HIGH-INDEX DAE SYSTEMS IN MODELING AND CONTROL OF CHEMICAL PROCESSES

Aditya Kumar and Prodromos Daoutidis

*University of Minnesota, Department of Chemical Engineering and Materials Science
Minneapolis, MN 55455, USA*

Abstract: Examples of chemical processes are presented that are modeled by high-index DAEs under quasi-steady-state assumptions of phase, thermal, reaction equilibrium etc. While the high index in these models can be avoided by replacing the equilibrium assumptions with rate expressions, the resulting models are not suitable for controller design. This is illustrated through simulations in two reactor examples.

Keywords: singular systems, fast-rate processes, equilibrium-based models

1. INTRODUCTION

The dynamics of many chemical processes are naturally described by coupled differential and algebraic equations (DAEs), where the differential equations arise from standard dynamic balances of mass, energy and momentum, and the algebraic equations include thermodynamic relations, empirical correlations and quasi-steady-state conditions. Often, the algebraic equations in the DAE models can be readily eliminated to obtain equivalent models comprised of ordinary differential equations (ODEs). However, there is a wide variety of chemical processes where the algebraic equations in the DAE models are singular in nature, implying the presence of a high index.

DAE systems with a high index are different from ODE systems, and there has been extensive research on the analysis of fundamental system-theoretic properties like solvability, stability etc. (Bajic, 1992; Dolezal, 1986; Rheinboldt, 1984) and the numerical simulation (Brenan *et al.*, 1989; Hairer *et al.*, 1989) of such systems. The majority of the research on the control of DAE systems had focused on linear systems (see e.g., (Campbell, 1982; Dai, 1989) and the references therein). More recently, the control of nonlinear high-index DAE systems has also been studied, where the controller design is addressed on the basis of suitable state-space realizations of the DAE system (Krishnan and McClamroch, 1994*b*; Kumar and Daoutidis, 1995) or a feedback modified system (Kumar and Daoutidis, 1996).

In this article, a few representative examples of chemical processes that are modeled by high-index DAEs, are presented. In these examples, the high index arises due to quasi-steady-state assumptions of phase, thermal, reaction and pressure equilibrium, corresponding to fast mass/heat transfer, fast reactions and fast vapor flow, respectively. The high index in these *equilibrium-based* models can be avoided by explicitly modeling the fast-rate phenomena, instead of employing the quasi-steady-state assumptions. However, such *rate-based* models of these fast-rate processes exhibit a stiffness/time-scale multiplicity, which makes them unsuitable for controller design purposes. More specifically, controllers designed on the basis of the rate-based models are highly ill-conditioned and may even lead to instability. On the other hand, controllers designed on the basis of the equilibrium-based models do not suffer from these problems. These control-relevant issues are illustrated through simulations in two examples.

2. PROCESSES MODELED BY HIGH-INDEX DAES

Chemical processes are naturally modeled by nonlinear DAE systems in the semi-explicit form:

$$\begin{aligned} \dot{x} &= f(x) + g(x)u + b(x)z \\ 0 &= k(x) + l(x)z \\ y_i &= h_i(x); \;\; i = 1, \ldots, m \end{aligned} \quad (1)$$

Financial support for this work from NSF, Grant CTS-9320402, is gratefully acknowledged.

where $x \in \mathbb{R}^n$ is the vector of differential variables, $z \in \mathbb{R}^p$ is the vector of algebraic variables, $u \in \mathbb{R}^m$ is the vector of manipulated inputs, y_i is the i-th output to be controlled, $f(x)$ and $k(x)$ are analytic vector fields of dimensions n and p, respectively, $b(x), g(x)$ and $l(x)$ are analytic matrices of appropriate dimensions, and $h_i(x)$ are analytic scalar functions. Clearly, the above DAE system has a high index $\nu_d > 1$ if the matrix $l(x)$ is singular. In what follows, some examples of chemical processes that are modeled by high-index DAEs in the form of Eq.1 are presented.

