ mol^{-1}$)
J	objective function
k	discrete sampling instance
k_j	rate constant for reaction j ($cm\,h^{-1}$)
M	control horizon
P	prediction horizon
r	setpoint
R	universal gas constant ($8.314\,J\,mol^{-1}K^{-1}$)
t	time (h)
t_f	batch reaction time (h)
T	reactor temperature (K)
u	manipulated variable
x	vector of state variables
x_i	dimensionless conc. of component i
x_0	initial state variable values
y	controlled variable
y_m	measured variable
$\hat{y}$	estimated variable

Greek letters

α_i	transfer coefficient at the electrode
β_i	step response coefficients
λ, Λ	weighting on the measured output
υ, Υ	weighting on the manipulated variables
Γ	discrete dynamic matrix
Φ	discrete dynamic matrix
Ψ	matrix of step response coefficients

Superscripts and subscripts

$'$	transpose
A	reactant
c	cathode
D	desired product
I	intermediate product
o	initial condition
U	undesired product

1. Introduction

Batch and semibatch reactors perform an important role in the production of low volume, high quality chemicals. This flexibility however, gives rise to challenging control problems that are due to the nonstationary and nonlinear nature of batch reactors, hard operating constraints, and multiple, prespecified setpoint trajectories. A large number of batch control schemes have been attempted including conventional feedback control, feed forward-feedback control [1], generic model control [2], adaptive regulators [3] and model-based control strategies [4, 5].

The latter controller designs have been shown to provide more robust control (better response to model uncertainty and disturbances) because a model of the process is used to predict the process performance into the future and it avoids significant online adaptation of the controller parameters which many of the other approaches require. Two specific model-based control strategies will be presented, quadratic dynamic matrix control (QDMC) and model predictive control (MPC) where the former is a specific implementation of the latter.

* Author to whom correspondence should be addressed.

0021-891X

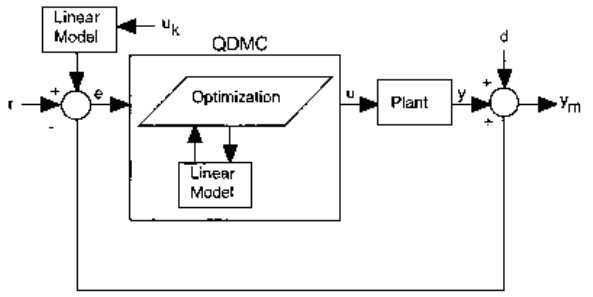

Fig. 1. Linear quadratic dynamic matrix control.

Garcia [4] employed QDMC with a convolution model (step response) of the process to control successfully a polymerization reactor that can be operated both as a semibatch and batch process. In that implementation, the convolution model is obtained by step testing a linear approximation of a nonlinear model (system of ordinary differential equations) of the process about the current operating point at every sample instance. The controller design is based on this model and the controller action is found as the least-squares solution to a quadratic error criterion with linear process inequality constraints (see Fig. 1).

In a related work, [5] employed a more complex design that uses an available nonlinear model in parallel with a linear model. Their objective is to not only improve the estimates of the future disturbances but also to separate the disturbance components into contributions from external disturbances (measurable and unmeasurable) and plant-model mismatch errors. In this fashion, the predictions of the future states from the linear model are repeatedly solved until they match those of the nonlinear model (see Fig. 2).

In both studies, the solution yields a suboptimal, nonlinear controller since the solution methodology uses linear programming to solve the optimization problem. Nevertheless, this approach can work quite well in instances where the disturbances do not move the process operations too far from the design operating point and the magnitude of the manipulated variable changes are small. For linear processes, it is well known that QDMC is essentially an optimal controller. It is our intent to provide sufficient incentive for practitioners to consider devising model-based control strategies for their application and objectives.

The organization of the paper is as follows. First, the relevant theories on quadratic dynamic matrix control (QDMC) and model predictive control are provided and used to design the controller. Second, a model of a batch electrochemical process [6, 7] where competing chemical and electrochemical reactions occur is developed and the control problem is outlined. It is assumed that optimal output trajectories are *a priori* specified and the task at hand is to design a controller to follow the trajectories. Finally, reactor performance responses are provided to illustrate the effectiveness of the controller in the presence of two types of disturbances.

2. Model-based control framework

In this section, an elementary overview of the theory is provided. Adequate references for in depth studies can be found in [4, 8]. Quadratic dynamic matrix control (QDMC) is a model-based predictive control (MPC) strategy that is an optimal predictive control method for sampled data systems. The basic idea is to formulate the control law as the solution to an optimization problem subject to constraints on the state, manipulated, and output variables. Moreover, the manipulated variable move is not only determined for the current operating point, it is also calculated for M (controller horizon) sample instances into the future [9, 10]. This strategy is based on the predicted and the future values of the outputs, over a prediction horizon $P(<\infty)$, when no additional changes in controller actions are specified for $M+1$ up to P steps into the future.

The predicted process behaviour is obtained by using an online model of the process. In this fashion, the projected errors between the desired trajectory and the predicted response can be used to adjust the future controller moves. Even though $M \geq 1$ future control moves are calculated, only the current control move is actually applied to the process; the procedure is repeated at the next sampling instant to determine the next controller action and so on. Figure 3 attempts to represent this scheme [5].

Clearly, the performance of the controller will be dependent on the quality of the online model. In classical control theory, transfer function models are used to represent the dynamic behaviour of a process that is assumed to be nearly linear, and to design an appropriate control strategy. Such models, however, require the model order to be *a priori* specified. In almost all cases involving chemical processes, the

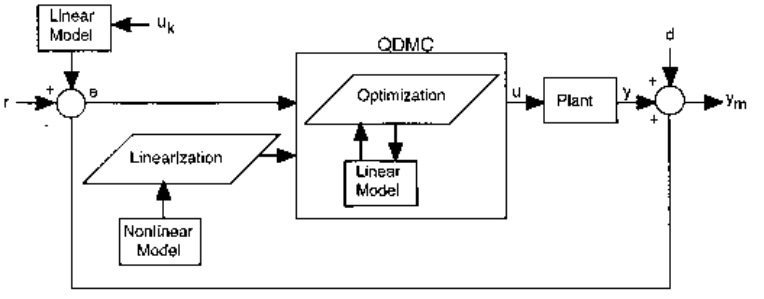

Fig. 2. Nonlinear quadratic dynamic matrix control.

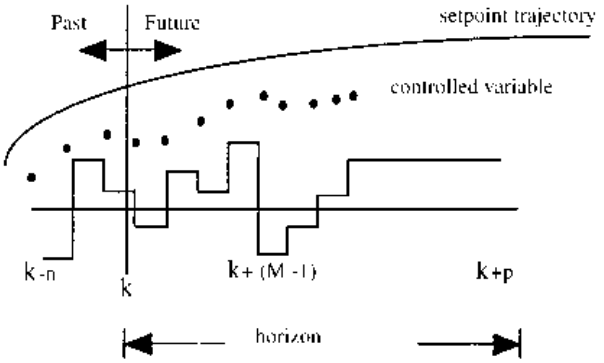

Fig. 3. Moving horizon approach of MPC.

additional transport lag parameter must also be determined. For processes having highly nonlinear dynamic behaviour, linear models cannot be specified to provide sufficient accuracy.

