Target Linearization and Model Predictive Control of Polymerization Processes

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Modeling and control of prototypical industrial polymerization processes are presented in the presence of disturbances and plant-model mismatch. The process model consists of the material balances for the reactor, energy balance for the reactor, and a simplified dynamic model for the downstream flash separator. The process model is used to develop a TLMPC that separates the target calculation from dynamic regulation in a closed-loop setting. The main feature of the proposed controller (TLMPC) is that the targets are calculated from the nonlinear model, and then used to get the correct linearized model. The linearized model is utilized to calculate the actual control moves for dynamic regulation, and develop an extended Kalman filter for state estimation. The resulting TLMPC controller does not have the computational burdens associated with a full NMPC and gives similar performance to NMPC. The TLMPC shows optimal plant startup, product grade transitions, and regulation around a set point for the polymerization process in comparison to a fixed LMPC. Robustness with respect to incorrect model kinetic parameters, initial state estimates and measured disturbances is also shown for TLMPC, as compared to LMPC.

Introduction

Control of polymerization processes is challenging because of inherent nonlinearities, infrequent measurements of product quality, and rapidly changing product requirements. Various forms of feedback controllers ranging from proportional-integral-derivative (PID) controller to nonlinear model predictive controller (NMPC) have been proposed. In this work, a target linearization model predictive controller (TLMPC) based on the nonlinear model is proposed for achieving process objectives in a prototypical polymerization process after demonstrating cases where a fixed linear model predictive controller fails to meet process objectives. The key feature of the proposed TLMPC is that it calculates the steady-state targets using a nonlinear model, and then solves the dynamic regulation problem using the appropriate linearized model. The steady-state targets are evaluated from the nonlinear model by taking into account the measured disturbances at the sample time. The nonlinear optimization problem for the target calculation is small as compared to the one for nonlinear dynamic regulation, and is solved on-line at every sample time. The linearized model is also obtained for the calculated targets at every sample time. This linearized model is used to obtain the control moves for appropriate feedback control. The effectiveness of TLMPC is shown for the typical scenarios of reactor startup, product grade change, and regulation around a set point.

The polymer control problem was initially tackled as an optimal control problem (Lapidus and Luus, 1967; Hicks et al., 1969; Yoshimoto et al., 1971). The optimal control synthesis methods have yielded good results in the laboratory. However, problems naturally arise in industrial reactors due to significant process disturbances and modeling errors. A large number of articles in polymer control are concerned with dynamic controllers like the proportional integral-derivative controller (PID) and adaptations of linear model predictive controller (LMPC). Control studies consisting of appropriately chosen loop pairings and tuned PID controllers have been reported in simulations and laboratory-scale experiments (Congalidis et al., 1989; Kim and Choi, 1991; Chien and Penlidis, 1994; Ellis et al., 1994). An adaptive control scheme with control moves optimal for the current time in-

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terval only is presented in a simulation study for methylmethacrylate polymerization (Houston and Schork, 1987). Model predictive controllers based on Volterra models have been evaluated in simulations (Maner et al., 1996; Maner and Doyle III, 1997). Dynamic Matrix Controller (DMC) is a popular form of LMPC that is widely used in the industry (Meziou et al., 1996). The PID controller and linear model predictive controllers may not be able to achieve process goals for a prototypical polymerization process with strong nonlinearities (Ray, 1986).

NMPC has the potential of improving the feedback control performance for polymerization processes; however, it comes with a significant addition to computational burden. Different techniques of linearization have been proposed to overcome the computational difficulties. Nonlinear control of polymer processes based on input-output linearization controller and gain scheduled controller has been reported in literature. Input-output feedback linearization has been studied by various researchers (Soroush and Kravaris, 1993; Alvarez et al., 1994; Wang et al., 1995; Kurtz and Henson, 1997). The feedback linearization approach does not incorporate constraints in the controller formulation thereby imposing performance limitations for many industrial situations. An approach that linearizes the model around the current operating point to obtain step response coefficients for DMC has been proposed (García, 1984). García's approach will not be able to handle plant-model mismatch adequately because it lacks a state estimator. A feedback controller based on a model arising from a Taylor series expansion of outputs has also been proposed (Mutha et al., 1997). Cluett's work does have an extended Kalman filter (EKF) to perform state estimation for multirate systems, but there are no measured disturbances in his controller. An EKF based nonlinear model predictive controller suggested by Lee and Ricker (1994) that requires a future reference trajectory for outputs has been implemented for polymerization control (Park et al., 2002; Prasad et al., 2002). NMPC has been formulated for batch reactors to control end-use properties within a region (Valappil and Georgakis, 2002). This batch controller has a significant computational burden because it uses the complete nonlinear model for state estimation, dynamic regulation, and target calculation. Ohshima has developed a control scheme (Wang et al., 2000ab; Seki et al., 2001) that also separates the target calculation from dynamic regulation. Ohshima's controller calculates an off-line trajectory of inputs and outputs instead of solving the nonlinear target optimization problem at every sample time by taking measured disturbances into account.

Nonlinear control based on gain scheduling also has been reported (Klatt and Engell, 1998; Doyle III et al., 1998). Standard scheduling methods have been used based on a set of linear models using Bayesian estimation for model selection to achieve nonlinear control (Banerjee and Arkun, 1998; Lakshmanan and Arkun, 1999). Gain scheduling has also been performed in a two-dimensional (2-D) space by explicitly accounting for the time variation of the scheduling variable (Doyle III et al., 1998). A set of linear models requires extensive bookkeeping.