2.1. *Multi-phase processes with fast mass transfer*

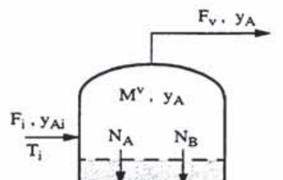

Fig. 1. A condenser

Consider the condenser in Fig. 1, where a gaseous mixture of two components A and B is fed at a molar flowrate F_i, composition y_{Ai} and temperature T_i. The mixture is cooled in the condenser to a temperature T, to facilitate a separation of the heavier component B into the liquid phase. If the inter-phase mass transfer between the liquid and vapor phases is fast, then the two phases can be assumed to be in equilibrium, and the following model of the process is derived:

$$\dot{M}^v = F_i - F_v - N_A - N_B \quad (2)$$

$$\dot{y}_A = \frac{1}{M^v}[F_i(y_{Ai} - y_A) - N_A(1 - y_A) + N_B y_A] \quad (3)$$

$$\dot{M}^l = N_A + N_B - F_l \quad (4)$$

$$\dot{x}_A = \frac{1}{M^l}[N_A(1 - x_A) - N_B x_A] \quad (5)$$

$$\dot{T} = \frac{1}{(M^l + M^v)c_p}[F_i c_p(T_i - T) - Q + (N_A + N_B)\Delta H^v] \quad (6)$$

$$0 = P_A^s x_A - P y_A \quad (7)$$

$$0 = P_B^s(1 - x_A) - P(1 - y_A) \quad (8)$$

$$0 = M^v RT - P(V_T - \frac{M^l}{\rho}) \quad (9)$$

where the dynamic material and energy balances yield the differential equations (Eq.2-6) for the molar holdups M^l, M^v and the mole fraction x_A, y_A of A, in the liquid and vapor phases, respectively, and the temperature T. The phase equilibrium relations (Eq.7,8) and the ideal gas equation for the vapor holdup (Eq.9) constitute the algebraic equations. It can be easily verified that the above DAE model is in the form of Eq.1 with the differential variables $x = [M^v, y_A, M^l, x_A, T]^T$ and the algebraic variables $z = [N_A, N_B, P]^T$. Furthermore, the algebraic equations (Eq.7-9) do not involve the inter-phase mole transfer rates N_A, N_B, and thus, the DAE model has a high index; it can be verified that the index is two. Such high-index DAE models arise in many processes, e.g. multi-phase reactors, distillation/absorption columns and reactive distillation columns, where the liquid/vapor phases are assumed to be in equilibrium.

2.2. *Reactors with fast and slow reactions*

Consider an isothermal CSTR, where a reactant A is fed at a volumetric flowrate F_i and concentration C_{Ai}, and the following first-order reactions occur in series $A \rightleftharpoons B$, $B \rightarrow C$. The reversible reaction $A \rightleftharpoons B$ is much faster than the irreversible one, and is thus essentially at equilibrium. Under this assumption of reaction equilibrium, the following model of the process is derived:

$$\dot{V} = F_i - F \quad (10)$$

$$\dot{C}_A = \frac{F_i}{V}(C_{Ai} - C_A) - R_A \quad (11)$$

$$\dot{C}_B = -\frac{F_i}{V}C_B + R_A - R_B \quad (12)$$

$$\dot{C}_C = -\frac{F_i}{V}C_C + R_B \quad (13)$$

$$0 = C_A - \frac{C_B}{K_{eq}} \quad (14)$$

$$0 = R_B - k_B C_B \quad (15)$$

where V is the liquid holdup in the reactor, and C_A, C_B, C_C are the molar concentrations of the corresponding components. The above DAE model is in the form of Eq.1 with the differential variables $x = [V, C_A, C_B, C_C]^T$, and the algebraic variables $z = [R_A, R_B]^T$, where R_A and R_B denote the reaction rates for the reversible and irreversible reactions, respectively. Clearly, the algebraic equations can not be solved for the reaction rate R_A for the fast reversible reaction. The reaction equilibrium relation in Eq.14 has to be differentiated once, to obtain a solution for R_A, and thus, the DAE model has an index two. Often in many chemical reactors, multiple reactions occur simultaneously, of which some are much faster than the others. The above example illustrates that in such processes, the assumption of reaction equilibrium for fast reversible reactions, and similarly the assumption of complete conversion for fast irreversible reactions, naturally leads to high-index DAE models.

