Control of Continuous Copolymerization Reactors

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Continuous solution copolymerization is an important industrial process in the manufacture of commodity and engineering plastics. The addition of comonomers and solvent, and the rate of heat exchange must be simultaneously manipulated to maintain safety, operability, and the product quality adequately, yielding a process with nonlinear behavior, strong and asymmetric input–output multivariable coupling, and potential for open-loop instability and state multiplicity, as shown in earlier dynamics and control studies. Accordingly, the key control objectives of the copolymerization reactor are: the compensation of interaction, the preclusion of input multiplicity and the robustness (i.e., tolerance to modeling and tuning errors) of the controller. In principle, these control issues should be considered within a nonlinear setting. Otherwise, the reactor may have to be operated with a conversion that is conservatively below what can be handled by standard mixing and heat-exchange equipment. To assess the inherent control possibilities and limitations of a given copolymerization reactor, a methodology to address the control problem is proposed such that the nonlinearity, interaction, input multiplicity, and robustness issues are explicitly confronted. The result is a linear multivariable interaction compensator whose tuning can be done with notions and tools from conventional control. This method is tested with the copolymerization of vinyl acetate with methyl methacrylate, dissolved in ethyl acetate.

Introduction

Solution copolymerization is an important industrial process in the manufacture of commodity and engineering plastics. The copolymer composition, conversion and chain-length distribution determine the quality attributes of the product, among which are the amount of volatile organic components; the glass transition temperature; the rheology of the polymeric solution; and the thermodynamic homogeneity of the copolymer phase. The control of the production rate and of the product grade (i.e., product quality properties), requires the regulation of the rates of insertion of the comonomer molecules into the growing copolymer chain (Leiza et al., 1993), which can be realized by manipulating the addition of the comonomers, the solvent, the initiator, or combinations of them.

In a series of papers (Hamer et al., 1981; Teymour and Ray, 1989, 1992a,b), the open-loop (i.e., without control) dynamics of a continuous solution copolymerization reactor have been studied, showing phenomena that are typically nonlinear: steady-state multiplicity, unstable steady states, limit cycles, parametric sensitivity, and bifurcation. Adebekun and Schork (1989a,b) studied the open-loop dynamics and the control problem of a continuous homopolymerization reactor, and reported the possibility of input multiplicity (Balakotaiah and Luss, 1985): the closed-loop reactor can reach its nominal output with nonunique control inputs, implying that the product grade may not reach its nominal design value. This observation indicates that the input-multiplicity question should also be a central point in the study of the control problem of the solution copolymerization case. Along an industrially oriented line of work, Congalidis et al. (1989) addressed the multiinput multioutput control problem in a solution-copolymerization reactor, with the minimization of interaction and the attainment of robustness being the key control objectives, and the choice of input–output pairing being the key decision. In this case, input multiplicity was not reported. Based on practical and physical considerations, a set of candidate input–output pairings were considered and

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their interaction and robustness were assessed by means of the relative gain array criteria and the condition number evaluated from an input–output linear transfer function of the reactor. With these criteria, an input–output configuration was chosen and realized by means of a conventional-type control scheme. The authors pointed out that the attainable settling time of the regulated output was limited by the impossibility of making the temperature loop, which had to be tuned conservatively due to interactions between loops, faster.

From the aforementioned works of Hamer et al. (1981), Teymour and Ray (1989, 1992a,b), Adebekun and Schork (1989a,b), and Congalidis et al. (1989), one is led to conclude that the nonlinearity, the interaction, the input multiplicity, and the robustness of the controller are the fundamental issues that determine the nature of the reactor control problem and of its solution, and that, in principle, understanding them should make possible a control scheme for operating the reactor under less conservative process designs.

In this work, we propose a methodology to address the feedback control problem of a solution-copolymerization reactor such that the nonlinearity, interaction, input multiplicity and robustness issues are explicitly confronted. Within a nonlinear control framework, it is established how the kind of copolymerization kinetics, reactor equipment, operation condition, and feedback control scheme, as well as the choice of regulated outputs determine the existence or nonexistence of input multiplicity, the kind and degree of the interaction and robustness problems, and the attainable settling times of the production rate and product grade. First, we address the problem as a nonlinear state-feedback control problem (i.e., assuming that the reactor states are known), yielding a test for the existence of a perfect interaction compensator that is robust and achieves linear, noninteractive, pole-assignable output dynamics with closed-loop state stability (i.e., without input multiplicity), as well as the structure of the underlying nonlinear state-feedback controller. This identifies the source and nature of the interaction, robustness, and input-multiplicity problems, as well as the possibility of handling them by feedback control. Then, with a practical implementation in mind, we present a procedure for assessing the functioning of a linear output-feedback (driven only by measured outputs) approximation or the rigorous nonlinear state-feedback controller. In particular, the construction of the linear output-feedback controller can be done with a rather simplified model, and its tuning resorts to notions and tools from conventional control, like the ones employed in industrial practice. Finally, these findings and their associated methodology are applied to the control of the continuous copolymerization of vinyl acetate with methyl methacrylate, dissolved in ethyl acetate (Hamer et al., 1981).

Copolymerization Reactor and Its Control Problem

Let us consider a continuous reactor where a solution copolymerization reaction takes place. Two monomers, solvent, and initiator are fed to the tank. The reactor is represented in Figure 1. The free-radical copolymerization is induced by radicals generated from the decay of the initiator. The reaction is strongly exothermic, heat being removed by means of a cooling jacket. If there is a significant gel effect, the conversion of comonomers into polymer is accompanied

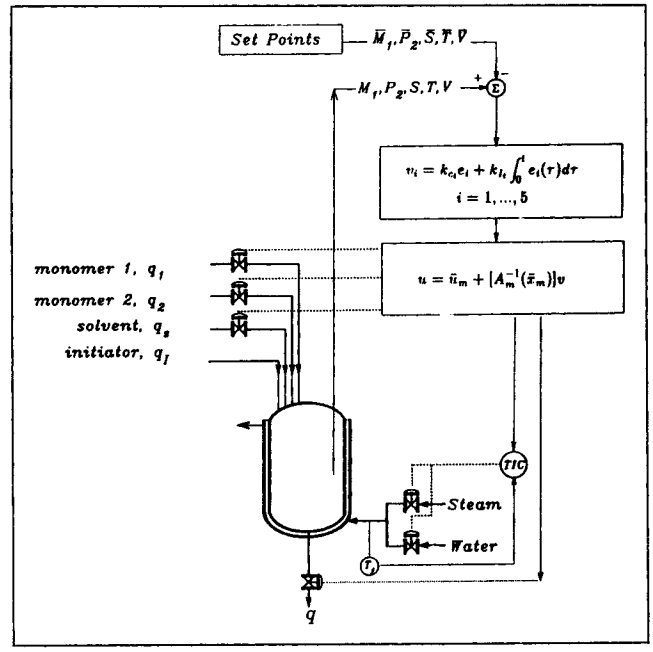

Figure 1. Copolymerization reactor and its linear output-feedback controller.

by a considerable increase in viscosity, which decreases the heat-removal capability. From standard kinetics, reaction engineering (Hamer et al., 1981), and viscous heat-exchange (Alvarez et al., 1990) modeling considerations, the reactor dynamics is given by the following set of nonlinear ordinary differential equations (see notation in Appendix A):

$$\dot{M}_1 = -R_1 - M_1(\epsilon_1 R_1 + \epsilon_2 R_2) + \frac{q_1}{V}(M_{1_e} - \epsilon_{m_1} M_1) - q_2 \frac{M_1}{V}\epsilon_{m_2} - q_s \frac{M_1}{V}\epsilon_s$$

$$\dot{M}_2 = -R_2 - M_2(\epsilon_1 R_1 + \epsilon_2 R_2) + \frac{q_2}{V}(M_{2_e} - \epsilon_{m_2} M_2) - q_1 \frac{M_2}{V}\epsilon_{m_1} - q_s \frac{M_2}{V}\epsilon_s$$

$$\dot{I} = -R_I - I(\epsilon_1 R_1 + \epsilon_2 R_2) + q_I \frac{I_e}{V} - q_1 \frac{I}{V}\epsilon_{m_1} - q_2 \frac{I}{V}\epsilon_{m_2} - q_s \frac{I}{V}\epsilon_s$$

$$\dot{S} = -S(\epsilon_1 R_1 + \epsilon_2 R_2) + \frac{q_s}{V}(S_e - \epsilon_s S) - q_1 \frac{S}{V}\epsilon_{m_1} - q_2 \frac{S}{V}\epsilon_{m_1}$$

$$\dot{P}_1 = R_1 - P_1(\epsilon_1 R_1 + \epsilon_2 R_2) - q_1 \frac{P_1}{V}\epsilon_{m_1} - q_2 \frac{P_1}{V}\epsilon_{m_2} - q_s \frac{P_1}{V}\epsilon_s$$

$$\dot{P}_2 = R_1 - P_2(\epsilon_1 R_1 + \epsilon_2 R_2) - q_1 \frac{P_2}{V}\epsilon_{m_1} - q_2 \frac{P_2}{V}\epsilon_{m_2} - q_s \frac{P_2}{V}\epsilon_s$$

$$\dot{T} = q_1\Omega_1 + q_2\Omega_2 + q_s\Omega_S - \gamma(T - T_j) + \beta_1 + \beta_2$$

$$\dot{V} = q_1\epsilon_{m_1} + q_2\epsilon_{m_2} + q_s\epsilon_s - q + V(\epsilon_1 R_1 + \epsilon_2 R_2),$$

where the effect of density changes with conversion has been accounted for by the contraction factors ϵ_1, ϵ_2 and the functions Ω_1, Ω_2, Ω_s. If no contraction is considered $\epsilon_1 = \epsilon_2 = 0$ and ϵ_{m_1}, ϵ_{m_2}, and ϵ_s become the expressions of the usual copolymerization model (Hamer et al., 1981). There are eight states in the reactor: $x_1 = M_1$ (concentration of monomer 1), $x_2 = M_2$ (concentration of monomer 2), $x_3 = I$ (concentration of initiator), $x_4 = S$ (concentration of solvent), $x_5 = P_1$ (concentration of converted monomer 1), $x_6 = P_2$ (concentration of converted monomer 2), $x_7 = T$ (reactor temperature), and $x_8 = V$ (volume). The control (manipulated) inputs are $u_1 = q_1$ (feed rate of monomer 1), $u_2 = q_2$ (feed rate of monomer 2), $u_3 = q_s$ (feed rate of solvent), $u_4 = T_c$ (coolant temperature), and $u_5 = q$ (exit flow rate). There are five control (regulated) outputs, three of which are specified: $y_3 = S$ (concentration of solvent), $y_4 = T$ (temperature), $y_5 = V$ (volume), and two are to be determined: $y_1 = h_1(x)$ and $y_2 = h_2(x)$.

R_I, R_1, R_2, β, and γ are smooth (infinitely differentiable) scalar-valued functions (see appendix) that correspond to rates of initiator decomposition, conversion of monomer 1, conversion of monomer 2, ratio of heat production to heat capacity, and ratio of heat exchange to heat capacity, respectively. R_1 and R_2 account for the autoacceleration phenomenon due to the gel effect (Hamer et al., 1981) and γ depends on the mixture viscosity, which in turn depends, via a free volume-type functionality (Alvarez et al., 1990), on the monomer conversion.