An important feature of industrial processes is that the key product quality measures often have long sampling times with associated delays. A two-tier cascade control scheme based on a linear model has been used by industrial researchers to deal with the unavailability of on-line key product quality variable measurements (Ogunnaike, 1994). In the two-tier system, the set points for the on-line outputs are based on the targets for the laboratory outputs. Hence, the infrequent laboratory measurements are not used in the feedback control loop. The long sampling times with associated delays for polymer quality measures have been considered using PID controllers (Kim and Choi, 1991; Ellis et al., 1994). The process model has also been directly used in a multirate state estimator for solution polymerization of styrene (Tatiraju et al., 1998). In this work, the infrequent product quality measures are used along with other on-line measurements by constructing the equivalent raised system for the multirate sampling control problem (Lee et al., 1992; Feuer and Goodwin, 1996).

In this article, the chain growth copolymerization of methyl methacrylate (MMA) and vinyl acetate (VA) with free radical kinetics is taken from the open literature (Congalidis et al., 1989) and modeled as a prototypical industrial solution copolymerization process. The process model is given realistic measured disturbances and plant-model mismatch based on our experience with an industrial copolymerization process (Bindlish et al., 1997; Bindlish, 1999), thereby making the feedback control problem representative of the challenges faced by industrial practitioners. The polymerization control problem is solved using a TLMPC which has the appropriate computational complexity. TLMPC gives a similar performance to NMPC with lesser computational burden, and is required because LMPC fails to attain feedback control objectives for typical industrial control scenarios. This article makes the following contributions to the field of polymerization control:

- Introduces TLMPC in which the state and input targets are updated by solving the full nonlinear model given the measured disturbances and polymer grade; the regulation problem is solved with the linearized model based on the current steady-state target.
- Defines a representative polymer control problem having multirate sampling with product property measurement delay and realistic process objectives, and measured and unmeasured disturbances.
- Demonstrates typical industrial startup and grade change cases in which fixed linear control laws fail and TLMPC succeeds.

Process Description and Model

The MMA-VA copolymerization process consists of a well-mixed reactor followed by a flash separator, both shown in Figure 1. The process is used to make various grades of copolymer. These grades are characterized by the mole fraction of monomer A (MMA) in the copolymer product and the intrinsic viscosity of the copolymer product. The copolymer composition and viscosity are varied for each grade depending upon market requirements. The feed to the reactor consists of the monomers (MMA and VA), initiator (AIBN), transfer agent (acetaldehyde), and inhibitor (m-dinitrobenzene) dissolved in a solvent (benzene). The transfer agent is used to provide better control of the polymer molecular weight. A coolant is employed for the removal of heat re-

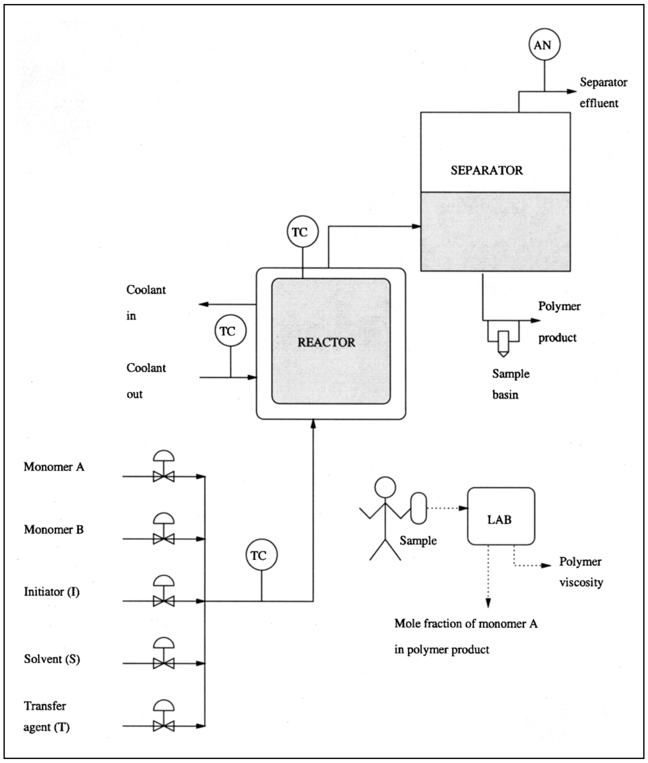


Figure 1. MMA-VA polymerization process.

leased due to polymerization. The polymer product is separated from the unreacted hydrocarbons in a downstream separator. A physical model based on first principles is proposed to enable us to capture the information in the process output measurements and further use it to develop appropriate feedback control strategies.

The flow rate and composition of the feed, and product streams, are measured along with the reactor temperature. The polymer production rate is obtained by an on-line measurement of the refractive index that yields conversion, thereby giving the production rate. An off-line laboratory measurement is also made for the polymer viscosity and composition. The polymer viscosity is measured using a capillary viscometer and the polymer product composition is measured using NMR spectroscopy. The polymer product quality measurements of viscosity and composition are characterized by large delays and sampling intervals.