2.3. *Staged processes with negligible pressure drop*

Consider the cascade of two gas phase CSTRs in Fig. 2. Gaseous reactant A is fed to the first reactor at a molar flowrate F_A and temperature T_A,

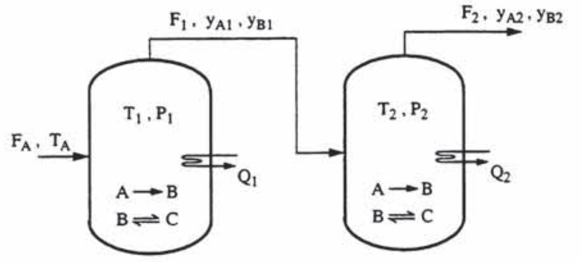

Fig. 2. A cascade of two gas-phase reactors

where the exothermic reactions $A \rightarrow B$, $B \rightleftharpoons C$ occur in series. The irreversible reaction $A \rightarrow B$ is quite slow, and thus, the first reactor is operated at a high temperature T_1 to promote the conversion of A. However, at a high temperature T_1, the fast reversible reaction $B \rightleftharpoons C$ is essentially at equilibrium and the production of C is restricted by this reaction equilibrium limitation. The gaseous mixture from the first reactor is fed to the second reactor, which is operated at a lower temperature T_2 to shift the reaction equilibrium in the forward direction and favor the production of C. The product stream from the second reactor is withdrawn at a molar flowrate F_2. On the other hand, the molar flowrate F_1 of the gaseous stream from the first to the second reactor is governed by the pressure drop $\Delta P = P_1 - P_2$ between the two reactors:

$$F_1 = (\frac{P_1}{RT_1})\frac{(\Delta P)^{4/7}}{\sigma} \tag{16}$$

where P_1/RT_1 is the molar density of the vapor. At a high operating pressure P_1, this density is high, and consequently a small pressure drop yields a high molar flowrate. Thus, the pressure drop between the reactors is essentially negligible.

Under the assumption of negligible pressure drop, and reaction equilibrium for the fast reversible reaction $B \rightleftharpoons C$, the following DAE model of the process is derived:

$$\begin{aligned}
\dot{M}_1 &= F_A - F_1 \\
\dot{y}_{A1} &= \frac{1}{M_1}[F_A(1-y_{A1}) - R_{B,1}] \\
\dot{y}_{B1} &= \frac{1}{M_1}[-F_A y_{B1} + R_{B,1} - R_{C,1}] \\
\dot{T}_1 &= \frac{1}{M_1 c_p}[F_A c_p(T_A - T_1) - R_{B,1}\Delta H_{R1} \\
&\qquad -R_{C,1}\Delta H_{R2} - Q_1] \\
\dot{M}_2 &= F_1 - F_2 \\
\dot{y}_{A2} &= \frac{1}{M_2}[F_1(y_{A1} - y_{A2}) - R_{B,2}] \\
\dot{y}_{B2} &= \frac{1}{M_2}[F_1(y_{B1} - y_{B2}) + R_{B,2} - R_{C,2}] \\
\dot{T}_2 &= \frac{1}{M_2 c_p}[F_1 c_p(T_1 - T_2) - R_{B,2}\Delta H_{R1} \\
&\qquad -R_{C,2}\Delta H_{R2} - Q_2] \\
0 &= R_{B,1} - k_{1,o}\exp(-E_1/RT_1)M_1 y_{A1} \\
0 &= y_{B,1} - \frac{y_{C,1}}{K_{eq}(T_1)} \\
0 &= R_{B,2} - k_{1,o}\exp(-E_1/RT_2)M_2 y_{A2} \\
0 &= y_{B,2} - \frac{y_{C,2}}{K_{eq}(T_2)} \\
0 &= M_1 RT_1 - P_1 V_{1T} \\
0 &= M_2 RT_2 - P_2 V_{2T} \\
0 &= P_1 - P_2
\end{aligned} \tag{17}$$