From a practical operation point of view, the following variables are important production and product-grade attributes:

$$P_r = M_1^w R_1 + M_2^w R_2$$

$$X_{ov} = (P_1 + P_2)/(M_1 + M_2 + P_1 + P_2)$$

$$C_I = R_1/(R_1 + R_2)$$

$$PM_w = \mu_2/\mu_1,$$

where

$$\mu_i = \sum_{k=1}^{\infty} \sum_{l=1}^{\infty} (k^i M_1^w + l^i M_2^w) D_{k,l}, \qquad i = 1, 2,$$

P_r is the production rate (kg/h); X_{ov} is the overall (mole basis) conversion; C_I is the instantaneous copolymer composition of monomer 1; PM_w is an average copolymer molecular weight based on the chain-length distribution, as defined in Congalidis et al. (1989); $D_{k,l}$ is the molar concentration (number of molecules/Avogadro's number) of copolymer chains with k and l units of monomers 1 and 2, respectively; M_i^w is the molecular weight of the ith monomer; and μ_1 and μ_2 are first and second moments of the copolymer chain-length distribution. The evaluation of PM_w requires the consideration of two additional differential equations for μ_1 and μ_2.

In compact notation, the preceding reactor control system can be written as follows:

$$\dot{x} = f(x) + G(x)u, \qquad y = h(x) x \in X, \qquad u \in U, \qquad y \in Y, \tag{1}$$

where $f(x)$, $h(x)$, and $g_i(x)$ are vector fields on some neighborhood X of the design nominal steady-state $\bar{x}$, and $G(x) = [g_1(x), \ldots, g_m(x)]$ is a matrix field. Associated to $\bar{x}$, there is a nominal input $\bar{u}$ and a nominal output $\bar{y}$ that satisfy the static version of Eq. 1:

$$0 = f(\bar{x}) + G(\bar{x})\bar{u}, \; \bar{y} = h(\bar{x}).$$

In open-loop operation (i.e., Eq. 1 with $u = \bar{u}$), the nominal steady-state $\bar{x}$ can be asymptotically (A) stable or unstable, unique or nonunique, or the center of a limit cycle (Hamer et al., 1981).

The on-line measurements of T and V can be done with standard instrumentation, and the compositions S, M_1, M_2, P_1, and P_2 can be determined from on-line measurements of density, refractive index, boiling point, and viscosity in conjunction (possibly using an observer) with mass balances and suitable density, refractive index, viscosity, and thermodynamics expressions derived from first principles or from correlations fitted to experimental data, as has been done in previous state-inference studies in polymerization reactors (Kozub and MacGregor, 1992). Moreover, due to the fact that the residence settling time (i.e., four times the residence time) of the continuous solution copolymerization reactors are much larger than the delay period of on-line chromatographic composition measurement instruments, the sampling theorem (Stephanopoulos, 1984) in conjunction with standard filtering-prediction schemes in signal processing (Papoulis, 1965) can yield good continuous-instantaneous estimates of the reactor compositions. These observations are consistent with the formulation of the reactor control problem in Congalidis et al. (1989).

Reactor control problem

Keeping in mind that we want to obtain an industrial-type (i.e., application-oriented) control scheme, the study of the control problem of the continuous free-radical copolymerization reactor in the present work is conducted under the following assumptions and requirements:

1. Temperature T, volume V, solvent concentration S, and two concentrations (to be determined) of the set (M_1, M_2, P_1, P_2) are available for feedback control.
2. The resulting control scheme must be a robust linear output-feedback controller.
3. Regulation of the nominal production rate and product grade must be accomplished indirectly, with fast recovery speeds.
4. Tuning of the multivariable (five-input, five-output) controller must be translated into the tuning of five closed-loop, low-order, noninteractive, linear single-input, single-output (SISO) systems (one for each output), and carried out in terms of ideas and vocabulary from (linear) conventional industrial control.

As mentioned in the introduction, the key problems that must be confronted in the design of any control scheme for the continuous copolymerization reactor are: (1) the potential for input multiplicity (Adebekun and Schork, 1989a), whose presence one expects to be favored by an operating regime with gel effect; (2) interaction conflict that limits the speed of recovery of the product grade (Congalidis et al.,

1989); and (3) lack of robustness, depending on the choice of inputs and outputs (Congalidis et al., 1989).

The present work is organized as follows. In the next section, a geometric control framework is set to address the problems of interaction and input multiplicity as parts of a nonlinear state-feedback problem. Within a suitable nonlinear state-feedback control scheme, we show in the fourth section how the selection of the regulated outputs determines (1) the degree of potential closed-loop interaction conflict and of tolerance to modeling errors; (2) a restricted plant system whose stability precludes the closed-loop input multiplicity; and (3) the attainable closed-loop settling time, which in turn determines the settling time of the production rate and of the product grade. At the end of that section, the assessment of the solvability and structure of the nonlinear state-feedback control problem (assuming all the states are known) lead to the design of an output-feedback controller (using only the output measurements). Finally, in the fifth section, we apply the aforementioned results to the solution copolymerization system studied in Hamer et al. (1981) and Congalidis et al. (1989).

Nonlinear Control Framework

In this section, we present the nonlinear geometric framework that will be used in the following section to address the reactor control problem. From Isidori's (1989) and Nijmeijer and Van der Schaft's (1990) textbooks we recall the statement (definition 1) and the solvability (theorem 1) of the nonlinear local state-feedback problem in geometric control theory, showing the possibility of a perfect interaction compensator. Then the need for a nonlinear local framework is motivated, leading to conditions (corollary 1) to preclude the possibility of closed-loop input multiplicity.

Definition 1. Let $\bar{x}$ be the (possibly open-loop unstable) nominal steady state of plant (Eq. 1). Its state-feedback problem is (locally) solvable if there exists a nonlinear controller

$$u = \mu(x) \tag{2}$$

that, applied to the plant, yields a closed-loop system

$$\dot{x} = f(x) + G(x)\mu(x), \qquad y = h(x), \tag{3}$$

where

The nominal steady-state $\bar{x}$ is a locally asymptotically (LA) stable steady state.

The output dynamics is linear, noninteractive, and pole assignable (LNPA).

In the following theorem, necessary and sufficient solvability conditions for the preceding problem are given. To state the theorem, some notation and definitions must be introduced. The directional derivative $L_f c$ of a scalar field $c(x)$ along the vector field $f(x)$, and its recursion formula, are given by

$$L_f^{i+1}c = L_f\left(L_f^i c\right), \quad L_f^0 c = c, \qquad L_f c = \sum_{i=1}^{n} \frac{\partial c}{\partial x_i} f_i.$$

From this definition, a set $\{\kappa_1, \ldots, \kappa_m\}$ of m positive integers determines the vectors

$$\phi_I(x) = \left[h_1, \ldots, L_f^{\kappa_1 - 1}h_1; \ldots; h_m, \ldots, L_f^{\kappa_m - 1}h_m\right]^T,$$

$$\dim \phi_I = \kappa = \kappa_1 + \cdots + \kappa_m \tag{4}$$

$$b(x) = [L_f^{\kappa_1}h_1, \ldots, L_f^{\kappa_m}h_m]^T, \qquad \dim b = m, \tag{5}$$

the decoupling matrix

$$A(x) = \mathrm{mat}\left[L_{g_j}L_f^{\kappa_i}h_i(x)\right], \qquad \dim A(x) = \kappa \times \kappa \tag{6}$$

and, provided $A(\bar{x})$ is nonsingular, the $(n-\kappa)$-dimensional zero-dynamics

$$\dot{x} = f(x) - G(x)A^{-1}(x)b(x), \qquad x \in X_{zI} \tag{7a}$$

$$X_{zI} = \{x \in X_{zI} | \phi_I(x) = \phi_I(\bar{x})\}, \qquad \dim X_{zI} = n - \kappa. \tag{7b}$$

Theorem 1. The state-feedback problem of plant (Eq. 1) is locally solvable if and only if there are m positive integers $\kappa_1, \ldots, \kappa_m$ such that

(i) $\kappa_1 + \ldots + \kappa_m = \kappa \le n$

(ii) $(\partial\phi_i/\partial u)_{(\bar{x})} = 0$

(iii) $|A(\bar{x})| \neq 0$

(iv) $\bar{x}$ is an LA-stable steady state of the zero-dynamics (Eq. 7).

If the maps $f(x)$, $G(x)$, and $h(x)$ of plant (Eq. 1) satisfy the conditions of the last theorem, the nonlinear controller (Eq. 2) is given by

$$\mu(x) = A(x)^{-1}\{K_c[\phi_I(x) - \phi_I(\bar{x})] - b(x)\}, \tag{8}$$

where K_c is a block-diagonal (bd) matrix with κ nonzero adjustable entries:

$$K_{c(n\times m)} = \mathrm{bd}[k_1, \ldots, k_m], \; k_{i(1\times\kappa_i)} = [k_{i1}, \ldots, k_{i\kappa_i}].$$

With this controller, the closed-loop plant (Eq. 3) has the following LNPA output dynamics:

$$\epsilon_i^{(\kappa_i)} - k_{i\kappa_i}\epsilon_i^{(\kappa_1 - 1)} - \cdots - k_{i1}\epsilon_i = 0,$$

$$\epsilon_i = y_i(t) - \bar{y}_i(t), \qquad 1 \le i \le m. \tag{9}$$

This equation says that the dynamics of each output is linear, of order κ_i, and independent of the other outputs. With a suitable choice of gains $k_{i1}, \ldots, k_{i\kappa_i}$, the κ_i poles (eigenvalues) of the ith output loop can be arbitrarily placed in the stable side of the complex plane. Thus, controller (Eq. 8) is a perfect nonlinear interaction compensator, and the structure of the matrix $A(x)$ characterizes the kind and degree of inherent input–output interaction of the plant. If the initial state x_0 of the closed-loop plant (Eq. 3) is in the manifold X_{zI} (Eq. 7b), the closed-loop dynamics (Eq. 3) of the plant coincides with its stable zero-dynamics (Eq. 7), the plant stays in X_{zI} and moves toward its nominal steady-state $\bar{x}$ (see Eq. 7a, and curve 1 in Figure 2) with the output y fixed at its nominal value $\bar{y}$ and the input u evolving toward its nominal value $\bar{u}$. If the initial steady state is not in the zero-dynamics manifold X_{zI} (see curve 2 in Figure 2), the nonlinear controller steers, with adjustable speed, the plant toward the hypersurface X_{zI}. Simultaneously, the zero dynamics steers the

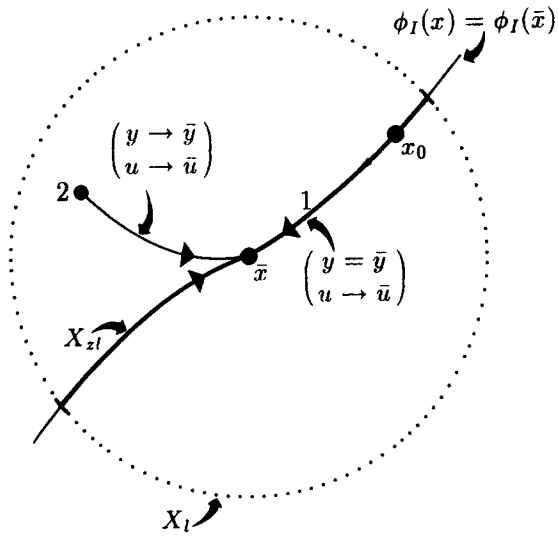

Figure 2. Reactor closed-loop dynamics in the neighborhood of the steady-state $\bar{x}$.

plant toward $\bar{x}$. As a result, the plant reaches asymptotically its nominal state $\bar{x}$.