2.1. *Linear systems*

In conventional QDMC the model of the process has a linear, time-invariant convolution (step or impulse) form that is obtained by step or pulse testing the process. Both the transfer function and time domain convolution approaches produce representations that can be inaccurate for processes that are nonlinear and nonstationary in nature.

Without loss of generality, consider a simple, linear process that has one input and one output. Such a system can be described by

$$\begin{aligned} \dot{\boldsymbol{x}} &= \boldsymbol{A}\boldsymbol{x} + \boldsymbol{B}u \\ y &= \boldsymbol{D}'\,\boldsymbol{x} \\ x_0 &= 0 \end{aligned} \tag{1}$$

where $\boldsymbol{x} \in \mathscr{R}^n$ is the vector of states, $\boldsymbol{A} \in \mathscr{R}^{n\times n}$ is the system dynamic information, u is the system input, $\boldsymbol{B}, \boldsymbol{D} \in \mathscr{R}^{n\times 1}$ and y is the output. All physical systems are subject to constraints that may arise from actuator limitations and safety considerations. Here, it is assumed that $u(t)$ is member of a set that is convex and every sequence in that set converges to an element that is a member of the set. Similarly, $x(t)$ is a member of a set that is convex and closed* and the origin lies in the interior of the set.

In computer control implementation, the output is sampled at some sampling interval, Δ, and because of the nature of digital-to-analog converters, the input applied to the system is constant over this interval (see Fig. 3). This necessarily results in a piecewise constant control input with period Δ. Thus, the set that $u(t)$ is a member of, is the class of piecewise constant control functions, such that $u(t) = u[k]$ for each value of t between successive sampling intervals, $k\Delta < t < (k+1)\Delta$.

For batch systems, the objective is to force the output to follow a desired trajectory, in control terminology this is called the *servo* problem. That is, under certain conditions, the control input, $u(t)$, can be found so that the output y as defined in Equation 1, can be made to approach asymptotically certain command inputs (desired values).

Through simple integration, the behaviour of the system (Equation 1) at every discrete interval k can be represented by

$$\begin{aligned} \boldsymbol{x}_{k+1} &= \Phi\boldsymbol{x}_k + \Gamma u_k \\ y_k &= \boldsymbol{D}'\boldsymbol{x}_k \\ \boldsymbol{x}_0 &= 0 \end{aligned} \tag{2}$$

where Φ contains the sampled-system dynamics. By successive substitution, the state vector $\boldsymbol{x}$ can be eliminated, yielding the convolution model,

$$\begin{aligned} y_k &= \boldsymbol{D}'\boldsymbol{\Phi}\boldsymbol{x}_{k-1} + \boldsymbol{D}'\boldsymbol{\Gamma}u_{k-1} \\ y_k &= \boldsymbol{D}'\boldsymbol{\Phi}^2\boldsymbol{x}_{k-2} + \boldsymbol{D}'\boldsymbol{\Phi}\boldsymbol{\Gamma}u_{k-2} + \boldsymbol{D}'\boldsymbol{\Gamma}u_{k-1} \\ &\vdots \quad\quad \vdots \\ y_k &= \sum_{\ell=1}^{N-1} \boldsymbol{D}'\boldsymbol{\Phi}^{\ell-1}\boldsymbol{\Gamma}u_{k-\ell+1} + \boldsymbol{D}'\boldsymbol{\Phi}^N\boldsymbol{x}_{k-N+1} \end{aligned} \tag{3}$$

The product of the terms multiplying the input u are the impulse response coefficients [11]. For stable systems and for N sufficiently large, the impulse coefficients are reasonably approximated by 0. Thus, a finite N is sufficient to describe the system.

A model based on the system response to a step input can be found as

$$\hat{y}_{\ell|k} = y_0 + \sum_{i=1}^{N} \beta_i \Delta u_{k+\ell-i} \tag{4}$$

where $\hat{y}_{\ell|k}$ is the output, ℓ samples instances into the future.

Equation 4 is commonly used in conventional QDMC implementation. It can be obtained from the convolution model, Equation 3, since the step response coefficients, β_ℓ, are the sum of the first ℓ impulse response coefficients; $\Delta u_k = u_k - u_{k-1}$ represents the change in the manipulated variable. Estimates of the output, $\hat{y}_{\ell|k}$, are a function of the present and past control moves. That is,

$$\hat{y}_{k+\ell|k} = \overbrace{y_0 + \sum_{i=\ell+1}^{N} \beta_i \Delta u_{k+\ell-i}}^{\text{past}} + \overbrace{\sum_{i=1}^{\ell} \beta_i \Delta u_{k+\ell-i}}^{\text{future}} + d_{k+\ell|k} \tag{5}$$

where $d_{k+\ell|k}$ represents any unmodelled effects (see Fig. 1).

The first term in this equation is the effect of the past inputs if all future control moves are the same as u_{k-1}, that is, no more control moves are made at sample instances $k, k+1, \ldots$. The second term represents the predicted behaviour due to future inputs. If the linear convolution model is a perfect representation of the process, then $d_{k+\ell|k}$ represents only external disturbances.

Since the above equation requires knowledge of the disturbances into the future, which is not *a priori*

* Given a ball with radius r and centre 0, if x is a member of this set, then the distance from x to the centre is less than or equal to r

known, it is reasonable to assume that future disturbances will be the same as it is at the current sample time k. That is,

$$d_{k|k} = d_{k+1|k} = d_{k+2|k} = \ldots = d_{k+\ell|k}$$

An estimate of d_k, can be obtained as the difference between the measured value of the output, $y_{m,k}$, and its model prediction $\hat{y}_k$,

$$d_{k|k} = y_{m,k} - \left(y_0 + \sum_{i=1}^{N} \beta_i \Delta u_{k-i} \right) \tag{6}$$

Once the predictions are defined, we may solve for the control moves subject to constraints using optimal control theory.

The optimal control problem for the system defined by Equation 1 with control input $u(t)$ and objective term

$$J = \int_0^T L(x,u)\mathrm{d}t + \mathscr{F}(x)$$

may, in principle, be solved using the Hamilton–Jacobi–Bellman partial differential equations [12]. In the QDMC formulation, solving this set of equations are avoided by solving an *open-loop* optimal control problem for a sequence of states and applying the control action assuming that the future disturbance will be the same as it is at the current sample time k.

This is accomplished by selecting a quadratic objective function that reflects the error between the predicted value of the output and the target value, r_k, and minimizing this over the set of admissible control changes subject to satisfying constraints on the inputs and outputs and the model, Equation 5,

$$\min_{\Delta u} J := \lambda \sum_{\ell=1}^{P} (r_{k+\ell} - \hat{y}_{k+\ell|k})^2 + v \sum_{\ell=1}^{M} \Delta u_{k+\ell-1}^2 \tag{7}$$

λ is a 'cost' on not achieving the desired value, and v is a 'penalty' for excessive changes in the manipulated variable. If the manipulated variable u is not penalized then unacceptably large control moves can result.