The copolymerization of methyl methacrylate (A) and vinyl acetate (B) is carried out using free-radical kinetics (Figure 2) in the presence of initiator (I), solvent (S), chain transfer agent (T) and inhibitor (Z) with model parameters taken from open literature (Congalidis et al., 1989). The polymerization reactions are assumed to be irreversible and first-order with respect to each reactant. The differential material balances including the rate expressions and the energy balance coupled with the equations for the physical phenomena constitute the dynamic process model. The following assumptions are made for developing the process model:

- Perfectly mixed tank
- Linear mixing rule for reactor density (Russell and Denn, 1972)

The mole balances for the reactor concentrations (c_k) are

$$\frac{dc_k}{dt} = \frac{c_{k,f} - c_k}{\theta_r} + R_k, \qquad k = a, b, i, s, t, z \tag{1}$$

in which $c_{k,f}$ is the feed concentration and R_k is the molar production rate. The reactor residence time (θ_r) is evaluated as

$$\theta_r = \frac{V_r}{Q_f} \tag{2}$$

The species molar concentrations in the feed ($c_{k,f}$) can be determined from molar feed rates ($F_{k,f}$) and volumetric feed rate (Q_f)

$$c_{k,f} = \frac{F_{k,f}}{Q_f}, \qquad k = a, b, i, s, t, z \tag{3}$$

The instantaneous polymerization rate (G_{pi}) can be evaluated using molar production rates (R_a and R_b) along with molecular weights (M_a and M_b)

$$G_{pi} = -R_a M_a - R_b M_b \tag{4}$$

The functions of states and kinetic parameters that appear in the process model are

$$\beta = \frac{(k_{pab} + k_{xab})c_b}{(k_{pba} + k_{xba})c_a} \tag{5}$$

$$l_1 = k_{caa} + 2\beta k_{cab} + k_{cbb}\beta^2 + k_{daa} + 2\beta k_{dab} + k_{dbb}\beta^2 \tag{6}$$

$$l_2 = (k_{za} + k_{zb}\beta)c_z \tag{7}$$

$$l_3 = -2k_i c_i \tag{8}$$

$$\alpha_1 = \frac{k_{paa}c_a}{\sum\limits_{j=a,b}\left(k_{paj}c_j + k_{caj}c_{j\bullet} + k_{daj}c_{j\bullet}\right) + \sum\limits_{j=a,b,s,t}\left(k_{xaj}c_j\right) + k_{za}c_z} \tag{9}$$

$$\alpha_2 = \frac{k_{pbb}c_b}{\sum\limits_{j=a,b}\left(k_{pbj}c_j + k_{cbj}c_{j\bullet} + k_{dbj}c_{j\bullet}\right) + \sum\limits_{j=a,b,s,t}\left(k_{xbj}c_j\right) + k_{zb}c_z} \tag{10}$$

$$k_{cba} = k_{cab} \tag{11}$$

$$k_{dba} = k_{dab} \tag{12}$$

$$c_1 = \frac{2k_i c_i + \sum\limits_{j=a,b} \sum\limits_{k=s,t} k_{xjk} c_{j\bullet} c_k}{k_{paa}(c_a + c_b)} + \frac{\sum\limits_{j=a,b} k_{xja} c_{j\bullet}}{k_{paa}} \tag{13}$$

$$c_2 = \frac{2k_i c_i + \sum\limits_{j=a,b} \sum\limits_{k=s,t} k_{xjk} c_{j\bullet} c_k}{k_{pbb}(c_a + c_b)} + \frac{\sum\limits_{j=a,b} k_{xjb} c_{j\bullet}}{k_{pbb}} \tag{14}$$

The model is simplified using the quasi-steady-state approximation for the live polymers because of their faster kinetic equilibration relative to the other species. The equations for the live and dead polymer chains are simplified by taking the zeroth, first, and second moments.

Dead copolymer composition

The molar concentrations of the two monomers in the dead copolymer (λ_a and λ_b) can be evaluated by using the following molar balances

$$\frac{d\lambda_a}{dt} = \frac{\lambda_{af} - \lambda_a}{\theta_r} - R_a \tag{15}$$

$$\frac{d\lambda_b}{dt} = \frac{\lambda_{bf} - \lambda_b}{\theta_r} - R_b \tag{16}$$

The mole fraction of monomer $A(Y_{ap})$ in the dead polymer can be evaluated as

$$Y_{ap} + \frac{\lambda_a}{\lambda_a + \lambda_b} \tag{17}$$

Energy balance

The reactor energy balance can be written for the continuous stirred tank reactor to obtain reactor temperature (T_r)

$$\frac{dT_r}{dt} = \frac{\left(-\Delta H_{paa} k_{paa} c_a c_{a\bullet} - \Delta H_{pba} k_{pba} c_a c_{b\bullet} - \Delta H_{pab} k_{pab} c_b c_{a\bullet} - \Delta H_{pbb} k_{pbb} c_b c_{b\bullet}\right)}{\rho_r C_{pr}} + \frac{T_f - T_r}{\theta_r} - \frac{U_r S_r (T_r - T_j)}{V_r C_{pr} \rho_r} \tag{18}$$

The energy balance takes into account only the propagation reactions for the live polymer chains ($A_{n,m}$. and $B_{n,m}$.)