As it stands, Theorem 1 guarantees the existence of some neighborhood $X_l \subset \mathbb{R}^n$ (Figure 2), (of undefined size and shape, not necessarily small) of the nominal steady-state $\bar{x}$ where the nonlinear controller works. However, as illustrated in Figure 3, the nominal steady-state $\bar{x}$ may not be unique in a given set $X \subset \mathbb{R}^n$. The nominal state $\bar{x}$ is LA-stable (see trajectories 1 and 2) in the neighborhood X_l, but some motions (see trajectory 4) born outside X_l do not settle at the nominal steady-state $\bar{x}$. In spite of reaching the prescribed nominal output $\bar{y}$, the closed-loop trajectory 4 reaches a state-input pair $(\bar{x}^*, \bar{u}^*)$ that is not the nominal design pair $(\bar{x}, \bar{u})$. In chemical engineering terminology (Luss and Balakotaiah, 1985), the comparison of the closed-loop input–output behaviors of trajectories 3 and 4 of Figure 3 show that the system has input multiplicity. In our nonlinear control framework, this input multiplicity is due to the fact that the nominal state $\bar{x}$ is not a unique attractor of the zero-dynamics in the manifold X_z of Figure 3. To address the preclusion of the closed-loop input-state multiplicity of

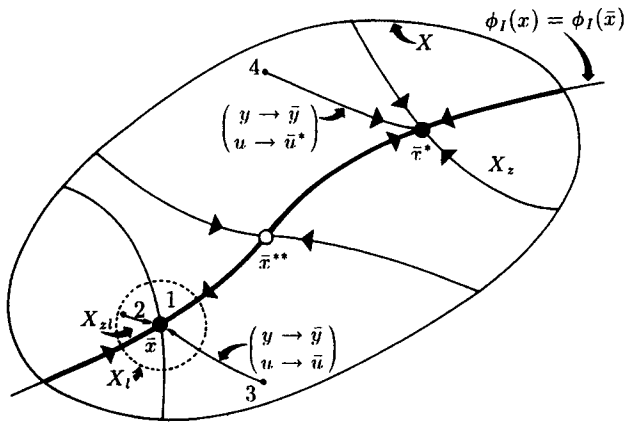

Figure 3. Reactor closed-loop dynamics in the state set X.

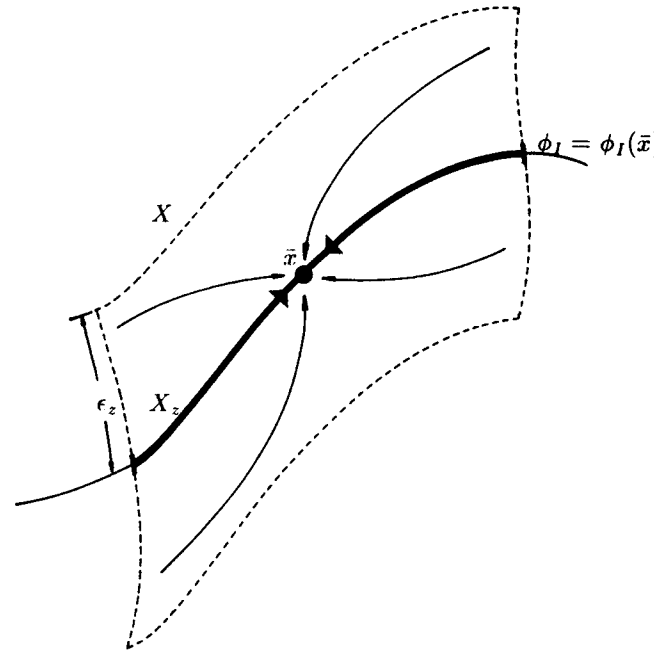

Figure 4. Reactor closed-loop dynamics in the neighborhood $N(X_z)$ of the zero-dynamics manifold X_z.

Figure 3, let us introduce the following variation of Theorem 1.

Corollary 1. Let the state-feedback problem of plant (Eq. 1) be locally solvable at the nominal steady-state $\bar{x}$, and let X_z be a given open, connected, invariant manifold of the $(n - \kappa)$-dimensional hypersurface $\phi_I(x) = \phi_I(\bar{x})$ (see Figure 4). Then there exists a strip-type neighborhood (see Figure 4)

$$N(X_z) = \{x \in X \mid \|\phi(x) - \phi(\bar{x})\| < \epsilon_z, \epsilon_z > 0\},$$

where $\bar{x}$ is a unique A-stable steady-state of the closed-loop system (Eq. 3) with LNPA output dynamics, if and only if

(i) Conditions (i), (ii), and (iii) of Theorem 1 hold at each point of the manifold X_z.

(ii) There exists a complementary map, $\phi_{II}(x) = [\phi_{\kappa+1}(x), \ldots, \phi_n(x)]^T$, such that the map $\phi(x) = [\phi_I^T(x), \phi_{II}^T(x)]^T$ is one-to-one on X_z.

(iii) The nominal steady-state $\bar{x}$ is a unique A-stable steady state of zero-dynamics (Eq. 7).

It must be kept in mind that (1) the dimensions, the shape, and the stability of the zero-dynamics manifold depend on the choice of the outputs of the control scheme; (2) the rate of approximation of the state x to the zero-dynamics manifold X_z (see Figure 4) can be modified by adjusting the gains of the nonlinear controller (Eq. 8), and (3) the evolution of the closed-loop plant along the zero-dynamics manifold cannot be modified by adjusting the control gains.

Approach to the Reactor Control Problem

State-feedback control

According to the statement of the problem in the second section and the nonlinear control framework of the last sec-

tion, our copolymerization reactor control problem consists of finding out for which output pair (h_1, h_2) the solvability conditions of corollary 1 are met with the reactor maps $f(x)$, $G(x)$, and the output maps $h_3(x) = x_4$ (solvent), $h_4(x) = x_7$ (temperature), and $h_5(x) = x_8$ (volume). First, to endow this mathematical problem with physical meaning, the solvability conditions of Corollary 1 will be specialized (in Proposition 1) to our reactor problem.

From mass and energy conservation principles and the second law of thermodynamics, the state x of the reactor evolves in the following invariant manifold X of $\mathbb{R}^8$:

$$X = X_1 \times \cdots \times X_8 \subset \mathbb{R}^8,$$

where x_i, $i = 1, \ldots, 8$ are open and bounded intervals in the real line

$$X_1 = (0, M_{1_e}), \quad X_2 = (0, M_{2_e}), \quad X_3 = (0, I_e),$$
$$X_4 = (0, S_e) \quad X_5 = (0, M_{1_e}/(1-\epsilon_1)),$$
$$X_6 = (0, M_{2_e}/(1-\epsilon_2)), \quad X_7 = (T_*, T^*), \quad X_8 = (0, V^*)$$

and

$$T_* = \min(T_{e*}, T_{j*}), \quad T^* = T_{ec}^* + \Delta H^* M/(V\rho^* C_p),$$
$$T_{ec}^* = \max(T_e^*, T_j^*), \quad \Delta H^* = \max(\Delta H_1, \Delta H_2),$$
$$\rho^* = \min[M_{1_e} M_1^w/(1-\epsilon_1), \quad M_{2_e} M_2^w/(1-\epsilon_2)].$$

The lower and upper bounds of the inlet and coolant temperatures (T_{e*}, T_e^*, T_{ec*}, and T_{ec}^*) are regarded as fixed design specifications. Thus, any physically meaningful reactor geometry must be circumscribed to the open and connected state-set X. Correspondingly, for a given pair (h_1, h_2) of output maps, the associated zero-dynamics invariant manifold X_{zl} (Eq. 7b) is given by

$$X_z = \{x \in X \mid \phi_I(x) = \phi_I(\bar{x})\}, \quad \dim \phi_I = 3 + \kappa_1 + \kappa_2 \le 8, \tag{10}$$

where

$$\phi_I(x) = \left[h_1(x), \ldots, L_f^{\kappa_1 - 1} h_1(x), h_2(x), \ldots, L_f^{\kappa_2 - 1} h_2(x), x_4, x_7, x_8\right]^T, \quad \kappa_1 + \kappa_2 \le 5, \quad \kappa_1, \kappa_2 \ge 1.$$

Proposition 1. The reactor control problem is solvable if and only if there are two positive integers, κ_1 and κ_2, such that the output maps $h_1(x)$ and $h_2(x)$ meet the following conditions:

(i) The decoupling matrix ($\kappa_1 + \kappa_2 \le 5$, $\kappa = 3 + \kappa_1 + \kappa_2$)

$$A(x)_{(\kappa,5)} = \begin{pmatrix} L_{g1} L_f^{\kappa_1 - 1} h_1 & L_{g2} L_f^{\kappa_1 - 1} h_1 & L_{g3} L_f^{\kappa_1 - 1} h_1 & 0 & 0 \\ L_{g1} L_f^{\kappa_2 - 1} h_2 & L_{g2} L_f^{\kappa_2 - 1} h_2 & L_{g3} L_f^{\kappa_2 - 1} h_2 & 0 & 0 \\ -SV^{-1}\epsilon_{m_1} & -SV^{-1}\epsilon_{m_2} & (S_e - S\epsilon_s)V^{-1} & 0 & 0 \\ \Omega_1 & \Omega_2 & \Omega_S & \gamma & 0 \\ \epsilon_{m_1} & \epsilon_{m_2} & \epsilon_s & 0 & -1 \end{pmatrix} \tag{11}$$

is nonsingular in the zero-dynamics manifold X_z (Eq. 10).

(ii) There is a complementary map, $\phi_{II}(x) = [\phi_{\kappa+1}(x), \ldots, \phi_n(x)]^T$, such that the map

$$\phi(x) = \begin{pmatrix} \phi_I(x) \\ \phi_{II}(x) \end{pmatrix}$$

is invertible on some strip neighborhood $N(X_z)$ (see Figure 4) of the zero-dynamics manifold X_z (Eq. 10).