which is in the form of Eq.1 with the differential variables $x = [M_1, y_{A1}, y_{B1}, T_1, M_2, y_{A2}, y_{B2}, T_2]^T$ and the algebraic variables $z = [R_{B,1}, R_{C,1}, R_{B,2}, R_{C,2}, P_1, P_2, F_1]^T$. In the above model, M_i is the molar holdup, y_{Ai}, y_{Bi} and $y_{Ci} = 1 - y_{Ai} - y_{Bi}$ are the mole fractions of A, B and C, T_i is the temperature, P_i is the pressure, and $R_{B,i}, R_{C,i}$ are the rates of production of B and C through the irreversible and reversible reactions, respectively, in the first ($i = 1$) and second ($i = 2$) reactors.

As in the previous example, the algebraic equations do not involve the rates $R_{C,1}$ and $R_{C,2}$ for the fast reversible reaction at equilibrium. Moreover, the vapor flow rate F_1 also does not appear in any of the algebraic equations, due to the negligible pressure drop assumption. Thus, the above DAE model has a high index; it can be verified that the index is two. This example illustrates that in high-pressure staged processes or process networks with a fast inter-stage gaseous flow, e.g. high-pressure absorption/distillation and reactive distillation columns, the assumption of negligible pressure drop between the individual stages leads to high-index DAE models.

3. FEEDBACK CONTROL BASED ON EQUILIBRIUM- VS. RATE-BASED MODELS

A common feature in the DAE models of the examples discussed in the previous section, is that the fast mass transfer, reactions, gaseous flow, etc., are assumed to be at quasi-steady-state equilibrium conditions, which leads to the high index. If these equilibrium assumptions are relaxed, and explicit rate expressions for the fast phenomena are included instead, then the resulting rate-based DAE models have an index one and are easily reduced to ODE models. More specifically, the rate expressions for fast reactions, mass/heat transfer, etc., involve large parameters of the form $(1/\epsilon)$, e.g. large reaction rate and mass/heat transfer coefficients, and the corresponding rate-based models are given by ODE systems in the form:

$$\begin{aligned}
\dot{x} &= f(x) + g(x)u + \frac{1}{\epsilon}b(x)k(x) \\
y_i &= h_i(x);\ i = 1, \ldots, m
\end{aligned} \tag{18}$$

where ϵ is a small positive parameter, and $f(x)$ and $k(x)$ are analytic vector fields of dimensions n and p, respectively ($p < n$).

Owing to the presence of the large parameter $(1/\epsilon)$, the system in Eq.18 exhibits a two-time-scale behavior, although it is not in the standard singularly perturbed form. The derivation of standard form singularly perturbed representations of such two-time-scale systems has been addressed in (Kumar *et al.*, 1996; Krishnan and McClamroch, 1994*a*) through a nonlinear, and possibly ϵ-dependent change of coordinates. While the numerical simulation of the rate-based ODE models in Eq.18 can be addressed through appropriate methods for stiff systems, such models are not suitable for controller synthesis. More specifically, standard inversion-based controllers designed on the basis of these models also involve the large process parameters, and thus, they are highly sensitive to even small modeling/measurement errors that are always present. Furthermore, such controllers may lead to instability even in the nominal system without any errors, if the fast phenomena lead to a slightly non-minimum phase behavior.

A standard approach to avoid these problems, is to design the controller on the basis of quasi-steady-state models that describe the slow dynamics of the process (Kokotovic *et al.*, 1986). For the fast-rate processes discussed previously, the fast dynamics corresponding to the fast reactions and heat/mass transfer are typically stable and can be ignored to obtain such models, which are precisely the high-index DAE models obtained under the quasi-steady-state equilibrium conditions in the previous section. The equilibrium-based models do not involve any of the large process parameters, and controllers designed on the basis of these models are well-conditioned and do not lead to instability in the presence of slightly non-minimum phase behavior.