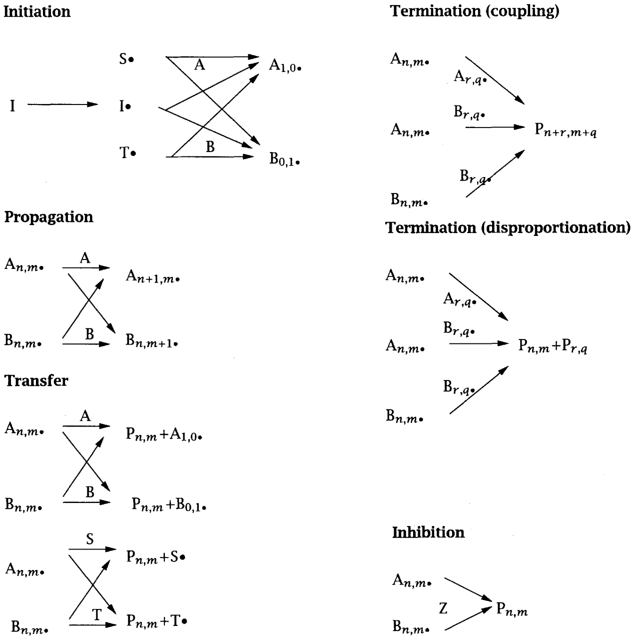
$$A_{n,m\bullet} + A \xrightarrow{k_{paa}} A_{n+1,m\bullet} \tag{19}$$

$$A_{n,m\bullet} + B \xrightarrow{k_{pab}} B_{n,m+1\bullet} \tag{20}$$

$$B_{n,m\bullet} + A \xrightarrow{k_{pba}} A_{n+1,m\bullet} \tag{21}$$

$$B_{n,m\bullet} + B \xrightarrow{k_{pbb}} B_{n,m+1\bullet} \tag{22}$$

because their reaction rates are far greater than the other reactions in the polymerization process (Figure 2).



A = MMA, B = VA, I = AIBN, S = Benzene
T = Acetaldehyde, Z = m-DNB
$A_{n,m\bullet}$ = Live polymer chain containing n units of A and m units of B terminating in A
$B_{n,m\bullet}$ = Live polymer chain containing n units of A and m units of B terminating in B
$P_{n,m}$ = Dead polymer chain containing n units of A and m units of B

Figure 2. Reaction mechanism for MMA-VA copolymerization.

Moments for the active and dead polymers

The intrinsic viscosity of the polymer, which is measured for the copolymer, can be obtained from the average molecular weight using the Mark-Houwink equation (Young and Lovell, 1991)

$$[\eta] = KM_v^a \tag{23}$$

where K and a are constants. In practice, a plot of log(η) vs. log(M_v) is linear over a wide range of molecular weight. The viscosity average molecular weight (M_v) is replaced by the weight average molecular weight (M_w) in Eq. 23, because they are close (Young and Lovell, 1991). The corresponding constants (K and a) are taken from literature (Brandrup and Immergut, 1975). The weight average molecular weight for the polymer is obtained using leading moments for the molecular weight distribution. Hence, there is no need in this work to evaluate explicitly the entire chain length distribution of the polymer. The moments for the molecular weight distribution can be calculated by employing generating functions (Ray, 1972). In this case, the leading moments for the polymer are obtained by explicitly evaluating the series summations involved.

The zeroth, first, and second moments of the molecular weight distribution for the dead polymer can be defined as

$$\psi_k^p = \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} (nM_a + mM_b)^k P_{n,m}, \qquad k = 0,1,2 \tag{24}$$

Leading moments of the differential molecular weight distribution are also used to describe the state of the live polymer chains. The zeroth, first, and second moments of the molecular weight distribution for the live polymer can be defined as

$$\psi_k^{a\bullet} = \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} (nM_a + mM_b)^k A_{n,m\bullet}, \qquad k = 0,1,2 \tag{25}$$

$$\psi_k^{b\bullet} = \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} (nM_a + mM_b)^k B_{n,m\bullet}, \qquad k = 0,1,2 \tag{26}$$

The number average molecular weight (M_n) and the weight average molecular weight (M_w) can be computed using the following relationships

$$M_n = \frac{\psi_1^p}{\psi_0^p} \tag{27}$$

$$M_w = \frac{\psi_2^p}{\psi_1^p} \tag{28}$$

The resulting equations for the zeroth, first, and second moments for the live polymers are

$$\psi_0^{a\bullet} = c_{a\bullet} \tag{29}$$

$$\psi_0^{b\bullet} = c_{b\bullet} \tag{30}$$

$$\psi_1^{a\bullet} = \frac{M_a c_1 \alpha_1}{(1-\alpha_1)^2} + \frac{\alpha_1 \gamma}{r_1}\left(\frac{\psi_1^{b\bullet}}{1-\alpha_1} + \frac{M_a \psi_0^{b\bullet}}{(1-\alpha_1)^2}\right) \tag{31}$$

$$\psi_1^{b\bullet} = \frac{M_b c_2 \alpha_2}{(1-\alpha_2)^2} + \frac{\alpha_2}{\gamma r_2}\left(\frac{\psi_1^{a\bullet}}{1-\alpha_2} + \frac{M_b \psi_0^{a\bullet}}{(1-\alpha_2)^2}\right) \tag{32}$$

$$\psi_2^{a\bullet} = M_a^2 c_1 \alpha_1 \frac{(1+\alpha_1)}{(1-\alpha_1)^3} + \frac{\alpha_1 \gamma}{r_1} \times \left(\frac{\psi_2^{b\bullet}}{1-\alpha_1} + \frac{M_a^2 \psi_0^{b\bullet}(1+\alpha_1)}{(1-\alpha_1)^3} + \frac{2M_a \psi_1^{b\cdot}}{(1-\alpha_1)^2}\right) \tag{33}$$

$$\psi_2^{b\bullet} = M_b^2 c_2 \alpha_2 \frac{(1+\alpha_2)}{(1-\alpha_2)^3} + \frac{\alpha_2}{\gamma r_2} \times \left(\frac{\psi_2^{a\bullet}}{1-\alpha_2} + \frac{M_b^2 \psi_0^{a\bullet}(1+\alpha_2)}{(1-\alpha_2)^3} + \frac{2M_b \psi_1^{a\bullet}}{(1-\alpha_2)^2}\right) \tag{34}$$