(iii) The nominal steady-state $\bar{x}$ is the unique A-stable attractor of the $[8-(\kappa_1+\kappa_2+3)]$-dimensional zero dynamics

$$\dot{x} = f(x) - G(x)A(x)^{-1} b(x), \quad x \in X_z \tag{12}$$

where the five entries of $b(x)$ are

$$b_1(x) = L_f^{\kappa_1} h_1$$
$$b_2(x) = L_f^{\kappa_2} h_2$$
$$b_3(x) = -S(\epsilon_1 R_1 + \epsilon_2 R_2)$$
$$b_4(x) = \beta_1 + \beta_2 - \gamma T$$
$$b_5(x) = V(\epsilon_1 R_1 + \epsilon_2 R_2).$$

If the output pair (h_1, h_2) meets the conditions of the last proposition, the nonlinear controller (8) is given by

$$u = [q_1, q_2, q_s, T_c, q]^T = A(x)^{-1}\{K_c[\phi_I(x) - \phi_I(\bar{x})] - b(x)\}, \quad x \in N_z \tag{13}$$

$$K_c = bd[k_1, k_2, k_3, k_4, k_5] \tag{14}$$
$$k_1 = [k_{11}, \ldots, k_{1\kappa_1}], \quad k_2 = [k_{21}, \ldots, k_{2\kappa_2}].$$

The corresponding LNPA output closed-loop dynamics (Eq. 9) is

$$(y_i - \bar{y}_i)^{\kappa_i} - k_{i\kappa_i}(y_i - \bar{y}_i)^{\kappa_i - 1} - \cdots - k_{i1}(y_i - \bar{y}_i) = 0, \quad i = 1, 2, \quad \kappa_1 + \kappa_2 \le 5 \tag{15a}$$

$$(y_3 - \bar{y}_3)^{(1)} - k_{31}(y_3 - \bar{y}_3) = 0, \text{ (solvent loop)} \tag{15b}$$

$$(y_4 - \bar{y}_4)^{(1)} - k_{41}(y_4 - \bar{y}_4) = 0, \text{ (temperature loop)} \tag{15c}$$

$$(y_5 - \bar{y}_5)^{(1)} - k_{51}(y_5 - \bar{y}_5) = 0, \text{ (volume loop).} \tag{15d}$$

As mentioned in the preceding section, controller 13 is a perfect interaction compensator. The information of the inherent open-loop interaction structure is contained in the decoupling matrix $A(x)$ (Eq. 11). If the model available for the control was perfect, the criteria to choose the output pair (h_1, h_2) would be to look for two positive integers, κ_1 and κ_2, such that $\kappa_1 + \kappa_2$ is as large as possible, meaning that the dimension of the zero-dynamics manifold (Eq. 10) is made as small as possible to reduce or rule out ($\kappa_1 + \kappa_2 = 5$) the potential of input multiplicity. If $\kappa_i > 1$ ($i = 1$ or 2), at least one of the polymerization rates, say $R_i(x)$, and its gradient $dR_i(x)$ appear in the decoupling matrix $A(x)$. In a practical control situation, the model available to design the control scheme is imperfect, meaning that approximations $A_m(\chi)$ of $A(\chi)$ and $b_m(\chi)$ of $b(\chi)$ are to be used (χ is a suitable estimate of the state). If the on-line control scheme is to be of an industrial type, a constant approximation, say, $A_m(\bar{x}_m)$ and $b_m(\bar{x}_m)$, may have to be used ($\bar{x}_m$ is the nominal state obtained from an approximated model). Knowing that the strong nonlinear input–output coupling of a copolymerization reactor can limit its closed-loop speed of response (Congalidis et al., 1989), one should avoid the presence of the polymerization rate functions in the decoupling matrix $A(x)$ to prevent potential closed-loop output interaction problems due to the magnification of errors when obtaining the inverse approximation, $A_m^{-1}(\chi)$ or $A_m^{-1}(\bar{x}_m)$, of the decoupling matrix $A(x)$, or equivalently, $\kappa_1 + \kappa_2 \geq 2$ should be as small as possible. This observation brings to light a trade-off between dynamic performance and robustness. As the relative degrees (κ_1 and κ_2) of the output map pair (h_1, h_2) are increased, the controllable (steerable) part of the reactor increases its dimension (yielding a faster settling time of product properties), the zero dynamics reduce their dimension (reducing the potential of input multiplicity), and there is a growth in the potential of instability or degradation of response due to modeling errors. The determination of the adequate compromise depends on the specific copolymerization system, the specific reactor, and on whether the reactor operates at mild or severe conditions (i.e., with or without significant gel effect).

In view of the interaction and robustness (Congalidis et al., 1989), and closed-loop stability (Adebekun and Schork, 1989a,b) problems associated with the continuous solution polymerization reactors, as well as of the assumptions and specifications in the statement of our reactor control problem, we shall focus on studying the four possible linear output pairs

$$(y_1,y_2) = (P_1,P_2),(M_1,M_2),(M_2,P_1),(M_1,P_2); \quad \kappa_1 = \kappa_2 = 1$$

that can be made from the set (M_1, M_2, P_1, P_2) of the reactor concentrations.

Case I: $(y_1, y_2) = (P_1, P_2)$. At first sight, the choice of regulating the compositions P_1 and P_2 of converted monomers seems justified for regulating the overall conversion and the copolymer composition. However, after testing condition (i) of Proposition 1, one concludes that the decoupling matrix $A(x)$ is singular, meaning that the state-feedback control problem does not have a solution because the reactor is not output controllable.

Case II: $(y_1, y_2) = (M_1, M_2)$. In this case, the decoupling matrix $A(x)$ (see Appendix B) is nonsingular,

$$|A(x)| = \frac{\gamma}{V^3}[M_{1_e}(\epsilon_s M_{2_e} S + \epsilon_{m_2} M_2 S_e) + M_{2_e} S_e(\epsilon_{m_1} M_1 - M_{1_e})] \neq 0,$$

and the zero dynamics are given by

$$\begin{pmatrix} \dot{I} \\ \dot{P}_1 \\ \dot{P}_2 \end{pmatrix} = \begin{pmatrix} -I(\epsilon_1 R_1 + \epsilon_2 R_2) - R_I + q_I I_e V^{-1} \\ -P_1(\epsilon_1 R_1 + \epsilon_2 R_2) + R_1 \\ -P_2(\epsilon_1 R_1 + \epsilon_2 R_2) + R_2 \end{pmatrix} + \begin{pmatrix} \epsilon_{m_1} I & \epsilon_{m_2} I & \epsilon_s I & 0 & 0 \\ \epsilon_{m_1} P_1 & \epsilon_{m_2} P_1 & \epsilon_s P_1 & 0 & 0 \\ \epsilon_{m_1} P_2 & \epsilon_{m_2} P_2 & \epsilon_s P_2 & 0 & 0 \end{pmatrix} \times V^{-1} A(x)^{-1} b(x)_{x=(\bar{M}_1,\bar{M}_2,I,\bar{S},P_1,P_2,\bar{T},\bar{V})^T}. \quad (16)$$

Case III: $(y_1, y_2) = (M_2, P_1)$. Here the decoupling matrix $A(x)$ is nonsingular (see Appendix B):

$$|A(x)| = -\frac{\gamma}{V^3}\epsilon_{m_1} M_{2_e} S_e P_1 \neq 0,$$

and the zero dynamics are

$$\begin{pmatrix} \dot{I} \\ \dot{M}_1 \\ \dot{P}_2 \end{pmatrix} = \begin{pmatrix} -I(\epsilon_1 R_1 + \epsilon_2 R_2) - R_I + q_I I_e V^{-1} \\ -M_1(\epsilon_1 R_1 + \epsilon_2 R_2) - R_1 \\ -P_2(\epsilon_1 R_1 + \epsilon_2 R_2) + R_2 \end{pmatrix} + \begin{pmatrix} \epsilon_{m_1} I & \epsilon_{m_2} I & \epsilon_s I & 0 & 0 \\ -M_{1_e} + \epsilon_{m_1} M_1 & \epsilon_{m_2} M_1 & \epsilon_s M_1 & 0 & 0 \\ \epsilon_{m_1} P_2 & \epsilon_{m_2} P_2 & \epsilon_s P_2 & 0 & 0 \end{pmatrix} \times V^{-1} A(x)^{-1} b(x)_{x=(M_1,\bar{M}_2,I,\bar{S},\bar{P}_1,P_2,\bar{T},\bar{V})^T}. \quad (17)$$

Case IV: $(y_1, y_2) = (M_1, P_2)$. The decoupling matrix $A(x)$ is nonsingular (see Appendix B):

$$|A(x)| = \frac{\gamma}{V^3}\epsilon_{m_2} M_{1_e} S_e P_2 \neq 0,$$

and the zero dynamics is

$$\begin{pmatrix} \dot{I} \\ \dot{M}_2 \\ \dot{P}_1 \end{pmatrix} = \begin{pmatrix} -I(\epsilon_1 R_1 + \epsilon_2 R_2) - R_I + q_I I_e V^{-1} \\ -M_2(\epsilon_1 R_1 + \epsilon_2 R_2) - R_2 \\ -P_1(\epsilon_1 R_1 + \epsilon_2 R_2) + R_1 \end{pmatrix} + \begin{pmatrix} \epsilon_{m_1} I & \epsilon_{m_2} I & \epsilon_s I & 0 & 0 \\ \epsilon_{m_1} M_2 & -M_{2_e} + \epsilon_{m_2} M_2 & \epsilon_s M_2 & 0 & 0 \\ \epsilon_{m_1} P_1 & \epsilon_{m_2} P_1 & \epsilon_s P_1 & 0 & 0 \end{pmatrix} \times V^{-1} A(x)^{-1} b(x)_{x=(\bar{M}_1,M_2,I,\bar{S},P_1,\bar{P}_2,\bar{T},\bar{V})^T}. \quad (18)$$

In Case I, there is no solution to the state-feedback problem because the reactor is not output-controllable. In cases II, III and IV, the output-controllability conditions, (i) and (ii), of proposition 1 are met, and the corresponding zero dynamics were identified. As expected from our low-interaction and robustness requirements (i.e., $\kappa_1 = \cdots = \kappa_5 = 1$), in all cases the decoupling matrix depends on the concentrations M_1, M_2, S, P_1, P_2, the volume V, the temperature T, the heat transfer function $\gamma(x)$, and not on the copolymerization rates $R_1(x)$ and $R_2(x)$. Provided the closed-loop instability (i.e., the input multiplicity) is ruled out, the preceding key observation must be regarded as a preamble that opens the possibility of handling the interaction compensation problem in the presence of inexorable modeling errors, as well as of errors associated with the implementation and tuning of the nonlinear state-feedback controller.

If the zero dynamics of cases II, III and IV happened to be unstable, or equivalently, to yield input multiplicity, output maps, h_1 and h_2, with higher relative degrees should be considered, and the solvability of the corresponding state-feedback problem should be tested according to Proposition 1. As mentioned before, using h_1 and h_2 with higher relative degrees may overcome the input-multiplicity problem at the cost of its inclination for stronger interaction and poorer tolerance to modeling errors, which in turn may lead to a redesign of the equipment and/or of the operation condition, in order to have a milder process (say, with more solvent and less gel effect). The consideration of this case goes beyond the scope of the present work. Here, it suffices to say that the general methodological approach is the same.

To handle modeling errors, the state-feedback controller (Eq. 13) will be equipped with integral action. If cases II, III or IV (i.e., with $\kappa_i = 1$, $i = 1, \ldots, 5$) meets the closed-loop stability condition (iii) of Proposition 1, the nonlinear controller (Eq. 8) and the LNPA output dynamics (Eq. 14) can be written as follows:

$$u = \varphi(x,v) = A^{-1}(x)[v - b(x)] \tag{19a}$$

$$v = K_c(y - \bar{y}) + K_I \int_0^t (y - \bar{y})(\tau)\, d\tau \tag{19b}$$

$$(y_i - \bar{y}_i)^{(2)} - k_{i1}(y_i - \bar{y}_i)^{(1)} - k_{Ii}(y_i - \bar{y}_i) = 0, \qquad 1 \le i \le 5. \tag{19c}$$

Without restricting the approach, let us consider output dynamics with an underdamped response (i.e., with two complex conjugate eigenvalues). By doing so, the condition number (i.e., the ratio of the modulus of the two eigenvalues) of each output error dynamics is set to unity, minimizing the effect of modeling errors. In terms of (1.98%)-settling times τ_i (i.e., the time for a second-order system to settle within 1.98% of the final minus initial values) and damping factors ξ_i, the gains of Eq. 19 are given by (Stephanopoulos, 1982)

$$k_{i1} = -\frac{8}{\tau_i}, \quad k_{Ii} = -\frac{16}{(\tau_i \xi_i)^2}. \tag{19d}$$

Linear output-feedback control

Once the solvability of the reactor state-feedback control problem has been established by testing the output controllability (i and ii) and closed-loop stability (iii) conditions of Proposition 1, the estimator-based nonlinear control design of Alvarez (1996) can be applied to obtain a dynamic nonlinear output-feedback controller with stability criteria and a tuning procedure. To comply with the application-oriented requirements of the statement of our reactor control problem, instead of building an estimator-based controller, let us consider a simpler output-feedback controller: the linear output-feedback approximation of the rigorous nonlinear state-feedback controller with integral action (Eq. 19). The viability of such a controller is backed by the low interaction and robustness features already endowed to the nonlinear state-feedback reactor design (Eq. 19) of the preceding subsection.