Remark: In some cases, the fast dynamics are unstable and a high-gain controller is required to stabilize them. The high gain in such controllers is another large parameter of the form $(1/\epsilon)$ and the corresponding quasi-steady-state assumption may lead to a higher index (see (Kumar and Daoutidis, 1996) for an example).

3.1. *Control of reactor with multiple reactions*

Consider the example in section 2.2, where it is desired to control the liquid holdup $(y_1 = V)$ and the concentration of the product $B\,(y_2 = C_B)$, using the inlet and outlet flowrates as the manipulated inputs $(u_1{=}F,\ u_2{=}F_i)$. A rate-based model of the process is derived by including the rate expression:

$$R_A = k_A(C_A - \frac{C_B}{K_{eq}})$$

for the fast reversible reaction $A \rightleftharpoons B$, where the reaction rate coefficient k_A is much larger than the coefficient k_B for the slow reaction $B \rightarrow C$. Thus,

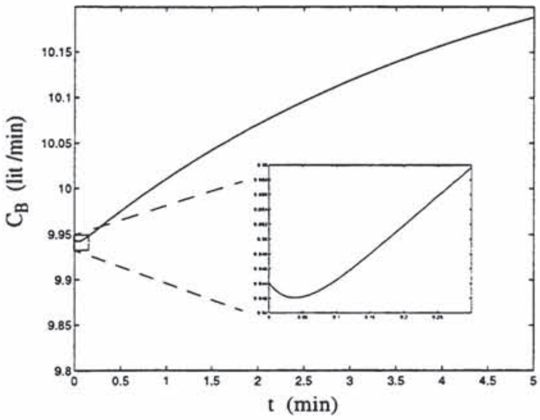

Fig. 3. Inverse response in C_B

defining the large parameter $k_A = 1/\epsilon$, the rate-based model of the form in Eq.18 is obtained with $n = 4$ and $p = 1$. The relative orders of the two outputs y_1 and y_2 with respect to the inputs u are $r_1{=}1$ and $r_2{=}1$, respectively, and an input/output linearizing controller was designed to enforce a first-order, decoupled input/output response in the closed-loop system. However, this controller leads to instability due to a slightly non-minimum phase behavior exhibited by the rate-based model (this is illustrated in Fig. 3, which shows an inverse response in y_2 in the initial boundary layer, for a step increase in u_2). On the other hand, the controller designed on the basis of the equilibrium-based index-two DAE model in section 2.2, yields excellent performance with stability. Fig. 4 shows the closed-loop profiles of y_2 and u_2 for a 10% increase in the setpoint y_{2sp}.