Equations 31, 32, 33, and 34 for the moments of live polymer are equivalent to those obtained in literature (Choi and Butala, 1991). The zeroth, first, and second moments for the dead polymer change due to polymerization reactions as

$$\frac{d\psi_0^p}{dt} = \frac{\psi_{0f}^p - \psi_0^P}{\theta_r} + \sum_{j=a,b} \sum_{k=a,b} \left(0.5k_{cjk} c_{j\bullet} c_{k\bullet} + k_{djk} c_{j\bullet} c_{k\cdot} + k_{xjk} c_{j\bullet} c_k\right) + \sum_{j=a,b} \left(\left(k_{xjt} c_t + k_{zj} c_z + k_{xjs} c_s\right) c_{j\bullet}\right) \tag{35}$$

$$\frac{d\psi_1^p}{dt} = \frac{\psi_{1f}^p - \psi_1^p}{\theta r} + \sum_{j=a,b} \sum_{k=a,b} \left(\left(k_{cjk} + k_{djk}\right) \psi_0^{j\bullet} \psi_1^{k\bullet} + k_{xjk} \psi_1^{j\bullet} c_k\right) + \sum_{j=a,b} \left(\left(k_{xjt} c_t + k_{zj} c_z + k_{xjs} c_s\right) \psi_1^{j\bullet}\right) \tag{36}$$

$$\frac{d\psi_2^p}{dt} = \frac{\psi_{2f}^p - \psi_2^p}{\theta_r} + \sum_{j=a,b} \sum_{k=a,b} \left(k_{cjk}\left(\psi_0^{j\bullet}\psi_2^{k\bullet} + \psi_1^{j\bullet}\right) + k_{djk} \psi_0^{j\bullet} \psi_{2\bullet}^{k} + k_{xjk} \psi_2^{j\bullet} c_k\right) + \sum_{j=a,b} \left(\left(k_{xjt} c_t + k_{zj} c_z + k_{xjs} c_s\right) \psi_2^{j\bullet}\right) \tag{37}$$

Equations 35, 36, and 37 for the moments of dead polymer are equivalent to those obtained in literature (Congalidis et al., 1989; Choi and Butala, 1991).

Separator balances

The separator is modeled as a first-order lag on the species concentrations ($c_{k,s}$) with constant level

$$\frac{dc_{k,s}}{dt} = \frac{c_{k,sf} - c_{k,s}}{\theta_s}, \qquad k = a,b,i,s,t,z \tag{38}$$

where the separator residence time (θ_s) can be evaluated as

$$\theta_s = \frac{V_s}{Q_f} \tag{39}$$

Model parameters

The kinetic rate constants are of the Arrhenius type

$$k = Ae^{(-E/RT)} \tag{40}$$

The values of the pre-exponential factors (A) and the activation energies (E) are obtained from literature (Congalidis et al., 1989) and listed in Table 1. The following assumptions are also made for the calculation of the cross termination rate constants (Congalidis et al., 1989)

$$k_{cba} = k_{cab} = \sqrt{k_{caa} k_{cbb}} \tag{41}$$

$$k_{dba} = k_{dab} = \sqrt{k_{daa} k_{dbb}} \tag{42}$$

The physical parameters (Table 2) and thermodynamic parameters (Table 3) for the process are also taken from literature.

Table 1. Kinetic Parameters for MMA-VA Copolymerization

Pre-exponential Factor			Activation Energy (kJ/kmol)	
A_i	4.5×10^{14}	s^{-1}	E_i	1.25×10^5
A_{caa}	4.209×10^{11}	$m^3/kmol\cdot s$	E_{caa}	2.69×10^4
A_{cbb}	1.61×10^9	$m^3/kmol\cdot s$	E_{cbb}	4.00×10^3
A_{daa}	0.0	$m^3/kmol\cdot s$	E_{daa}	0.0
A_{dbb}	0.0	$m^3/kmol\cdot s$	E_{dbb}	0.0
A_{paa}	3.207×10^6	$m^3/kmol\cdot s$	E_{paa}	2.42×10^4
A_{pab}	1.233×10^5	$m^3/kmol\cdot s$	E_{pab}	2.42×10^4
A_{pba}	2.103×10^8	$m^3/kmol\cdot s$	E_{pba}	1.8×10^4
A_{pbb}	6.308×10^6	$m^3/kmol\cdot s$	E_{pbb}	2.42×10^4
A_{xaa}	32.08	$m^3/kmol\cdot s$	E_{xaa}	2.42×10^4
A_{xab}	1.234	$m^3/kmol\cdot s$	E_{xab}	2.42×10^4
A_{xas}	86.6	$m^3/kmol\cdot s$	E_{xas}	2.42×10^4
A_{xat}	2085.0	$m^3/kmol\cdot s$	E_{xat}	2.42×10^4
A_{xba}	5.257×10^4	$m^3/kmol\cdot s$	E_{xba}	1.8×10^4
A_{xbb}	1577.0	$m^3/kmol\cdot s$	E_{xbb}	1.8×10^4
A_{xbs}	1514.0	$m^3/kmol\cdot s$	E_{xbs}	1.8×10^4
A_{xbt}	4.163×10^5	$m^3/kmol\cdot s$	E_{xbt}	2.42×10^4
A_{za}	2.2	$m^3/kmol\cdot s$	E_{za}	0.0
A_{zb}	1.13×10^5	$m^3/kmol\cdot s$	E_{zb}	0,0