The tuning parameters for this formulation are the prediction horizon P, the controller horizon M, and any weighting terms on the cost or penalty portions of the objective function. With the controller in place, there are no assurances of the closed-loop stability of the system, regardless of whether the system being controlled is linear or nonlinear. For an excellent discussion on this issue see the work by Mayne [13].

By disregarding the constraints, the solution to Equation 7 reduces to the standard least-squares problem which can be solved explicitly. The reader is referred to the work by Morshedi *et al.*, [14] for the proof of this claim.

The QDMC formulation is one approach of the more general MPC framework. In MPC, the open-loop optimal control problem is solved using dynamic programming and yields a time-varying, nonlinear, control law. However, while an optimal solution may be found, it does not imply stability because of the assumption of a finite horizon (cf. linear quadratic optimal controller). It has been shown that stability can be achieved by varying the tuning parameters [15]; how to change them, however, largely remains a trial and error process.

In practice, the choice of the horizon is selected to be long sufficiently, so as to ensure that the process has settled close to its equilibrium state at time $k + P$, implicitly satisfying the stability constraint. If the horizon is chosen to be *infinite* then stability can be guaranteed but the resulting optimal control problem cannot be solved in general. Several researchers have addressed this issue by reformulation of the objective function or by developing specific conditions that guarantee stability; the details can be found in [16–18].

2.2. *Nonlinear systems*

The above discussion has been carried out with the basic assumption that the process to be controlled is linear. However, all real processes are nonlinear to varying degrees and batch processes are more so than most.

A general nonlinear system to be controlled is given by the following set of ordinary differential equations,

$$\begin{aligned} \dot{\boldsymbol{x}} &= f(\boldsymbol{x}, \boldsymbol{u}) \\ \boldsymbol{y} &= \boldsymbol{g}(\boldsymbol{x}) \\ \boldsymbol{x}_0 &= \boldsymbol{x}(0) \end{aligned} \tag{8}$$

where $\boldsymbol{f}$ and $\boldsymbol{g}$ are vector-valued, nonlinear functions of the states $\boldsymbol{x} \in \mathscr{R}^n$, the inputs $\boldsymbol{u} \in \mathscr{R}^m$ and the outputs $\boldsymbol{y} \in \mathscr{R}^q$. The properties of the admissible set of $\boldsymbol{u}$ and $\boldsymbol{x}$ still hold.

If the process is nonstationary and sufficiently nonlinear, such as a batch process, then a single linear model may not suffice to represent its behaviour over the batch cycle. Thus, the values of the step response coefficients, β_i, will depend on the values of $\boldsymbol{x}$ and $\boldsymbol{u}$ at each sampling instant, meaning that they are 'local' rather than 'global' coefficients.

A means of obtaining the step response model coefficients is to linearize the system of nonlinear equations about some meaningful point, $\{x_s, u_s\}$; then step test the resulting linear model. The effect of the past inputs and the disturbance are computed using the step response model.

The linearization/step testing procedure must necessarily be repeated at each sampling instant, generating a bank of step response models, each valid within a small neighbourhood of the operation. Related work includes that of Garcia [4] and Gattu and Zafiriou [19] on a semibatch and batch chemical process.

The general MPC strategy allows the use of state-space models directly, thereby avoiding the step testing and step response coefficients calculations [15,20]. However, to circumvent solving a nonlinear programming problem, the linearization step will be retained.

A linear approximation to Equation 8 can be obtained by retaining only the first order terms in a Taylor series expansion about some nominal state, $\boldsymbol{x}_s$ and manipulated variable $\boldsymbol{u}_s$,

$$\begin{aligned} \dot{\bar{\boldsymbol{x}}} &= \boldsymbol{A}\bar{\boldsymbol{x}} + \boldsymbol{B}\bar{\boldsymbol{u}} \\ \boldsymbol{y} &= \boldsymbol{D}'\bar{\boldsymbol{x}} \\ \bar{\boldsymbol{x}}_0 &= 0 \end{aligned} \tag{9}$$

where $\boldsymbol{A}$ and $\boldsymbol{B}$ are the Jacobian matrices. $\bar{\boldsymbol{x}}$ and $\bar{\boldsymbol{u}}$ are the deviations of the state and manipulated variable values from their nominal values, respectively. A suitable linear, discrete, state-space model, obtained by integration of Equation 9, will be used in the MPC strategy to provide current and future estimates of the output.

In the case of multiple inputs, Equation 7 becomes

$$\begin{aligned} \min_{\Delta u} J := &\sum_{\ell=1}^{P} e'_{q,k+\ell}\boldsymbol{\Lambda}e_{q,k+\ell} + \sum_{\ell=1}^{M}\left[\boldsymbol{u}'_{k+\ell-1}\boldsymbol{\Upsilon}_1\boldsymbol{u}_{k+\ell-1}\right. \\ &\left.+\Delta\boldsymbol{u}_{k+\ell-1}\boldsymbol{\Upsilon}_2\Delta\boldsymbol{u}_{k+\ell-1}\right] \end{aligned} \tag{10}$$

The resulting controller moves are those that minimize $\Delta\boldsymbol{u}$ subject to satisfying equality and inequality constraints imposed by the system and the allowable limits on the inputs and outputs. As before, $\boldsymbol{\Lambda}$ is a weighting matrix that relates the relative importance of the errors, $\boldsymbol{\Upsilon}_1$ and $\boldsymbol{\Upsilon}_2$ are penalties on using too 'fast' a control action, and $\boldsymbol{e}$ is the predicted deviation from the desired value.

2.3. *Controllability and stability*

Controllability and stability are readily established for linear systems [21]. However, for nonlinear systems the proof is almost always system dependent. In the case where there the nonlinear system is approximated by local, linear models, it is enough to show that each local model is asymptotically controllable.

Definition: A linear system is *asymptotically controllable* if and only if the unstable modes (eigenvalues) are controllable.

Asymptotic controllability of each linear system implies 'asymptotic stability' of each linear system, but more importantly it ensures that the nonlinear system, Equation 8 is locally asymptotically stable as the following theorem indicates [22].

Theorem: Assume that the pair $(\boldsymbol{x}_0, \boldsymbol{u}_0)$ *is such that for the time-invariant, continuous-time system* $\sum : \dot{\boldsymbol{x}} = f(x,u)$, *with* $\boldsymbol{x} \in \mathscr{R}^n$ *and* $\boldsymbol{u} \in \mathscr{R}^m$,

$$f(\boldsymbol{x}_0, \boldsymbol{u}_0) = 0$$

Assume that the linearization of $\sum$ *is asymptotically controllable. Then* $\sum$ *is locally asymptotically controllable (to* $\boldsymbol{x}_0$*). Moreover, there exists in that case an* $n \times m$ *matrix* $\boldsymbol{K}$ *such that the closed-loop system*

$$\dot{\boldsymbol{x}} := f(\boldsymbol{x}, \boldsymbol{u}_0 + \boldsymbol{K}(\boldsymbol{x} - \boldsymbol{x}_0))$$

is locally asymptotically stable. The proof can be found in Appendix 1.

The system defined by Equation 2 will be open-loop stable if and only if the continuous system, Equation 9 is stable.

3. Controller design

Two model predictive controller designs are developed. The first employs the kth linear model to provide the output predictions and the disturbance estimate over the prediction horizon P. Thus, at any given sampling instant, the kth model is *fixed* and is used to estimate $\hat{y}_{k+\ell}, \{\ell = 1, 2, \ldots, P\}$.