In terms of a Taylor-series expansion about the nominal operation (i.e., $x = \bar{x}$, and $v = \bar{v} = 0$) the nonlinear state-feedback controller (Eq. 19a) can be expressed as follows:

$$u = \bar{u} + [A^{-1}(\bar{x})]v + \frac{\partial}{\partial x}[A^{-1}(x)b(x)]_{x=\bar{x}}(x - \bar{x}) + O(v, x - \bar{x}), \tag{20}$$

where $O(v, x - \bar{x})$ denotes quadratic and higher order terms. As pointed out before, with $\kappa_1 = \kappa_2 = 1$ the decoupling matrix $A(x)$ does not depend on the model-sensitive polymerization rates, and therefore one can expect that the essential information of the perfect nonlinear compensator (Eq. 13) is retained by the constant approximation $A_m(\bar{x}_m)$ of $A(x)$. For a given output $\bar{y}$, the solution of the steady-state version of the model-based closed-loop system (Eq. 3)

$$0 = f_m\{\bar{x}_m, \varphi(\bar{x}_m, 0)\}, \qquad \bar{y} = h_m(\bar{x}_m)$$
$$0 = \varphi\{\bar{x}_m, 0\} - \bar{u}_m$$

yields the approximation $(\bar{u}_m, \bar{x}_m)$ of the nominal input-state pair $(\bar{u}, \bar{x})$. Thus, the first-order truncation

$$u = \bar{u}_m + A_m^{-1}(\bar{x}_m)v, \quad v = K_c(y - \bar{y}) + K_I \int_0^t (y - \bar{y})(\tau)\, d\tau$$
$$K_c = \mathrm{diag}[k_{11}, \ldots, k_{51}], \qquad K_I = \mathrm{diag}[k_{I1}, \ldots, k_{I5}] \tag{21}$$

of the nonlinear controller (Eq. 19) seems a reasonable candidate for output-feedback linear controller. Following well-known arguments in control practice, the integral action is meant to eliminate the steady-state offset and to endow tolerance of the control scheme to modeling errors due to the overlooking, in controller (Eq. 21), of the linear error term in $x - \bar{x}$, and of the higher order terms in $x - \bar{x}$ and v, as well as to parametric and functional errors in the approximations $A_m(\bar{x}_m)$ and $\bar{u}_m$. A formal consideration of the key stability and robustness issues associated with the candidate linear output-feedback-approximated controller (Eq. 21) goes beyond the scope of the present work. Thus, we will limit ourselves to applying the linear output-feedback control scheme (Eq. 21) to the specific case example of the next section, and to test it with simulations.

The zero-dynamics associated with the candidate output-feedback controller (Eq. 21) are

$$\dot{x} = f(x,\bar{u}), \qquad x \in \{x \in \mathbb{R}^8 | \phi_I(x) = \phi_I(\bar{x})\}. \tag{22}$$

Observe that these zero dynamics are a particularization of the rigorous zero-dynamics (Eqs. 16, 17, or 18), when the nonlinear map $\varphi^{-1}(x, v)$ in Eq. 19a is replaced by its first-order approximated truncation (Eq. 21).

Application to the Copolymerization of VAM-MA (EA-Solvent, AIBN-Initiator)

In this section, we consider a particular copolymerization reactor to illustrate the application of the proposed control design technique. For this purpose, let us recall the solution copolymerization reactor of Congalidis et al. (1989), but with some modifications: no transfer agent and inhibitor are fed to the tank; there is no recycle of solvent and monomers; the reactor volume is doubled; and the massic conversion of the monomers is increased (from 20% to 45%) 2.25 times, the gel effect on both the copolymerization kinetics (Hamer et al., 1981) and the heat-transfer coefficient (Alvarez et al., 1990) are accounted for. As we shall see, this reactor and its operating condition retain the essence of the control problem, and are drastic in the sense that, due to the gel effect, the kinetics autoacceleration and its corresponding heat-removal limitation lead to an open-loop unstable reactor. By doing so, the proposed control scheme will be subjected to a severe test.

Nominal operation

As mentioned in the introduction, the copolymerization system is MMA (monomer 1), VA (monomer 2), EA (solvent), and AIBN (initiator). The functionalities and the parameters of the copolymerization kinetics were taken from Congalidis et al. (1989), and the functionalities of the viscosity and the heat-transfer coefficient were adapted from Alvarez et al. (1990). To look at the response of the copolymer average molecular weight PM_w, the reactor model was run with the two additional differential equations for the moments μ_1 and μ_2 of the chain-length distribution.

As indicated by Congalidis et al. (1989), their operating condition was such that the gel effect was not active, the heat-exchange coefficient was independent of the fraction of the solids (i.e., mass fraction of the copolymer), and the open-loop reactor had a unique and stable steady state. In their control study, a monomer feed rate $\bar{q}_1 + \bar{q}_2 = 108$ kg/h ($\bar{q}_1 = 18$ kg/h, $\bar{q}_2 = 90$ kg/h) was processed in a $\bar{V} = 1$ m^3 reactor at $\bar{T} = 353$ K, $\bar{C}_I$ (mass fraction of copolymer) = 0.56, yielding $\bar{P}_r$ (production rate) = 23 kg/h, and PM_w (molecular weight) = 35,000. To have a more difficult operating condition with a more severe control problem, let us redesign the preceding nominal operation as follows. The production rate is approximately doubled, and the copolymer composition of the more reactive monomer is halved, while the molecular weight and the mass fraction of the solvent are maintained. This is $\bar{P}_r = 52$ kg/h; $\bar{C}_I = 0.318$; $\overline{PM}_w = 36{,}688$; and $\bar{C}_s = 24.4$. For this to happen, the fraction of solids $\bar{C}_s$ must be (approximately) doubled, favoring the possibility of having the gel effect, autoaccelerated kinetics, and viscosity increase, and the heat-transfer coefficient decrease, as well as open-loop instability.

Based on the idea of the aforementioned redesign, the following nominal operating condition was obtained from the reactor model. Pure monomers and solvent at 300 K are fed to the reactor, and the initiator is diluted in a stream of solvent. The nominal control input vector is

$$\begin{aligned}\bar{u} &= \left[\bar{q}_1, \bar{q}_2, \bar{q}_s, \bar{T}_c, \bar{q}\right]^T \\ &= [1.9263\times10^{-2}\ \text{m}^3/\text{h},\ 9.6315\times10^{-2}\ \text{m}^3/\text{h}, \\ &\quad 3.8526\times10^{-2}\text{m}^3/\text{h},\ 330.0\ \text{K},\ 1.4263\times10^{-1}\ \text{m}^3/\text{h}]^T,\end{aligned}$$

the nominal steady state is

$$\begin{aligned}\bar{x} &= \left[\bar{M}_1, \bar{M}_2, \bar{I}, \bar{S}, \bar{P}_1, \bar{P}_2, \bar{T}, \bar{V}\right]^T \\ &= [0.0119\ \text{kmol/m}^3,\ 4.6737\ \text{kmol/m}^3,\ 0.0019\ \text{kmol/m}^3, \\ &\quad 2.7623\ \text{kmol/m}^3,\ 1.2347\ \text{kmol/m}^3,\ 2.6368\ \text{kmol/m}^3, \\ &\quad 334.92\ \text{K},\ 2.0\ \text{m}^3]^T,\end{aligned}$$

and the nominal production rate P_r of copolymer, the overall conversion X_{ov}, the composition of monomer 1 C_I, and the copolymer average molecular weight PM_w are

$$\bar{P}_r = 52\ \text{kg/h}, \quad \bar{X}_{ov} = 0.453, \quad \bar{C}_I = 0.319, \qquad PM_w = 36{,}688.$$

From standard bifurcation-stability numerical analysis (Hamer et al., 1981), it follows that the preceding nominal steady state is open-loop unstable and nonunique. Specifically, the open-loop reactor has at least five steady states, two of which being extinction and unwanted ignition attractors.

As mentioned in the second section, $\bar{P}_r$, $\bar{X}_{ov}$, $\bar{C}_I$, and $\overline{PM}_w$ are production- and product-grade specifications that the control scheme must regulate indirectly; and the kind, speed, and tolerance to the modeling errors of their recovery trajectories constitute an assessment of the performance of the control scheme. Thus, our control problem consists of finding out whether the nominal open-loop unstable reactor can be stabilized by means of feedback control, with interaction compensation, tolerance to modeling errors, closed-loop steady-state uniqueness, and responses of the production and grade variables that are faster than the ones in previous studies.

The previous section shows that the candidate configurations II, III and IV meet the output-controllability (i.e., conditions i and ii) property of proposition 1, and the stabilities of their zero dynamics must be assessed (in the next subsection) to decide on their input uniqueness. Then, for the configurations that produce input uniqueness we will investigate whether this property can be carried over to the reactor with the linear output-feedback approximation of the nonlinear state-feedback controller. Finally, in the subsection on closed-loop reactor dynamics, the related findings will be corroborated with numerical simulation tests.

A-priori assessment of closed-loop state stability and dynamics

With regard to the stability of the zero dynamics (or equivalently, of the closed-loop reactor under nonlinear state-feedback control) of cases II, III and IV, the results are shown in the top three bifurcation diagrams of Figure 5, where the overall conversion X_{ov} is plotted vs. the bifurcation parameter $\alpha = k_d/\bar{k}_d$, which is a factor that multiplies the value of the nominal preexponential factor $\bar{k}_d$ of the initiator decomposition constant. As can be seen in Figure 5, the zero dynamics of cases II and IV have unique and stable nominal steady states that correspond to the nominal one. Otherwise, in case III the zero dynamics do not have a unique steady state, and the one that corresponds to the nominal operation is unstable. The plot shows an unwanted extinction stable steady state, and no additional effort was made to determine the full set of steady states. Finally, case III is rejected because it means a closed-loop operation where the nominal steady state is unstable, meaning that the reactor cannot reach its nominal production and product-grade specifications. If a rigorous nonlinear state-feedback controller (Alvarez, 1996) is to be used, the control schemes of cases II and IV lead to a closed-loop operation where the nominal steady state is stable, implying that the reactor reaches its nominal production and product-grade specifications.