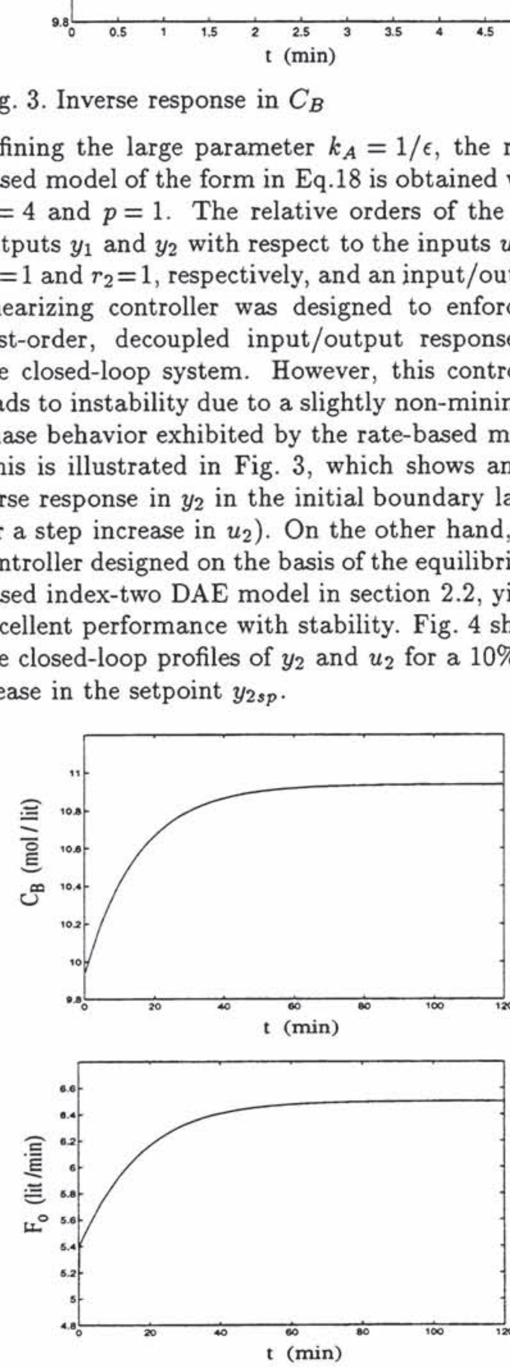

Fig. 4. Closed-loop performance of index-two DAE model-based controller

3.2. *Control of gas-phase reactor cascade*

Consider the cascade of reactors in section 2.3, where it is desired to control $y_1 = y_{A1}$ and $y_2 = y_{C2}$

using the manipulated inputs $u_1 = Q_1$ and $u_2 = Q_2$. A detailed, rate-based model of the process is derived by explicitly including the rate expression:

$$R_{C,i} = k_{2,o} \exp(-E_2/RT_i) M_i \left(y_{B,i} - \frac{y_{C,i}}{K_{eq}(T_i)}\right)$$

for the fast reversible reaction $B \rightleftharpoons C$ in the two reactors, and the pressure drop correlation in Eq.16. At the nominal operating temperatures T_1 and T_2 of the two reactors, the reaction rate coefficient $k_{2,o} \exp(-E_2/RT_i)$ and the parameter $(P_1/\sigma RT_1)$ in the pressure drop correlation are large. Defining the large parameter $k_{2,o} \exp(-E_2/RT_1) = 1/\epsilon$, the rate-based model in the form of Eq.18 is obtained with $n=8$ and $p=3$.

The performance of a controller designed on the basis of the rate-based model was compared with that of a controller designed on the basis of the equilibrium-based index-two DAE model in section 2.3. More specifically, on the basis of the rate-based model, the relative orders were found to be $r_1 = 2$ and $r_2 = 2$. Thus, a controller was designed to induce linear second-order, decoupled input/output responses in the closed-loop system. The relative orders on the basis of the equilibrium-based model were found to be $r_1=2$ and $r_2=1$, and a controller was designed to induce a second- and first-order linear response in the two outputs y_1 and y_2, respectively. Fig. 5 shows the closed-loop responses for a 10% decrease in y_{1sp} and a 10% increase in y_{2sp}, under the two controllers. Clearly, both controllers yield excellent performance in this nominal case without any modeling errors.

However, the controller designed on the basis of the rate-based ODE model explicitly involves the large process parameters and is thus highly sensitive to even small modeling errors. The performance of the controller was studied for the same setpoint changes, in the presence of a 5% error in $c_p, \Delta H_{r1}, \Delta H_{r2}$. The controller calculates very large control action and leads to instability. For the same modeling errors, the controller designed on the basis of the equilibrium-based model yields an excellent performance with stability. The corresponding profiles in Fig. 6 clearly show that the controller is well-conditioned and the calculated manipulated input profiles are close to those in the nominal case (see Fig. 5).

4. CONCLUSIONS

A wide variety of fast-rate processes with fast mass/heat transfer, fast reactions, etc., are naturally modeled by high-index DAEs under quasi-steady-state assumptions of phase, thermal, reaction and pressure equilibrium. For these processes, detailed rate-based models with explicit rate expressions for the fast phenomena are given by index-one DAEs or ODEs. Owing to the presence of large parameters in the rate expressions, the ODE models exhibit a stiffness/time-scale-multiplicity and are not suitable for controller design. On the other hand, the equilibrium-based high-index DAE models that describe the slow dynamics of the process, are well-suited for the design of well-conditioned controllers.