The second scheme, uses a bank of linear models obtained from the *offline* evaluation of the Jacobian about the nominal process variables' profiles to provide the projected estimates. At sampling instance k, the kth model is used to provide the estimate $\hat{y}_k$. To obtain the future estimates at time k, the $(k+1)$ model is used to obtain $\hat{y}_{k+1|k}$; the $(k+2)$ model is used to obtain $\hat{y}_{k+2|k}$ and so on.

In what follows, FMPC (fixed model predictive control) will be used to refer to scheme I and MMPC (moving model predictive control) will be used for scheme II.

4. Electrochemical reactor

Electrochemical (EC) batch reactors are used in a number of processes such as in the production of specialty inorganic and organic chemicals. EC systems differ significantly from conventional chemical systems in that dynamic manipulation of voltage or current is much faster responding as compared to the conventional manipulation of temperature or flowrate in chemical reactors (transport delays). It is also possible to use control strategies in which the potential or current is rapidly perturbed since the electrical 'inertial'† effect is much less than that of the thermal or mass transfer effects. There are however, difficulties in the control of EC batch reactors. Most noticeably, there appears to be only a single manipulated variable, that is, the potential or the current.

In this particular application, the selected reactions, which occur at the surface of the electrodes, involve the competing chemical/electrochemical reactions,

$$\begin{aligned} \mathrm{A} &\rightarrow \mathrm{I} \quad \text{electrochem} \\ \mathrm{I} &\rightarrow \mathrm{D} \quad \text{chem} \\ \mathrm{I} &\rightarrow \mathrm{U} \quad \text{electrochem} \end{aligned}$$

where I is an intermediate, D is the desired product, and U is the undesired product. This general operational mechanism has been used to represent the reduction of nitrobenzene to *p*-aminophenol and aniline, through an intermediate, phenylhydroxylamine [6, 7, 23]. In this reaction sequence where there are competing chemical and electrochemical reactions, it is extremely difficult to control the con-

† inductance and capacitance effects tend to limit the rates of change in voltages and currents

centration of the product obtained by the chemical route due to a lack of sensitivity to the manipulated variable. This example is used to illustrate model-based concepts applied to an electrochemical reactor and not to emphasize the particular chemistry.

Several simplifying assumptions have been made about the transport and kinetic processes: (i) the reactor is well-mixed with mass transfer resistance occurring as a result of diffusion through the Nernst diffusion layer; (ii) the homogeneous chemical reaction I → D follows first-order kinetics in the bulk and consumes an inconsequential amount of reactant in the thin Nernst diffusion layer; (iii) the capacitance of the double layer is negligible; (iv) the current distribution is uniform; and (v) the charge transfer reactions are irreversible with a first-order dependence on reactant concentration.

The component continuity equations that describe the production or consumption of a component are:

$$\begin{aligned}
\dot{c}_{\mathrm{A}} &= -ak_1 c_{\mathrm{As}} e^{-\alpha_1 \mathscr{F} E} \\
\dot{c}_{\mathrm{I}} &= ak_1 c_{\mathrm{As}} \mathrm{e}^{-\alpha_1 \mathscr{F} E} - ak_2 c_{Is} \mathrm{e}^{-\alpha_2 \mathscr{F} E} - k_3 c_I \\
\dot{c}_{\mathrm{D}} &= k_3 c_{\mathrm{I}} \\
\dot{c}_{\mathrm{U}} &= ak_2 c_{\mathrm{Is}} \mathrm{e}^{-\alpha_2 \mathscr{F} E} \\
c_{\mathrm{A}}(0) &= c_{\mathrm{A}}^0
\end{aligned} \tag{11}$$

where the subscript s represents the surface concentration, $\mathscr{F} = F/RT$ where F is the Faraday constant, E is the electrode potential, k_j is the reaction rate constant for reaction j, a is the electrode surface area per unit volume of the reactor, and α_j is the transfer coefficient of the desired reaction. Values for the mass-transfer coefficient of species i indicate the relative degree to which the reactions are limited by the kinetics or the mass-transfer resistance. In this study it will be assumed that the mass-transfer resistance of component A is twice as great as that of I,

$$\begin{aligned}
k_{\mathrm{mA}} &= 4k_{\mathrm{m}} \\
k_{\mathrm{mI}} &= 2k_{\mathrm{m}} \\
1 &< k_{\mathrm{m}} < 10^5
\end{aligned}$$

The total batch time t_f is set at 4 h and the sample time is 3.6 s. The values of all parameters can be found in [7].

The optimal time-varying electrode potential which maximizes the production of D is found for the condition of $\alpha_1 > \alpha_2$ [7]. In this study, the objective will be to track the prescribed desired concentration profile D, that is a result of the optimal time-varying electrode potential, but in the presence of disturbances.

The optimal concentration profiles are shown in Fig. 4. Observe, that the yield of D is about six times that of U and that at the end of the batch cycle, there remains 32% of the intermediate I but $< 2\%$ A. In Fig. 5, the potential is ramped from an initial value of 1 V to the steady value of 0.2 V.

The surface concentrations can be eliminated from Equation 11 by applying a mass balance across the Nernst diffusion layer,

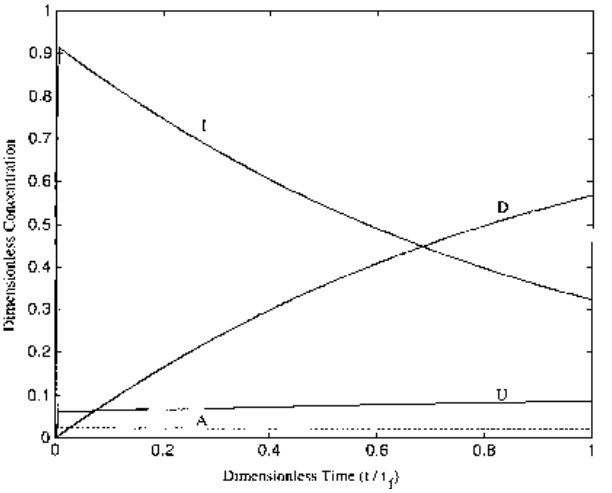

Fig. 4. Dimensionless concentration profiles using the optimal electrode potential. (A) feed component, (I) intermediate component, (D) desired product, (U) undesired product.