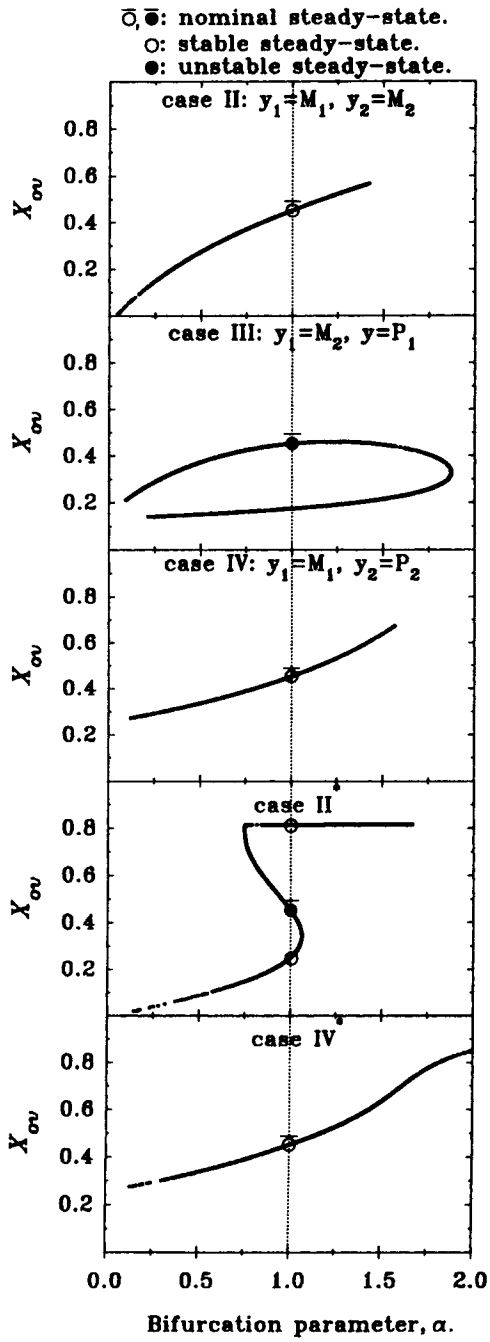

Figure 5. Multiplicity of the nominal zero-dynamics of cases II, III and IV with nonlinear state-feedback control; cases II* and IV* with linear output-feedback approximations of the state-feedback controller.

The two bottom plots of Figure 5 (cases II* and IV*) show the bifurcation diagrams of the modified zero dynamics (Eq. 22) that result when the nonlinear state-feedback controllers (Eq. 19) of cases II and IV are replaced by their linear output-feedback approximations (Eq. 21). As can be seen, in case IV* there is closed-loop stability with input-state uniqueness, but not in case II*. Hence, case IV* (i.e., $y_1 = M_1$ and $y_2 = P_2$) of the controller (Eq. 21) is an appropriate linear output-feedback controller for the reactor operating condition of the preceding subsection.

By construction, the nonlinear controller (Eq. 19) of case IV can steer, with adjustable speed, the reactor outputs (M_1, P_2, S, T, V) to their nominal values. The settling times of the variables P_r, X_{ov}, and C_I are limited by the convergence rate of the stable zero dynamics (Eq. 17), which in turn are limited by the residence settling time $\bar{\tau}_r$,

$$\bar{\tau}_r = 4\bar{\theta}, \qquad \bar{\theta} = \frac{\bar{V}}{\bar{q}}, \qquad \bar{q} = \bar{q}_1 + \bar{q}_2 + \bar{q}_s,$$

where $\bar{\theta}$ is the nominal residence time. The settling time of $\overline{PM}_w$ should be larger (slower), say $\approx 1.5\bar{\tau}_r$, as its evolution implies a cascade interconnection of the preceding dynamics with that of the chain-length distribution. These estimates of attainable response settling times are about three times faster than the ones reported in Congalidis et al. (1989), and must be regarded as the limit of what can be obtained with case IV* of the linear output-feedback controller (Eq. 21).

Closed-loop reactor dynamics

First, the reactor is operated with the nonlinear state-feedback controllers (Eq. 19) of cases II, III and IV, respectively. The gains (Eq. 19d) of the second-order closed-loop LNPA dynamics (Eq. 19c) were tuned according to the following settling times:

$$\tau_1 = \tau_2 = 0.224\bar{\tau}_r, \qquad \tau_3 = 0.335\bar{\tau}_r, \qquad \tau_4 = 0.056\bar{\tau}_r,$$

$$\tau_5 = 0.02225\bar{\tau}_r,$$

where $\bar{\theta} = 11$ h is the nominal reactor residence time. The proportionality constants in the preceding equations were

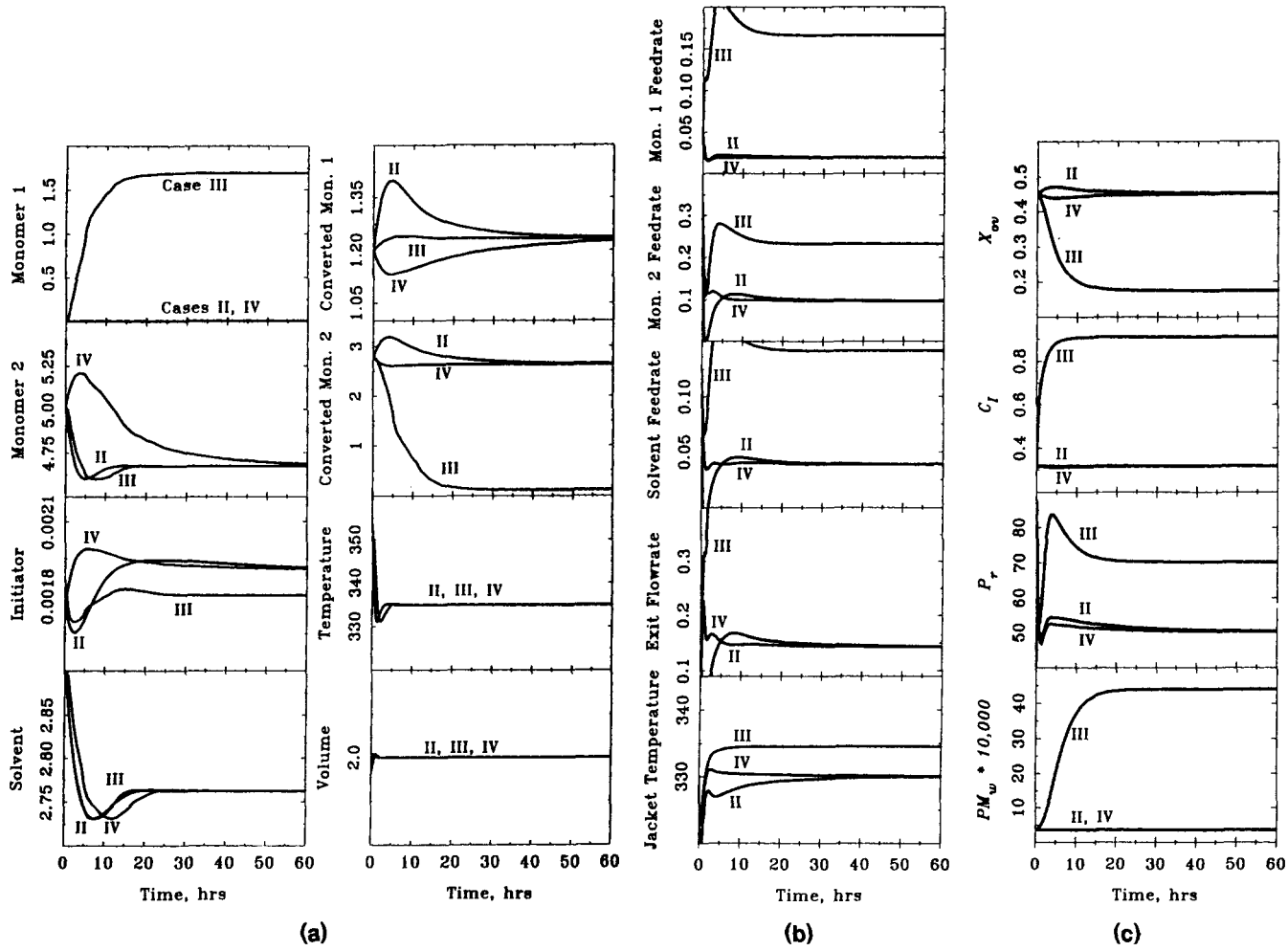

Figure 6. Responses of the closed-loop reactor with the nonlinear state-feedback control of cases II, III and IV with state-feedback control: (a) states (kmol/m^3, K, m^3); (b) control inputs (m^3/h, K); and (c) quality attributes.

tuned to be as small (fast) as possible, while producing reasonable control inputs. At the cost of a slower response, the control actions can be made arbitrarily smooth by making the proportionality constants sufficiently large. The initial reactor state is in the neighborhood of the nominal (open-loop unstable) steady state:

$$x(0) = [0.0121, 4.7906, 0.0019, 2.8313, 1.2655, 2.7027, 343.29, 2.05]^T \neq \bar{x}.$$

The corresponding state and input responses are shown in Figures 6a and 6b. As expected from the conclusions of the preceding subsection, control schemes II and IV (with stable zero dynamics; see Figure 5) steer the reactor to its nominal steady state, with the input reaching its nominal design value. Otherwise, control scheme III takes the reactor to its nominal output $[\bar{M}_2, \bar{P}_1, \bar{S}, \bar{T}, \bar{V}]$, but the state-input pair reaches an unwanted value (i.e., there is input multiplicity; see Figure 6b). This is consistent with Figure 5, from which we know that the nominal steady state is not an attractor of the zero dynamics of case III. The corresponding evolution of P_r, X_{ov}, C_I, and PM_w, are shown in Figure 6c. Observe that, in case III with input multiplicity, the reactor reaches erroneous values in its production rate and product grade. It must be pointed out that the response settling times of cases II and IV are consistent with the estimates drawn in the preceding subsection.

Next, the closed-loop reactor of cases II and IV is operated with the linear output-feedback approximation (Eq. 21) (cases II* and IV*) of the state-feedback controller (Eq. 19). The gains of the closed-loop design dynamics (Eq. 19c) were the ones of cases II and IV, and the corresponding responses are shown in Figure 7. As expected from Figure 5, in case IV* the reactor reaches its nominal operating condition, but not in case II*, where the reactor reaches an undesired extinction state. The outputs settle in their nominal values, but the production rate and product grade do not.

Case IV*-me of Figure 7 shows the responses of case IV* when the output-feedback controller (Eq. 21) is designed and built with modeling errors. For this reason, the following erroneous parameters were used to evaluate the nominal input $\bar{u}_m$ and the constant matrix $A_m(\bar{x}_m)$ of controller depicted in Eq. 21. The activation energy of the heat-producing propagation rates, the initiator efficiency, and the heat-transfer coefficient were underestimated by 15, 10 and 20%, respectively,