Table 2. Physical Parameters for MMA-VA Copolymerization

Physical Parameter	Value
Reactor volume (V_r)	1.0 m^3
Reactor heat-tranfer area (S_r)	4.6 m^2
Separator volume (V_s)	4.0 m^3

Controller Design

The objective of the feedback controller is to maintain the product specifications of copolymer composition and viscosity, along with production rate requirements. The copolymerization control problem (Figure 1) for this process has multiple inputs (feed rates and coolant temperature) and multiple outputs (reactor temperature, separator effluent composition, production rate, copolymer viscosity, and comonomer/monomer ratio in copolymer). There are no requirements for reactor temperature and separator effluent composition, but these output measurements are used to update the state estimates of the process model.

The ratio of the two monomers in the feed can be used to alter the composition of the copolymer product. The feed rate of MMA is maintained constant (0.3 kg/min), whereas the feed rate of VA is manipulated to achieve the required copolymer properties. The amount of initiator that initiates the chain polymerization process by forming free radicals is also maintained constant (0.003 kg/min). The amount of solvent (0.6 kg/min) that is fed to the polymerization reactor is dictated by the solubilities of the feed constituents and also by heat-transfer requirements for the process. The copolymerization control problem will then be characterized by:

- Inputs: Vinyl acetate (VA) feed rate, Acetaldehyde feed rate, coolant temperature; and
- Outputs: separator effluent composition, reactor temperature, polymer production rate, copolymer viscosity, and copolymer composition.

The separator effluent compositions, reactor temperature, and polymer production rate are on-line measurements that are available at every sampling time. The copolymer compostion and viscosity are laboratory measurements with delay. The polymer production requirements are dictated by the market demand and capacity of the polymer finishing line. The polymer composition and viscosity are also product quality measures that are set according to market needs. The feed rate of m-DNB that acts as an inhibitor along with the feed temperature are the measured disturbances entering the process. This feedback control problem for the polymerization process is challenging because of the following factors:

- Lack of on-line measurements for key product quality measures;
- Significant plant-model mismatch and disturbances; and
- Frequent polymer grade changes, including plant startup.

A model predictive controller is developed next to achieve the necessary feedback during plant startup and product grade change for the polymerization process. The nonlinear model is used to develop the target linearization model predictive controller (TLMPC). The process model can be written as

$$\frac{dx}{dt} = f(x,u,d,t) \tag{43}$$

$$y = h(x) \tag{44}$$

where

$$x = \left[c_a, c_b, c_i, c_s, c_t, c_z, \lambda_a, \lambda_b, T_r, \psi_0^p, \psi_1^p, \psi_2^p, c_{a,s}, c_{b,s}, c_{s,s}\right]^T \tag{45}$$

$$u = \left[G_{b,f}, G_{t,f}, T_j\right]^T \tag{46}$$

Table 3. Thermodynamic Parameters for MMA-VA Copolymerization

Thermodynamic Parameter	Value		Reference
Overall heat-transfer coefficient (U_r)	6.0×10^{-2}	$kJ/m^2\cdot s\cdot K$	Congalidis et al. (1989)
Heat of reaction 19 ($-\Delta H_{paa}$)	54.0×10^3	kJ/kmol	Congalidis et al. (1989)
Heat of reaction 21 ($-\Delta H_{pba}$)	54.0×10^3	kJ/kmol	Congalidis et al. (1989)
Heat of reaction 20 ($-\Delta H_{pab}$)	86.0×10^3	kJ/kmol	Congalidis et al. (1989)
Heat of reaction 22 ($-\Delta H_{pbb}$)	86.0×10^3	kJ/kmol	Congalidis et al. (1989)
Specific heat of MMA (C_{p_a})	1.9	$kJ/kg\cdot K$	Mark et al. (1985)
Specific heat of VA (C_{p_b})	1.7556	$kJ/kg\cdot K$	Gallant (1968)
Specific heat of benzene (C_{p_s})	1.672	$kJ/kg\cdot K$	Perry et al. (1984)
Specific heat of polymer (C_{p_p})	1.44	$kJ/kg\cdot K$	Brandrup and Immergut (1975)
Density of MMA (ρ_a)	940.0	kg/m^3	Brandrup and Immergut (1975)
Density of VA (ρ_b)	931.7	kg/m^3	Brandrup and Immergut (1975)
Density of benzene (ρ_s)	878.65	kg/m^3	Brandrup and Immergut (1975)

$$d = \left[G_{a,f}, G_{i,f}, G_{s,f}, G_{z,f}, T_f \right]^T \tag{47}$$

$$y = \left[c_{a,s}, c_{b,s}, c_{s,s}, T_r, G_{pi}, Y_{ap}, [\eta] \right]^T \tag{48}$$

A linear process model is obtained by evaluating the Jacobian of the nonlinear model at the steady state using Maple (Redfern, 1996). The discretized linear model in terms of deviation variables is written as

$$x_{k+1} = Ax_k + Bu_k + B^m d_k \tag{49}$$

$$y_k = Cx_k \tag{50}$$

In the feedback controller simulations, the plant is represented by the nonlinear model simulation along with an additive output disturbance ($p(t)$) that is based on our experiences with an industrial copolymerization (Bindlish, 1999; Bindlish et al., 2003). The variation of output disturbance ($p(t)$) with time is used to give the following plant representation for simulations

$$\frac{dx}{dt} = f(x,u,d) \tag{51}$$

$$y = h(x) + p(t) \tag{52}$$

The following two models are used in the feedback controller:

- fixed linear model (LMPC); and
- re-linearized model for the calculated targets at every sample time (TLMPC), to carry out plant startup, polymer grade changes and regulation around a setpoint.