$$\begin{aligned}
k_{\mathrm{mA}}(c_{\mathrm{A}} - c_{\mathrm{As}}) &= k_1 c_{\mathrm{As}} \mathrm{e}^{-\alpha_1 \mathscr{F} E} \\
k_{\mathrm{mI}}(c_{\mathrm{I}} - c_{\mathrm{Is}}) &= k_2 c_{\mathrm{Is}} \mathrm{e}^{-\alpha_2 \mathscr{F} E} - k_1 c_{\mathrm{As}} \mathrm{e}^{-\alpha_1 \mathscr{F} E}
\end{aligned}$$

where $k_{\mathrm{m}i}$ is the mass-transfer coefficient of species i. Introduction of the following dimensionless terms

$$\begin{aligned}
x_i &:= \frac{c_i}{c_{\mathrm{A}}^0} & k_i^* &:= ak_i t_{\mathrm{f}} \\
\zeta_{\mathrm{A}} &:= \frac{k_{\mathrm{mA}}}{k_1 u_1 + k_{\mathrm{mA}}} & \zeta_I &:= \frac{k_{\mathrm{mI}}}{k_2 u_2 + k_{\mathrm{mI}}} \\
u_1 &:= \mathrm{e}^{-\alpha_1 \mathscr{F} E} & u_2 &:= \mathrm{e}^{-\alpha_2 \mathscr{F} E} \\
t^* &:= \frac{t}{t_{\mathrm{f}}}
\end{aligned}$$

into Equation 11 yields the following dimensionless form,

$$\begin{aligned}
\dot{x}_{\mathrm{A}} &= (k_1^* \zeta_{\mathrm{A}} u_1) x_{\mathrm{A}} \\
\dot{x}_{\mathrm{I}} &= \left(k_1^* u_1 \zeta_{\mathrm{A}} - k_2^* \frac{k_1}{k_{\mathrm{mI}}} \zeta_{\mathrm{A}} \zeta_I u_1 u_2 \right) x_{\mathrm{A}} - \left(k_3^* + k_2^* \zeta_{\mathrm{I}} u_2 \right) x_{\mathrm{I}} \\
\dot{x}_{\mathrm{D}} &= k_3^* x_{\mathrm{I}} \\
\dot{x}_{\mathrm{U}} &= (k_2^* \zeta_{\mathrm{I}} u_2) x_{\mathrm{I}} + \left(k_2^* \frac{k_1}{k_{\mathrm{mI}}} \zeta_{\mathrm{A}} \zeta_{\mathrm{I}} u_1 u_2 \right) x_{\mathrm{A}} \\
x_{\mathrm{A}}(0) &= 1.0
\end{aligned} \tag{12}$$

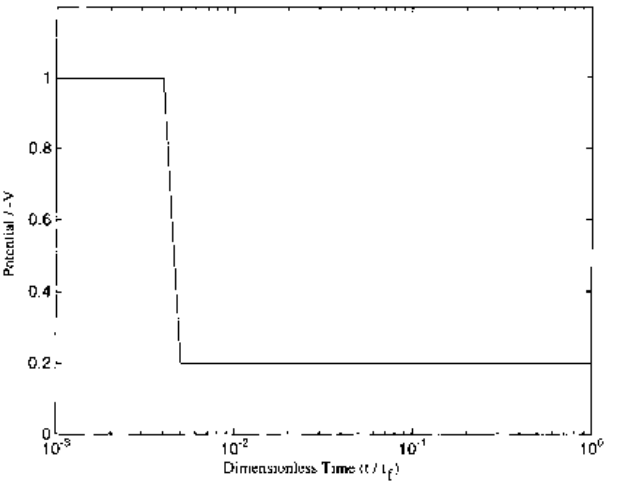

Fig. 5. Optimal electrode potential.

Control variables u_1 and u_2 are not independent, thus there is only one manipulated variable,

$$u_1 = u_2^{\alpha_1/\alpha_2} = u_2^{\alpha}$$

The electrode potential at the cathode is *a priori* constrained by

$$E_{c,max} \leq E \leq \infty$$

to avoid undesirable electrolyte and/or solvent electrochemistry. This necessarily constrains the control variables, $\{u_1, u_2\}$:

$$0 \leq u_1 \leq u_{1,max} \quad u_{1,max} = e^{-\alpha_1 \mathscr{F} E_{c,max}}$$
$$0 \leq u_2 \leq u_{2,max} \quad u_{2,max} = e^{-\alpha_2 \mathscr{F} E_{c,max}}$$

The linearized form (see Appendix 2) of Equation 12, *a priori* evaluated at every sample instance using the optimal variables' values, is used in the FMPC and MMPC strategy.

4.1. *Controllability and stability of EC reactor*

Let the controlled variable be the desired product x_D. Then, from Equation 9, $\boldsymbol{D}' = [0\,0\,1\,0]$ and the rank of $\boldsymbol{A}$ is found to be two. If the system is completely controllable, the linear rank condition should indicate a rank of four, which is the number of states [21]. However, the rank is found to be three, implying that *at least* one of the states is uncontrollable.

A similarity transformation can be found that will yield a matrix fraction description of the controllable and uncontrollable states [24]. Moreover, a particular choice of the transformation can be made so that the undesirable product, x_U, is also the uncontrollable state. Analysis reveals that the uncontrollable state results from having common terms in the denominator and numerator polynomial of the transfer function[‡] cancelling each other. Here, this cancellation involves a common root at zero; explaining the rank deficiency.

There is almost but not quite another cancellation of common terms that occur near zero. That is, besides the previous zero root, the denominator polynomial has a second one; however, the numerator polynomial has a root that is nearly but *not exactly* at zero. The implication of this is that although in theory there are three controllable states, one is nearly uncontrollable. Thus, from a practical perspective, only two of the states can be controlled effectively.

The problem with the uncontrollable state does not severely affect our ability to control the EC system. First, it is known by the above theorem to be locally asymptotically stable from an input/output perspective. Thus, control of the desired product is possible. Second, the process only operates for a finite time. Therefore, the uncontrollable states will not have sufficient time to create serious operational problems meaning that we can control the desired product but not the undesired one.

[‡] Laplace transform of Equation 9

5. Results and discussion

Both a fixed model (FMPC) and a moving model (MMPC) based predictive controller designs, are implemented on the electrochemical batch reactor (EC). The FMPC strategy uses the kth linear, dynamic state-space model to provide the current estimate of the output at time k and the predictions $\{k+\ell, \ell = 1, 2, \ldots, P\}$ into the future. In contrast, the MMPC strategy uses the $\{k, k+1, \ldots, P\}$ linear, dynamic state-space models to provide the current and future estimates.

The objective will be to maintain the desired product profile (see Fig. 4), as determined from the optimal open-loop studies, by changing the electrode potential *in the presence of* two different disturbances imposed independently on the system. These disturbances are (i) the presence of a small amount (10%) of I at the start of the batch cycle and (ii) a reduction in the available electrode area (40%) which can occur in EC reactors due to the 'clogging' of the porous electrodes or the formation of films in the case of ordinary electrodes.

The initial presence of I and any applied potential will preferentially produce x_U; while the reduction in the electrode surface area will prevent the expected production of I within the allowed batch time hence, both the production of x_D and x_U will be affected. This latter disturbance demonstrates the robustness of the controller design to model uncertainty.

The prediction horizon, P, is chosen to be 3 and that of the control horizon, M, to be 2. It is found that increasing P has a destabilizing effect on the closed-loop process performance. There are two reasons for this. First, the assumption of keeping the last control action constant over the remainder of the prediction horizon is not reasonable. This assumption is valid for setpoint tracking, but in the servo case such as this, a constant control effort will result in large deviations from the desired trajectory. Second, this effect is compounded by the fact that as the errors increase, the linear models become less valid. The latter fact also supports the subsequent conclusion that the performance of MMPC is better than that of FMPC.