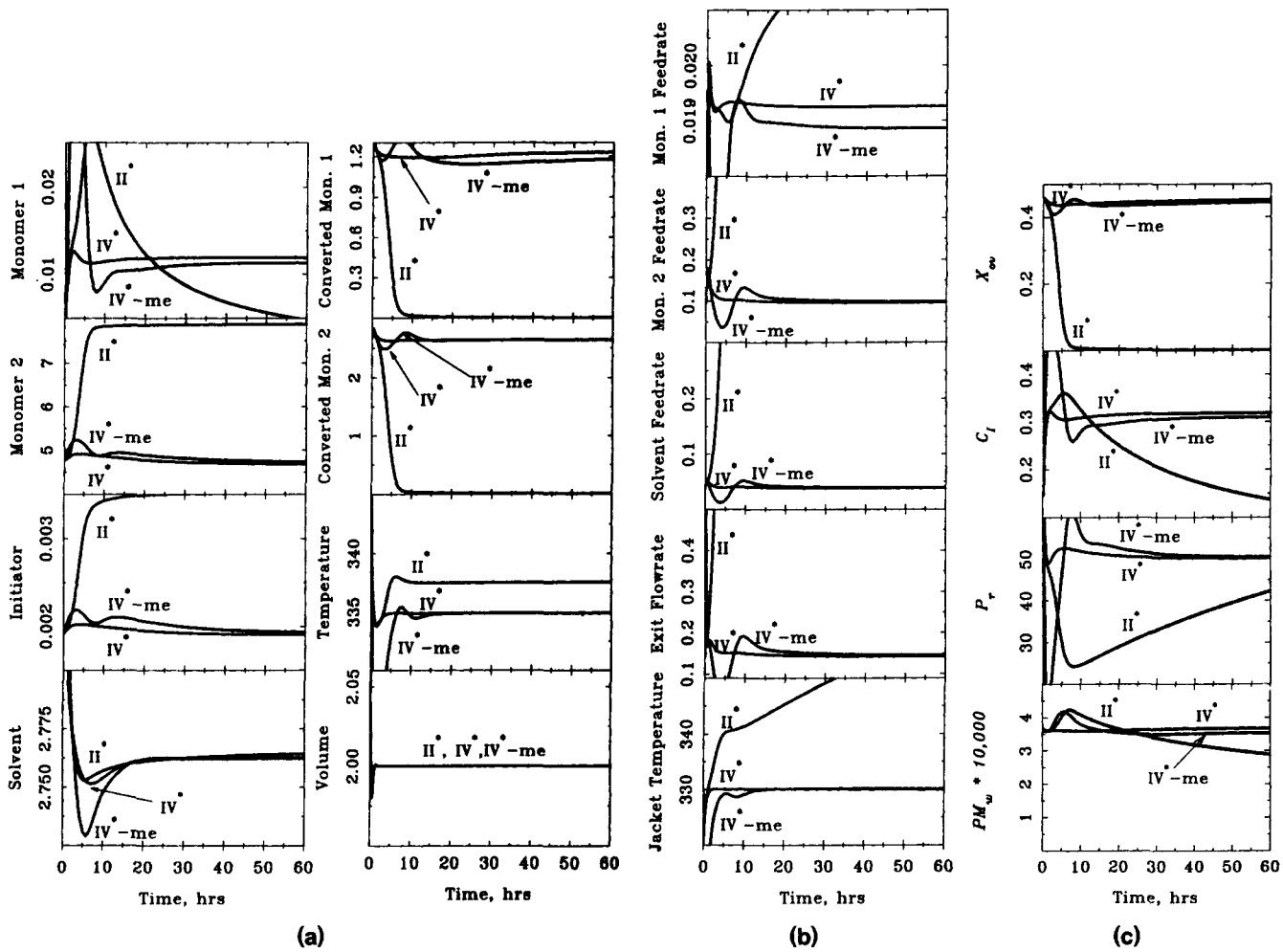

Figure 7. Responses of the closed-loop reactor with the linear output-feedback approximation control of cases II*, IV* ($\bar{u}$ and $A(x)$ evaluated with perfect model), and IV*-me ($\bar{u}$ and $A(x)$ evaluated with an erroneous model): (a) states (kmol/m^3, K, m^3); (b) control inputs (m^3/h, K); and (c) quality attributes.

and the contraction factors were neglected. In this case, the zero-dynamics of the linear output-feedback controller (Eq. 21) still has the nominal operation as an attractor, but is not unique. However, its domain of attraction seems to be sufficiently large for a practical situation. It must be kept in mind that extreme operating conditions and modeling errors have been chosen on purpose, in order to subject the proposed control approach to a severe test. According to the approximated model, the nominal state and input are given by

$$\bar{x}_m = [0.01187 \text{ kmol/m}^3,\ 6.91039 \text{ kmol/m}^3,\ 0.00335 \text{ kmol/m}^3,\ 2.76226 \text{ kmol/m}^3,\ 0.19051 \text{ kmol/m}^3,\ 2.63679 \text{ kmol/m}^3,\ 334.92 \text{ K},\ 2.0 \text{ m}^3]^T$$

$$\bar{u}_m = [1.87814 \times 10^{-2} \text{ m}^3/\text{h},\ 9.3907 \times 10^{-2} \text{ m}^3/\text{h},\ 3.94891 \times 10^{-2} \text{ m}^3/\text{h},\ 297.0 \text{ K},\ 1.46195 \times 10^{-1} \text{ m}^3/\text{h}]^T.$$

As shown in Figure 7 (case IV*-me), the linear output-feedback controller (Eq. 21), with modeling errors, is able to stabilize the reactor about the desired nominal operation, with the outputs reaching the desired nominal values, and the quality variables reaching their target values with rather small offsets. In particular, the molecular weight has an offset that is similar to the standard deviations of the measurement equipment. In a practical operation, this offset could be reduced by means of a suitable adjustment of model parameters. As expected from conventional control practice, the presence of modeling errors and integral action yield a response that is more oscillatory and sluggish than the response of the perfect model-based controller. With more tuning and testing, this response could be improved. As mentioned before, the capacity of the controller to tolerate modeling errors is due to the fact that the decoupling matrix is independent of the polymerization rate functions, which in turn is a consequence of having required the relative output degrees to be equal to one. The reactor and its conventional-type control scheme (i.e., case IV*-me) are represented in Figure 1.

In industrial practice, it is very important to change the operating conditions as safely and quickly as possible, such that the production of off-specification polymer is minimized (Oshima et al., 1994). With this idea, the linear output-feedback controller with modeling errors (case IV*-me) will be tested with setpoint changes in the production rate and production grade. Let us assume that the reactor is operating with the linear output-feedback controller (Eq. 21) with mod-

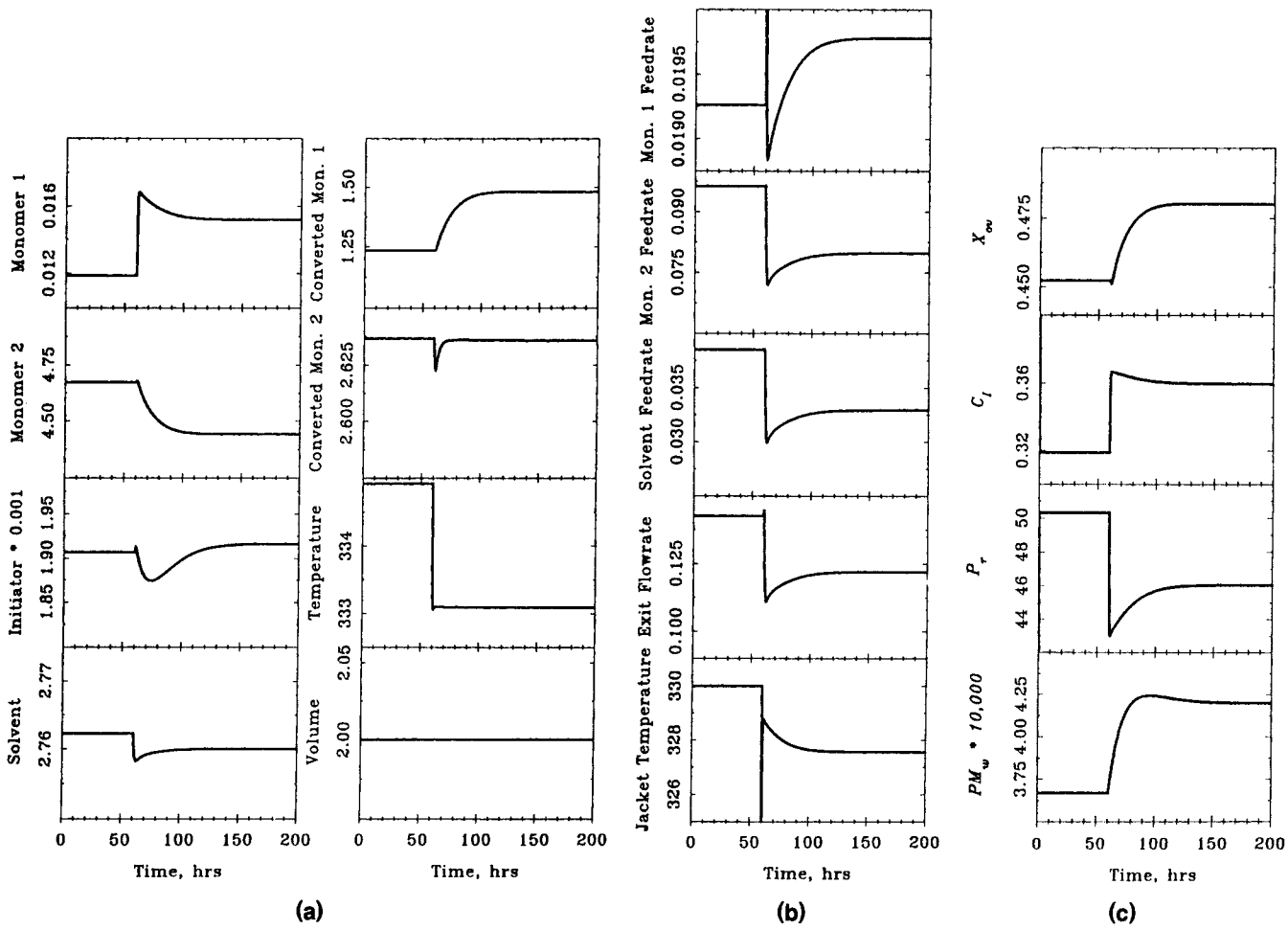

Figure 8. Responses of the closed-loop reactor with the linear output-feedback approximation control of case IV* with modeling errors (IV*-me), subjected to a set-point change: (a) states (kmol/m^3, K, m^3); (b) control inputs (m^3/h, K); and (c) quality attributes.

eling errors, yielding the nominal production rate and grade of the previous runs. Suddenly, at time $t = 60$ h the reactor is required to increase the instantaneous composition by 12.8%, the molecular weight by 14.5%, and decrease the production rate by 11.5%. Based on the approximated model, the corresponding setpoint $\bar{y}$ was generated, as were the new nominal values of inputs and states, $\bar{u}_m$ and $\bar{x}_m$. According to Eq. 21, the new values of u_m and x_m constitute updated information that should be feedforwarded to the controller. However, to test the controller, such updated feedforwarding will not be done. The response of the closed-loop reactor is shown in Figure 8. As can be seen, the settling times of the states, production rate, and grade closely resemble the attainable limiting values estimated in the previous subsection. In particular, composition C_I and production rate P_r reach their target values in about 1.5 nominal residence settling times (that is, $\approx$60 h), and the molecular weight settles in about 2 nominal residence settling times (that is, $\approx$80 h). In terms of nominal residence times, the responses are about four times faster than in Congalidis et al. (1989), keeping in mind that our redesigned operating condition implies a higher conversion with gel effect, lower temperature, and larger residence time.

The main differences between the proposed control scheme and the one presented in Congalidis et al. (1989) are the output maps. Congalidis et al.'s control scheme regulates P_r, C_I, PM_w, and T, and in our case M_1, P_2, S, T, and V are regulated. These authors concluded that the temperature loop had to be tuned conservatively. In cases II* and IV*, the temperature loop does not have interaction problems, because the control scheme is underlied by a perfect compensator design. Compared to Congalidis et al.'s conventional control scheme, case IV* is a linear approximation of a perfect compensator that retains the basic interaction information. This is why case IV* does not present temperature loop interaction problems.

Implementation Guidelines

The application of the proposed control design methodology requires a model of the reactor, as well as standard numerical solvers of nonlinear algebraic and differential equations. To determine the set of steady states and their stabilities, the algebraic solver can be the basis for a continuation method to generate a bifurcation diagram. These numerical tools are conventional, and their application to the study of

an open-loop reactor can be seen in Hamer et al. (1981) and references therein. As regards the reactor model, it must include the gel effect and its implications on the copolymerization kinetics (Teymour and Ray, 1992a), and on the heat-transfer coefficient (Alvarez et al., 1990).