TLMPC formulation

The existing model predictive control formulation based on an infinite horizon linear quadratic regulator (Muske and Rawlings, 1993; Scokaert and Rawlings, 1998) is used with some modifications to obtain a TLMPC for the polymerization process. The nonlinear model is re-linearized at a steady state obtained from the target calculation to obtain a new linear model at every sample time. The linear model is used to evaluate the controller moves for dynamic regulation, and develop an extended Kalman filter for state estimation. The effectiveness of the feedback controller is evaluated by tracking grade transitions, which may include plant startup, in the presence of measured and unmeasured disturbances entering the process. The TLMPC target tracking problem can be divided into four parts as follows:

Target Calculation. The steady-state targets for the states (x_{ss}) and inputs (u_{ss}) can be evaluated using a nonlinear program to minimize the deviations of the outputs from their targets ($\bar{y}$) and the deviation of the inputs from their desired values ($\bar{u}$). The nonlinear target tracking optimization problem can then be written as follows

$$\min_{x_{ss}, u_{ss}} \Phi = \left(y_{ss} + \hat{p} - \bar{y} \right)^T Q_s \left(y_{ss} + \hat{p} - \bar{y} \right) + (u_{ss} - \bar{u})^T R_s (u_{ss} - \bar{u}) \tag{53}$$

in which Q_s and R_s are positive definite weighting matrices. The governing constraints for the problem are

$$f(x_{ss}, u_{ss}, d) = 0 \tag{54}$$

$$y_{ss} = h(x_{ss}) \tag{55}$$

$$u_{\min} \le u_{ss} \le u_{\max} \tag{56}$$

The estimate of the output disturbance ($\hat{p}$) has been accounted for in the above formulation. The linear model is re-evaluated at the steady-state target (x_{ss}, u_{ss}) for the current measured disturbance (d).

Measured Disturbances. A feedforward control of the measured disturbances is based on the linearized model obtained at the steady-state target (x_{ss}, u_{ss}, d)

$$x^m_{k+1} = x^m_k + B^m d_k \tag{57}$$

$$y^m_k = C^m x^m_k \tag{58}$$

where

$$x^m = \left[c_a, c_b, c_i, c_s, c_t, c_z, \lambda_a, \lambda_b, T_r, \psi^p_0, \psi^p_1, \psi^p_2, c_{a,s}, c_{b,s}, c_{s,s} \right]^T \tag{59}$$

$$d = \left[G_{a,f}, G_{i,f}, G_{s,f}, G_{z,f}, T_f \right]^T \tag{60}$$

$$y^m = \left[c_{a,s}, c_{b,s}, c_{s,s}, T_r, G_{pi}, Y_{ap}, [\eta] \right]^T \tag{61}$$

The above feedforward linear model for the measured disturbances is obtained at every sample time.

Regulator. The receding horizon regulator can be expressed as the following open-loop quadratic objective function

$$\min_{u^N} \Phi_k = \sum_{j=0}^{\infty} \left(y^T_{k+j} Q y_{k+j} + u^T_{k+j} R u_{k+j} + \Delta u^T_{k+j} S \Delta u_{k+j} \right) \tag{62}$$

where Q is the weighting matrix on the outputs, R is the weighting matrix on the inputs, and S is the weighting matrix on the rate of change of inputs. The linear model corresponding to the x_{ss}, u_{ss}, d given in the target calculation is used as the regulation forecaster, resulting in a simple QP to be solved on-line in the regulator. The upper and lower linits for the inputs representing physical limitations for the process are also present. The optimization problem is formulated with a finite number of decision variables to make the problem tractable. The inequality constraints are active for this finite number of sample times along the prediction horizon. In that case, the state and input trajectories approach the set point exponentially and one can calculate cost-to-go by using the unconstrained feedback law for all future times (Gilbert and Tan, 1991; Rao and Rawlings, 1999) resulting in the following expression for Eq. 62

$$\min_{u^k \ldots u^{k+N}} \Phi_k = x^T_{k+N} \overline{Q} x_{k+N} + \Delta u^T_{k+N} S \Delta u_{k+N} + \sum_{j=0}^{N-1} \left(x^T_{k+j} C^T Q C x_{k+j} + u^T_{k+j} R u_{k+j} + \Delta u^T_{k+j} S \Delta u_{k+j} \right) \tag{63}$$

The vector of future open-loop control moves $(u^k \cdots u^{k+N})$ is computed from Eq. 63 and the first input value is injected into the plant. This process is repeated at each subsequent time interval with feedback using the plant measurements to update the state estimate. The terminal state penalty matrix $(\overline{Q})$ for a stable system can be determined from the discrete time algebraic Riccati equation.