Figures 6 and 7 illustrate the reactor performance and the manipulated variable profile (electrode potential), respectively using the FMPC and MMPC strategy under ideal conditions. The 'ref' line is the desired trajectory. The FMPC strategy produces 20% less D while producing 12% more of the undesired product. In contrast, MMPC produces 9.5% less D as compared to the reference value but, 12% more than FMPC.

Although the final production value of U by the MMPC strategy is the same as FMPC, it is worthwhile noting that the production of U after 80% of the batch cycle is complete exhibits a ramp-like growth. This is due to the zero eigenvalue which produces an integrating effect. Thus, while for most of the batch cycle, MMPC maintained a low pro-

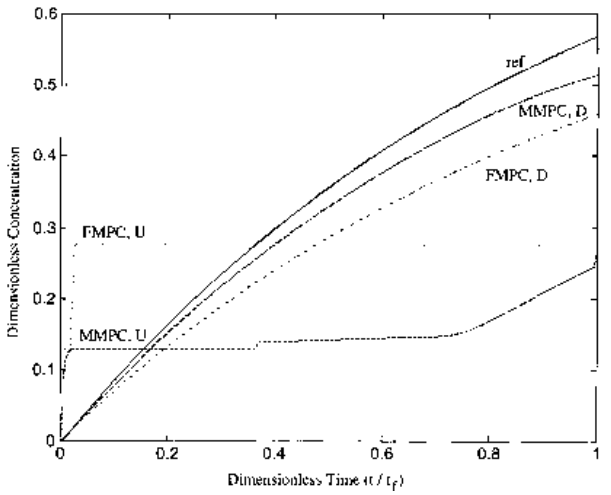

Fig. 6. Dimensionless concentration profiles with no disturbance present. Key: (· · · · · ·) FMPC strategy; (_ _ _ _) MMPC strategy, (D) desired product, (U) undesired product; (——) reference trajectory for desired product.

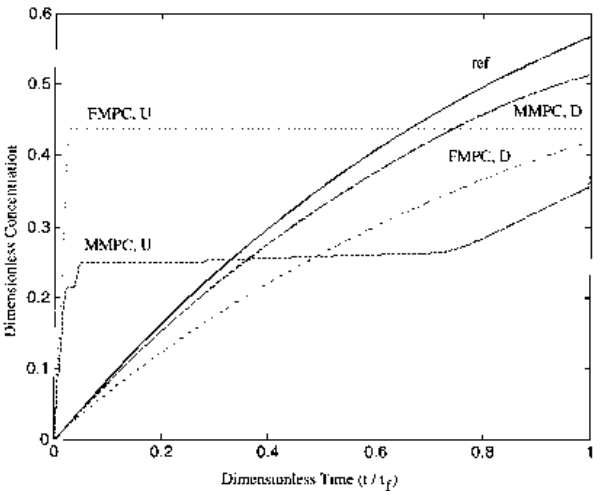

Fig. 8. Dimensionless concentration profiles in the presence of 10% increase in the intermediate component I. Key: (· · · · · ·) FMPC strategy; (_ _ _ _) MMPC strategy, (D) desired product, (U) undesired product); (——) reference trajectory for desired product.

duction rate of U, because only two of the states are controllable, it is unable to suppress completely the unfavourable production of U over the entire batch cycle and simultaneously maintain the desired production of D.

The electrode potential has a final value of −0.34 and 0.33 V for FMPC and MMPC strategy, respectively. In contrast to the reference electrode potential, both FMPC and MMPC controller actions are more aggressive early into the batch, and in the case of MMPC later on as well. Thus, we can conclude that the model predictive controller actions are selected so as to make an adjustment at every opportunity.

Figures 8 and 9 demonstrate the reactor performance and the electrode potential, respectively, when there is 10% I at the start of the batch cycle; I acts as a contaminant. The FMPC strategy produces 22% and 35% less D as compared to MMPC and the reference value, respectively. MMPC performs the same as in the ideal case (previous results) which indicates that having a more accurate model of the process will lead to better rejection of the disturbance. The production of U by the FMPC strategy is 23% more than that of the MMPC strategy. The final electrode potentials are −0.69 and 0.65 V for FMPC and MMPC, respectively.

Figures 10 and 11 represent the reactor performance and the electrode potential, respectively, for a 40% reduction in the electrode area. This case demonstrates the controller's robust performance to plant/model mismatch. MMPC produces 22% more D and 56% less U as compared to FMPC. The electrode potential profiles of FMPC are similar to those in Fig. 9; however, they are more aggressive in the MMPC strategy for the 10% increase in I. This implies that the presence of I has a greater effect on the reactor performance than the deterioration of the electrode area. Nevertheless, the FMPC and the MMPC controllers provided good, robust performance in both cases.

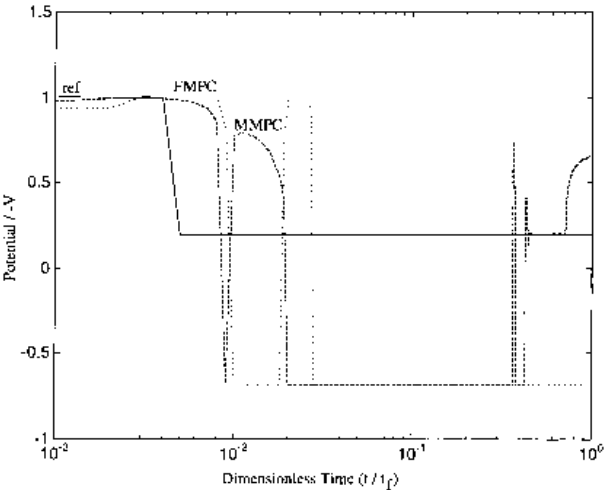

Fig. 7. Optimal electrode potential with no disturbance present. Key: (· · · · · ·) FMPC strategy; (_ _ _ _) MMPC strategy; (——) optimal electrode potential profile.

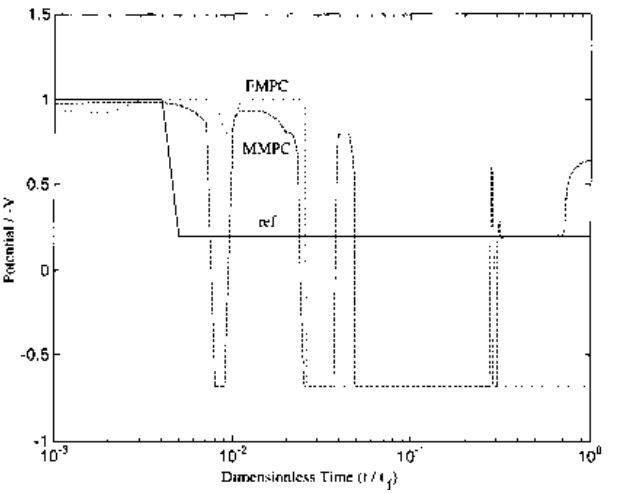

Fig. 9. Optimal electrode potential in the presence of 10% increase in the intermediate component I. Key: (· · · · · ·) FMPC strategy; (_ _ _ _) MMPC strategy; (——) optimal electrode potential profile.