For a given production rate and product grade, a candidate reactor operating condition must be established, and its open-loop dynamics (i.e., uniqueness/multiplicity, settling times, parametric sensitivity, etc.) must be assessed. Then, for each of the three output pairs (M_1, M_2), (M_2, P_1), and (M_1, P_2), the zero dynamics with unity output degrees must be studied, rejecting the cases where the operating state is not robustly stable with a sufficiently large region of attraction. If the set of admissible output pairs (y_1, y_2) is nonempty, test the robust stability of the modified zero dynamics (Eq. 22) associated to the linear output-feedback controller (Eq. 21). If at least one of the candidate control configurations has robustly stable modified zero dynamics (Eq. 22), the conventional-type reactor control problem is solvable. Otherwise, two alternatives may be considered: (1) to redesign the reactor in order to have a more moderate operating condition (say, reduce the fraction of solids, modify the residence time, etc.); or (2) to try higher relative degrees in y_1 and or y_2, with the understanding that the resulting interaction and robustness problems may be difficult to handle with a conventional-type control scheme.

Conceptually speaking, the design or redesign of the operating condition, equipment, and control scheme must be seen as three aspects of a unified design methodology, the rigorous consideration of which is an open problem that goes beyond the scope of the present work.

Conclusions

The control problem of a continuous-solution copolymerization reactor has been addressed with nonlinear control tools in conjunction with considerations from copolymerization kinetics and polymer reactor engineering. First, it was established how the kind of copolymerization kinetics, the reactor equipment, the operation condition, and the choice of control outputs determined the basic solvability of the control problem; identifying criteria to guarantee closed-loop input-state uniqueness, the kind and degree of the interaction problem, and the attainable closed-loop dynamics that set the response time of the product grade. With the proposed approach, the source and nature of the input-multiplicity, interaction, robustness, and sluggish response problems were identified, yielding a procedure with criteria to establish whether, for a given choice of outputs, those problems are solvable or not. For the outputs that led to a solution, the proposed approach yielded a design-tuning procedure to build and tune the rigorous nonlinear state-feedback controller (to be implemented with a suitable closed-loop observer) and its practically oriented linear output-feedback approximation. If the problem was not solvable, the methodology gave guidelines to choose a different set of outputs and/or to redesign the reactor and operating condition.

The crucial linear, noninteractive, pole-assignation (LNPA) property that underlay the solvability of the nonlinear geometric control problem enabled us to have a tuning procedure that was simple in the sense that it could be carried out with vocabulary and techniques from conventional control for linear SISO systems. In particular, to obtain a fast closed-loop response with reasonable control action, the choice of the control gains was guided by the dominant reactor dynamics, and integral action was included to handle modeling errors. By doing so, the design-tuning procedure was made appealing to the industrial control practitioner. The specification of having a nonlinear proportional feedback controller with first-order closed-loop dynamics led to a linear proportional-integral output-feedback control scheme with tolerance to modeling errors, as the underlying perfect interaction compensator design did not depend on the copolymerization kinetics.

The proposed control approach was applied to a copolymerization system that was studied earlier with conventional linear control techniques, and where the key interaction, robustness, and input multiplicity problems had manifested themselves. Compared with the previous control study, the proposed linear output-feedback controller yielded a faster closed-loop response to changes in production rate and production grade, in spite of dealing with a more difficult operating condition. The proposed control methodology showed how nonlinear control theory can be fruitfully blended with polymerization engineering notions and tools to yield practically oriented control schemes that incorporate considerations from equipment and process designs.

Acknowledgments

This work was carried out with the support of the Mexican Science Technology Council (CONACYT-59279) and of the Centro de Investigación en Polímeros (CIP) for S. Padilla, and from CIP fellowship program for J. Alvarez.

Notation

C_{p_1} = monomer 1 liquid heat capacity, kJ/kg·K
C_{p_2} = monomer 2 liquid heat capacity, kJ/kg·K
C_{p_s} = solvent liquid heat capacity, kJ/kg·K
I_e = molar density of initiator, kmol/m^3
M_1^w = molecular weight of monomer 1, kg/kmol
M_2^w = molecular weight of monomer 2, kg/kmol
M_{1_e} = molar density of monomer 1, kmol/m^3
M_{2_e} = molar density of monomer 2, kmol/m^3
P_1 = monomer 1 in polymer form, kmol/m^3
P_2 = monomer 2 in polymer form, kmol/m^3
q_I = feedrate of initiator, m^3/s
S_e = molar density of solvent, kmol/m^3
S^w = molecular weight of solvent, kg/kmol
T_e = feed-rate temperature, K
γ = overall heat-transfer coefficient, kJ/m^2·s·K

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Appendix A: Model Functionalities

$$R_1 = [(k_{p_{aa}} + k_{x_{aa}})P + (k_{p_{ba}} + k_{x_{ba}})Q]M_1$$

$$R_2 = [(k_{p_{ab}} + k_{x_{ab}})P + (k_{p_{bb}} + k_{x_{bb}})Q]M_2$$

$$R_I = k_d I$$

$$\beta_1 = [(-\Delta H_{p_{aa}})(k_{p_{aa}} + k_{x_{aa}})PM_1 + (-\Delta H_{p_{ba}})(k_{p_{ba}} + k_{x_{ba}})QM_2](V\rho C_p)^{-1}$$

$$\beta_2 = [(-\Delta H_{p_{ab}})(k_{p_{ab}} + k_{x_{ab}})PM_2 + (-\Delta H_{p_{bb}})(k_{p_{bb}} + k_{x_{bb}})QM_2](V\rho C_p)^{-1}$$

$$\epsilon_{m_1} = (\rho_1 \rho^{-1} - M1_e \omega_1)\omega^{-1}$$

$$\epsilon_{m_2} = (\rho_2 \rho^{-1} - M2_e \omega_2)\omega^{-1}$$

$$\epsilon_s = (\rho_s \rho^{-1} - S_e \omega_3)\omega^{-1}$$

$$\epsilon_1 = (\omega_1 - \omega_4)\omega^{-1}$$

$$\epsilon_2 = (\omega_2 - \omega_5)\omega^{-1}$$

$$\omega = 1 - \omega_1 + \omega_2 + \omega_3 + \omega_4 + \omega_5$$

$$\omega_1 = M_1^w(1 - \rho\rho_1^{-1})M_t^{-1}$$

$$\omega_2 = M_2^w(1 - \rho\rho_2^{-1})M_t^{-1}$$

$$\omega_3 = S^w(1 - \rho\rho_s^{-1})M_t^{-1}$$

$$\omega_4 = M_1^w\left(1 - \rho\rho_{P_1}^{-1}\right)M_t^{-1}$$

$$\omega_5 = M_2^w\left(1 - \rho\rho_{P_2}^{-1}\right)M_t^{-1}$$

$$M_t = M_1 M_1^w + M_2 M_2^w + SS^w + P_1 M_1^w + P_2 M_2^w$$

$$\Omega_1 = \rho_1(V\rho C_p)^{-1}(T_e - T)$$

$$\Omega_2 = \rho_2(V\rho C_p)^{-1}(T_e - T)$$

$$\Omega_I = \rho_I(V\rho C_p)^{-1}(T_e - T)$$

$$\Omega_s = \rho_s(V\rho C_p)^{-1}(T_e - T)$$

Heat-transfer correlations (see Bondy and Lippa (1983))
Reactor diameter, $Dr = 1.3$ m
Impeller diameter, $Da = 0.7$ m
Agitation speed, $N = 140$ rpm

$$k = 1.93 \times 10^{-4} \text{ kJ/m·s·K}$$

$$\alpha_v = 4.14 \times 10^{25}$$

$$b_v = 9.6763$$

$$c_v = 1.99$$

$$a_a = 0.1678$$

$$b_a = 0$$

$$c_a = -1.23577$$

$$t_g = 387.15$$

$$a_e = 0.03$$

$$a_h = 0.74$$

$$b_h = 2/3$$

$$c_h = 1/3$$

$$d_h = 0.14$$

$$a = a_a + b_a(1 - T/t_g) + c_a(1 - T/t_g)^2$$

$$e = \exp[-2.3x_m/(a + a_e x_m)]$$

$$a_{tc} = a_a + b_a(1 - T_j/t_g) + c_a(1 - T_j/t_g)^2$$

$$e_{tc} = \exp[-2.3x_m/(a_{tc} + a_e x_m)]$$

$$\mu = a_v(T - 273)^{-b_v} e^{c_v}$$

$$\mu_j = a_v|(T_j - 273)^{-b_v} e_{tc}^{c_v}$$

$$R_e = \rho N Da^2/\mu$$

$$P_r = \mu C_p/k$$

if $Re \leq 400$, then $a_h = 0.54$, else $a_h = 0.74$

$$h = a_h \frac{k}{Dr} R_e^{b_h} P_r^{c_h} \frac{\mu d_h}{\mu_j}$$

Heat transfer area, $A = \pi Dr^2/4 + 4V/Dr$

$\gamma = hA/(\rho V C_p)$

Appendix B: Decoupling Matrices for Cases II, III and IV

Case II.

$$A(x)=V^{-1}\begin{pmatrix} M_{1_e}-M_1\epsilon_{m_1} & -M_1\epsilon_{m_2} & -M_1\epsilon_s & 0 & 0 \\ -M_2\epsilon_{m_1} & M_{2_e}-M_2\epsilon_{m_2} & -M_2\epsilon_s & 0 & 0 \\ -S\epsilon_{m_1} & -S\epsilon_{m_2} & S_e-S\epsilon_s & 0 & 0 \\ V\Omega_1 & V\Omega_2 & V\Omega_S & V\gamma & 0 \\ V\epsilon_{m_1} & V\epsilon_{m_2} & V\epsilon_s & 0 & -V \end{pmatrix}$$

Case III.

$$A(x)=V^{-1}\begin{pmatrix} -M_2\epsilon_{m_1} & M_{2_e}-M_2\epsilon_{m_2} & -M_2\epsilon_s & 0 & 0 \\ -P_1\epsilon_{m_1} & -P_1\epsilon_{m_2} & -P_1\epsilon_s & 0 & 0 \\ -S\epsilon_{m_1} & -S\epsilon_{m_2} & S_e-S\epsilon_s & 0 & 0 \\ V\Omega_1 & V\Omega_2 & V\Omega_S & V\gamma & 0 \\ V\epsilon_{m_1} & V\epsilon_{m_2} & V\epsilon_s & 0 & -V \end{pmatrix}$$

Case IV.

$$A(x)=V^{-1}\begin{pmatrix} M_{1_e}-M_1\epsilon_{m_1} & -M_1\epsilon_{m_2} & -M_1\epsilon_s & 0 & 0 \\ -P_2\epsilon_{m_1} & -P_2\epsilon_{m_2} & -P_2\epsilon_s & 0 & 0 \\ -S\epsilon_{m_1} & -S\epsilon_{m_2} & S_e-S\epsilon_s & 0 & 0 \\ V\Omega_1 & V\Omega_2 & V\Omega_S & V\gamma & 0 \\ V\epsilon_{m_1} & V\epsilon_{m_2} & V\epsilon_s & 0 & -V \end{pmatrix}$$

Manuscript received Feb. 8, 1996, and revision received Aug. 22, 1996.