REFERENCES

Bajic, V. B. (1992). *Lyapunov's Direct Method in The Analysis of Singular Systems and Networks*. Shades Technical Publications. Hillcrest, Natal.

Brenan, K. E., S. L. Campbell and L. R. Petzold (1989). *Numerical Solution of Initial-Value Problems in Differential-Algebraic Equations*. Elsevier Science Pub. Co. Inc.. New York.

Campbell, S. L. (1982). *Singular Systems of Differential Equations II*. Vol. 61 of *Research Notes in Mathematics*. Pitman Books Ltd.. London.

Dai, L. (1989). *Singular Control Systems*. Vol. 118 of *Lecture Notes in Control and Information Sciences*. Springer-Verlag. Berlin, Heidelberg.

Dolezal, V. (1986). Generalized solutions of semistate equations and stability. *Circ. Syst. Sig. Proc.* **5**, 391–403.

Hairer, E., C. Lubich and M. Roche (1989). *The Numerical Solution of Differential-Algebraic Systems by Runge-Kutta Methods*. Vol. 1409 of *Lecture Notes in Mathematics*. Springer-Verlag. Berlin, Heidelberg.

Kokotovic, P. V., H. K. Khalil and J. O'Reilly (1986). *Singular Perturbations in Control: Analysis and Design*. Academic Press. London.

Krishnan, H. and N. H. McClamroch (1994*a*). On the connection between nonlinear differential-algebraic equations and singularly perturbed control systems in nonstandard form. *IEEE Trans. Automat. Contr.* **39**, 1079–1084.

Krishnan, H. and N. H. McClamroch (1994*b*). Tracking in nonlinear differential-algebraic control systems with applications to constrained robot systems. *Automatica* **30**, 1885–1897.

Kumar, A. and P. Daoutidis (1995). Feedback control of nonlinear differential-algebraic-equation systems. *AIChE J.* **41**, 619–636.

Kumar, A. and P. Daoutidis (1996). Dynamic feedback regularization and control of nonlinear differential-algebraic-equation systems. *AIChE J.* **42**, 2175–2198.

Kumar, A., P. D. Christofides and P. Daoutidis (1996). Singular perturbation modeling of nonlinear two-time-scale processes with non-explicit time-scale separation. In: *Proceedings of 13th IFAC World Congress*. San Francisco, CA, vol. M. pp. 1–6.

Rheinboldt, W. C. (1984). Differential-algebraic systems as differential equation on manifolds. *Math. Comput.* **43**, 473–482.