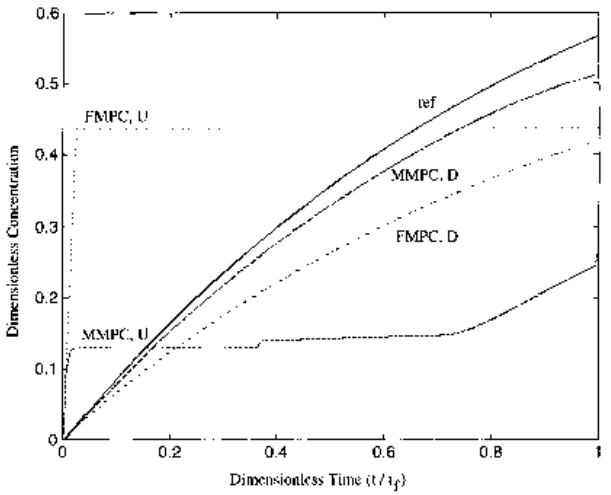

Fig. 10. Dimensionless concentration profiles in the presence of 40% increase in the electrode area. Key: (·····) FMPC strategy; (_ _ _ _) MMPC strategy, (D) desired product, (U) undesired product; (——) reference trajectory for desired product.

6. Summary

This work has demonstrated two model predictive controller designs that use linear, dynamic, statespace models to control the production of a component in an electrochemical/chemical reaction network in a batch reactor. These models were developed from the linearization of a nonlinear first principles model of the reactor, one at each sample time k.

The first design, FMPC, employs only the kth linear, dynamic, state-space model to predict the kth and future estimates; the other, MMPC, uses the kth, $(k+1)$, up to the $(k+P)$ model to provide the current and future estimates. In addition the MMPC linear models are evaluated not at the current state nor online, rather they are *a priori* determined offline using the desired trajectory profile.

This key point has two implications. First, the offline calculations are less burdensome for online implementation of more complex systems and second, for highly nonlinear systems, the current state should only be used to evaluate the current linear model because the linear model is only valid within a neighbourhood of that point. The use of the kth measurement to evaluate the $(k+1)$st model and so on presumes that the system is time-invariant, which it is not appropriate for almost all batch process. Similarly, to use the kth linear model to represent the complete time-dependent behaviour of a nonstationary process does not produce good controller results. This latter point is demonstrated by the results obtained for the FMPC controller strategy.

The performance of the MMPC controller was found to be superior to the FMPC design for the reasons stated above. Although, the MMPC controller did not exactly track the desired profile, it provided good reactor performance in the presence of a disturbance and excellent robustness to one example of plant/model mismatch.

The model predictive control framework unlike the conventional feedback controllers is attractive because it uses optimization to determine the optimal control action in the presence of constraints on the states, the outputs and the manipulated variables for the current sampling instance and up to M moves into the future. It also provides an indication of the process performance if no more control action is taken after M moves.

In the case of the electrochemical system studied, it was found that $P > 3$ had a destabilizing effect on the closed-loop process performance (cf. continuous processes $P = 30$). This can be understood when one considers that approximate models are being used to represent the highly nonlinear process. Thus, holding the last control action constant will result in large deviations from the desired profile.

An analysis of the controllability of the electrochemical system was also presented as it was relevant to the controller design. In particular, it was noted that of the four states, only two can be controlled in practice. This corroborates the performance of both controllers.

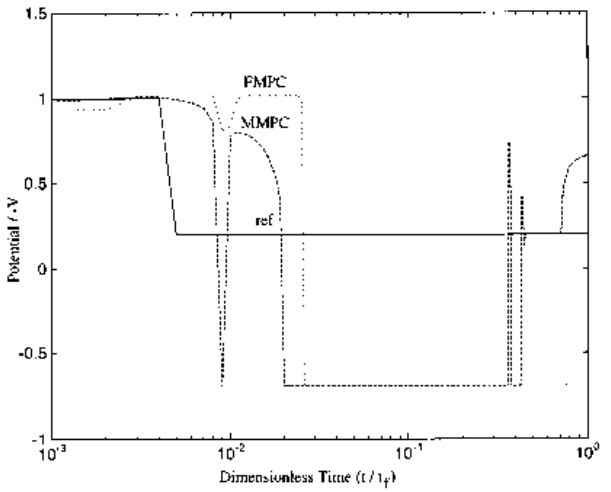

Fig. 11. Optimal electrode potential in the presence of 0% increase in the electrode area. Key: (·····) FMPC strategy; (_ _ _ _) MMPC strategy; (——) optimal electrode potential profile.

Acknowledgement

The authors would like to acknowledge Dr M. J. Piovoso of DuPont, Central Science & Engineering and Prof. P. S. Fedkiw of North Carolina State University for their assistance. Funding was provided by NSF grant number CTS-95-03437.

References

[1] A. Jutan and A. Uppal, *Ind. Eng. Chem. Process. Des. Dev.* **23** (1984) 597–602.
[2] B. J. Cott and S. Macchietto, *Ind. Eng. Chem. Res.* **28** (1989) 1177–84.
[3] W. R. Cluett, S. L. Shah and D. G. Fisher, *Chem. Eng. Comm.* **38** (1985) 67–78.
[4] C. E. Garcia, 'Quadratic/dynamic matrix control of nonlinear processes – an application to a batch reaction process', *in* AIChE Nat. Meeting (1984).
[5] T. Peterson, E. Hernandez, Y. Arkun and F. J. Schork, *CES* **47**(4) (1992) 737–53.

[6] R. Bakshi and P. S. Fedkiw, *J. Appl. Electrochem.* **23** (1993) 715–27.
[7] R. Bakshi and P. S. Fedkiw, *ibid.* **24** (1994) 1116–23.
[8] C. E. Garcia and M. Morari, *Ind. Eng. Chem. Process. Des. Dev.* **21**(2) (1993) 308–23.
[9] C. E. Garcia, D. M. Prett, and M. Morari, *Automatica* **25**(3) (1989) 335–48.
[10] J. B. Rawlings, E. S. Meadows and K. R. Muske, Nonlinear model predictive control: a tutorial and survey, *in* 'ADCHEM ' 94' *Proc., Kyoto, Japan* Elsevier, Amsterdam (1994).
[11] D. M. Prett and C. E. Garcia, 'Fundamental Process Control', Butterworth, Stoneham, MA, (1984).
[12] A. E. Bryson and Y. Ho, 'Applied Optimal Control', John Wiley, New York (1975).
[13] D. Q. Mayne, Optimization in modelbased control, *in* 'DYCORD$^{+'}$ 95', *Proc., Helsingφr, Denmark* (edited by J. B. Rawlings), Danish Automation Society, (1995) pp. 229–42.
[14] A. M. Morshedi, *in* 'Chemical Process Control III' (edited by M. Morari and T. J. M^{c}Avoy), Elsevier, New York, (1986), CACHE publication.
[15] J. H. Lee, M. Morari and C. E. Garcia, *Compt. Chem. Engng.* **4** (1994) 707–17.
[16] J. B. Rawlings and K. R. Muske, *IEEE Trans. Autom. Control.* **38** (1993) 1512–16.
[17] S. Keerthi and E. Gilbert, *J. Optim. Theory Appl.,* **57** (1988) 265–93.
[18] D. Q. Mayne and H. Michalska, *IEEE Trans. Autom. Control.* **35** (1990) 814–24.
[19] G. Gattu and E. Zafiriou, On the stability of nonlinear quadratic dynamic matrix control, *in* 'DYCORD^{+}' 92', *Proc., College Park, MD* (1992) pp. 303–8.
[20] S. Li, K. Y. Lim and D. G. Fisher, *AIChE J.* **35**(2) (1983) 241–49.
[21] W. J. Rugh, 'Linear Systems Theory', Prentice-Hall, Englewood Cliffs, NJ (1993).
[22] E. D. Sontag, 'Mathematical Control Theory: Deterministic Finite Dimensional Systems', Springer-Verlag, New York (1990).
[23] T. R. Nolen and P. S. Fedkiw, *J. Appl. Electrochem.* **20** (1990) 370–76.
[24] T. Kailath, 'Linear Systems', Prentice-Hall, Engelwood Cliffs, NJ (1981).