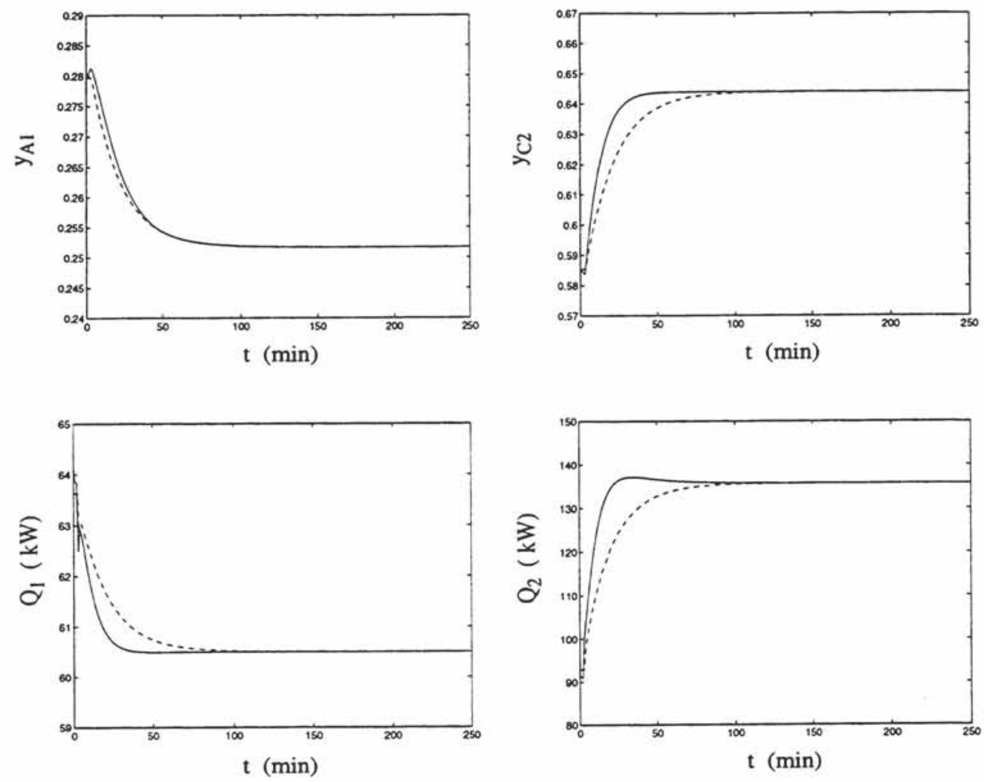

Fig. 5. Comparison of performances of the controllers based on the equilibrium-based (solid) and rate-based (dashed) models, in the nominal system

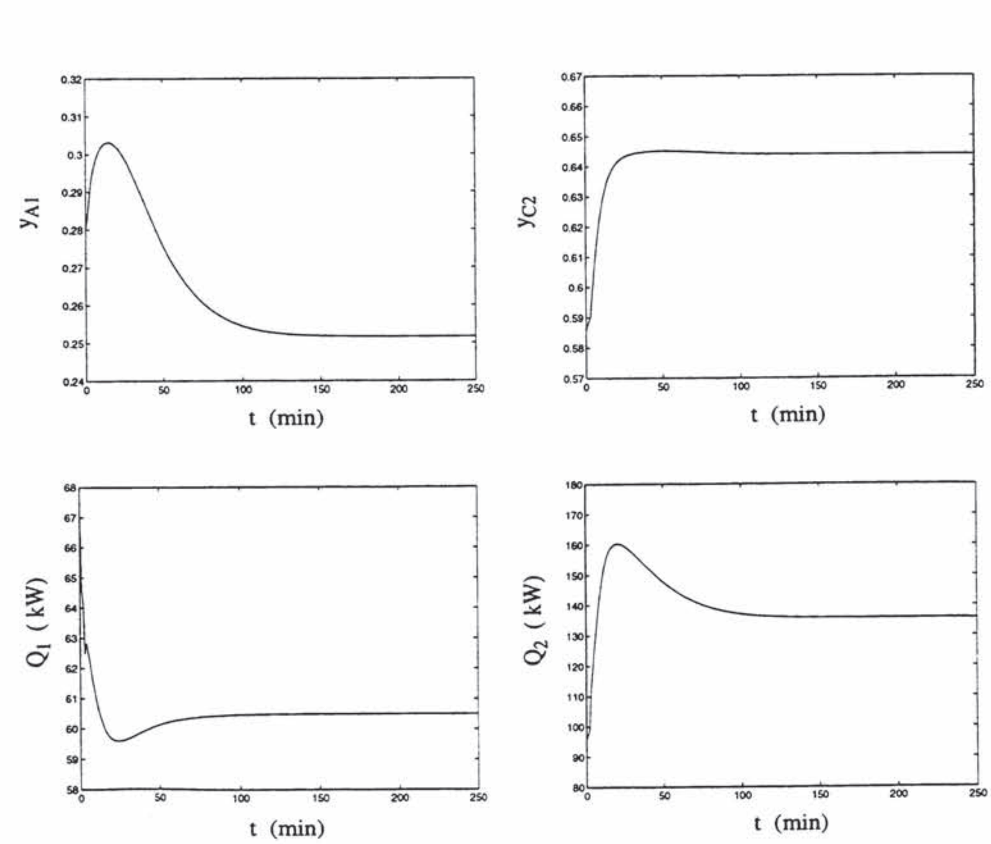

Fig. 6. Performance of the controller based on the equilibrium-based index-two DAE model, in the presence of modeling errors