Appendix A

Proof:
It is enough to prove that

$$\dot{x} = f_{cl}(x) := f(x,\, u^0 + F(x - x^0))$$

is locally asymptotically stable as stabilizability implies controllability. That is, a linear system is asymptotically controllable if and only if its unstable modes are controllable.

$$\Sigma : \dot{x} = f(x,\, u)$$

linearize:

$$f(x,\, u) = A(x - x^0) + B(u - u^0) + g(x - x^0,\, u - u^0)$$

$$\Sigma_{cl} : \dot{x} = f_{cl}(x) := f(x,\, u^0 + F(x - x^0))$$

$$f_{cl}(x) = A(x - x^0) + B(u^0 + F(x - x^0) - u^0) + g(x - x^0,\, u^0 + F(x - x^0) - u^0)$$

$$f_{cl}(x) = A_{cl}(x - x^0) + \gamma(x - x^0,\, F(x - x^0))$$

where

$$A_{cl} := A + BF$$

$$\gamma(x - x^0,\, F(x - x^0)) := g(x - x^0,\, F(x - x^0))$$

$$\lim_{\| \alpha, \beta \| \to \infty} \frac{\| \gamma(\alpha, \beta) \|}{\| \alpha, \beta \|} = 0$$

For nonlinear system, the best one can do is to establish that the system is asymptotically stable or asymptotically controllable is to use the Lyapunov function method.

Choose the Lyapunov function such that $V(x)$ is proper at x^0.

$$V(x) := (x - x^0)^T P (x - x^0)$$

on $\mathscr{R}^n$, $x \in \mathscr{O}$ where $\mathscr{O}$ is some neighbourhood about x^0, that is a compact* subset of χ. P is a positive definite, symmetric matrix, $P > 0$, such that

$$A_{cl}^T P + P A_{cl} = I$$

and is sufficient for

$$V(x) := x^T P x$$

to be a Lyapunov function for Σ_{cl}

$V(x)$ proper at x^0 for small enough ε implies

$$\{x \in \chi | V(x) \le \varepsilon\} \qquad \varepsilon > 0$$

$V(x)$ must also be positive definite on $\mathscr{O}$. By the above choice of $V(x)$ it does,

$$V(x^0) = 0 \qquad x^0 = 0$$
$$V(x) > 0 \qquad \forall x \in \mathscr{O} \qquad x \neq 0$$

For each $x \neq x^0 \in \mathscr{O}$ there is some finite time $\sigma \in \mathscr{T}, \sigma > 0$ and some control $u(\cdot) \in \mathscr{U}$, the admissible set for all x

$$\nabla V(x) \cdot f_{cl}(x) < 0 \qquad \forall t \in [0,\, \sigma] \tag{A1}$$

whenever $(x - x^0)$ is small enough.

By the continuity of $\nabla V(x)$ and $f_{cl}(x)$, the dot product in Equation (A1) is given by

$$\nabla V(x) f_{cl}(x) = - \| x - x^0 \|^2 + 2(x - x^0)^T P \gamma(x - x^0) \le 0$$

It remains to show that the second term is less than or equal to zero. Using the Cauchy-Schwarz inequality, we can write

$$|2(x - x^0)^T P \gamma(x - x^0)| \le \| x - x^0 \| \qquad \| 2P\gamma(x - x^0) \|$$

Defining $c := 2P$,

$$|2(x - x^0)^T P \gamma(x - x^0)| \le c \| x - x^0 \| \quad \| \gamma(x - x^0) \|$$

$$\frac{|2(x - x^0)^T P \gamma(x - x^0)|}{\| x - x^0 \|^2} \le c \frac{\| \gamma(x - x^0) \|}{\| x - x^0 \|}$$

For $\| (x - x^0) \|$ small enough and since $\gamma(x - x^0)$ is of order $0(x - x^0)$ then

$$\frac{\| \gamma(x - x^0) \|}{\| x - x^0 \|} \to 0$$

Hence, it follows that

$$\nabla V(x) \cdot f_{cl}(x) < 0$$

Appendix B

The linearization of Equation 12 yields,

$$\nabla_x \mathbf{f}|_{x_s, u_s} = \begin{pmatrix} -k_1^* \zeta_A u_2^\alpha & 0 & 0 & 0 \\ \zeta_A u_2^\alpha (k_1^* - k_2^* \frac{k_1}{k_{ml}} \zeta_I u_2) & -(k_3^* + k_2^* \zeta_I u_2) & 0 & 0 \\ 0 & k_3^* & 0 & 0 \\ k_2^* \frac{k_1}{k_{ml}} \zeta_A \zeta_I u_2^{1+\alpha} & k_2^* \zeta_I u_2 & 0 & 0 \end{pmatrix}$$

$$\frac{\partial \mathbf{f}}{\partial u_2}|_{x_s, u_s} = \begin{pmatrix} -k_1 x_A u_2^\alpha \left(\frac{\mathrm{d}\zeta_A}{\mathrm{d}u_2} + \zeta_A \frac{\alpha}{u_2}\right) \\ k_1 x_A u_2^\alpha \left(\frac{\mathrm{d}\zeta_A}{\mathrm{d}u_2} + \zeta_A \frac{c}{u_2}\right) - k_2 \frac{k_1}{k_{ml}} u_2^{\alpha+1} \left(\zeta_I \frac{\mathrm{d}\zeta_A}{\mathrm{d}u_2} \right. \\ \left. + \zeta_A \frac{\mathrm{d}\zeta_I}{\mathrm{d}u_2} + \zeta_A \zeta_I \frac{\alpha+1}{u_2}\right) x_A - k_2 \left(\zeta_I + u_2 \frac{\mathrm{d}\zeta_I}{\mathrm{d}u_2}\right) x_I \\ 0 \\ k_2 \left(\zeta_I + u_2 \frac{\mathrm{d}\zeta_I}{\mathrm{d}u_2}\right) x_I + k_2 \frac{k_1}{k_{ml}} u_2^\alpha \left(\zeta_A \zeta_I (\alpha + 1) \right. \\ \left. + \zeta_I u_2 \frac{\mathrm{d}\zeta_A}{\mathrm{d}u_2} + \zeta_A u_2 \frac{\mathrm{d}\zeta_I}{\mathrm{d}u_2}\right) x_A \end{pmatrix}$$

*A space χ is said to be compact if every open cover $\mathscr{O}$ of χ contains a finite subcollection that also covers χ