State Estimation. The output disturbance model is used to incorporate feedback to obtain state estimates. The estimates of the states $(\hat{x})$ and the output disturbances $(\hat{p})$ can then be obtained using the extended Kalman filter for the nonlinear model

$$\hat{x}_{k+1,k} = f(\hat{x}_{k,k-1}, u_k, d_k) + L_x(y_k - h(\hat{x}_{k,k-1})) \quad (64)$$

$$\hat{p}_{k+1,k} = L_p(y_k - h(\hat{x}_{k,k-1})) \quad (65)$$

The above outlined filter $(L = [L_x, L_p])$ is then used to provide feedback. The copolymer composition and viscosity are not measured at every sample time and have an associated delay. The time-varying linear system can be reformulated as a time invariant raised system (Feuer and Goodwin, 1996) to obtain the filter gain. The laboratory measurements from the plant are adjusted for the delay (k_d) and then used to update the estimate of the associated output disturbance $(\hat{p})$

$$\hat{p}_{k-k_d+1,k-k_d} = y_{k-k_d} - h(\hat{x}_{k-k_d,k-k_d-1}) \quad (66)$$

The above TLMPC is compared with the LMPC for feedback control of polymerization processes during plant startup, polymer grade change, and regulation around a setpoint.

MPC results

The MMA-VA copolymerization process is nonlinear and is a likely candidate for TLMPC feedback to achieve process objectives. Three different scenarios for polymerization control have been examined:

- Plant startup;
- Polymer grade change; and
- Regulation at a fixed setpoint.

In these feedback control simulations, the plant is represented by the nonlinear model simulation along with an additive output disturbance $(p(t))$ that is based on our experience with datasets obtained from industrial polymerization processes. The variation of output disturbance $(p(t))$ with time is used to give the following plant representation for simulations

$$\frac{dx}{dt} = f(x(t), u(t), d(t), t)$$

$$y = h(x) + p(t)$$

The measured disturbances (d) also vary with time and are based on our experience with industrial datasets. These realistic disturbances (d) and plant-model mismatch (p) are used to mimic a typical polymerization process in the following simulations. The time varying output disturbances for regulated measurements and measured disturbances that are used in the feedback control simulations are shown in Figures 3 and 4.

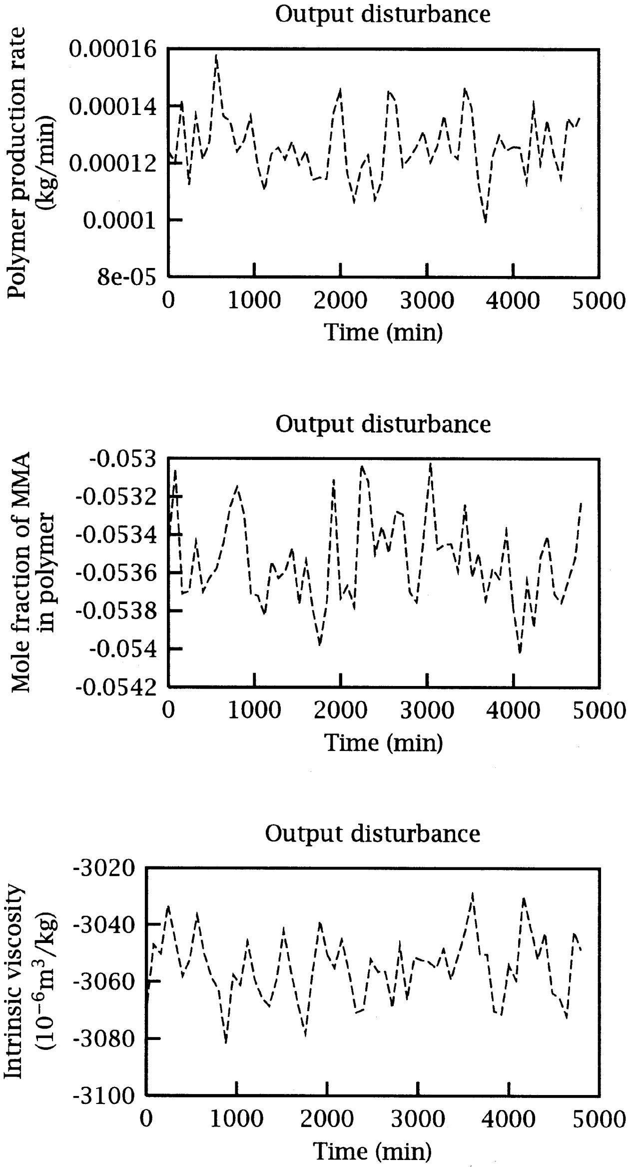


Figure 3. Output disturbance in regulated outputs.

The feedback control simulations also take into account that copolymer composition and viscosity have a larger sampling time with associated delay. The feedback controllers are tuned appropriately to give the desired control action. The quadratic penalty on the deviation of the outputs (Q, Q_s) from their desired values $(\bar{y})$ is set to its nominal value to get the same relative penalty for each output (Eq. 62)

$$Q = Q_s = \frac{1}{\bar{y}^2} \quad (67)$$

The quadratic penalty on the deviation of the inputs (R, R_s) from their desired values $(\bar{u})$ is set to zero, and the quadratic penalty on the rate of change of inputs (S) is set after taking into account the lower bound $(u_{\min})$ and upper bound $(u_{\max})$ into consideration

$$R = R_s = 0 \quad (68)$$

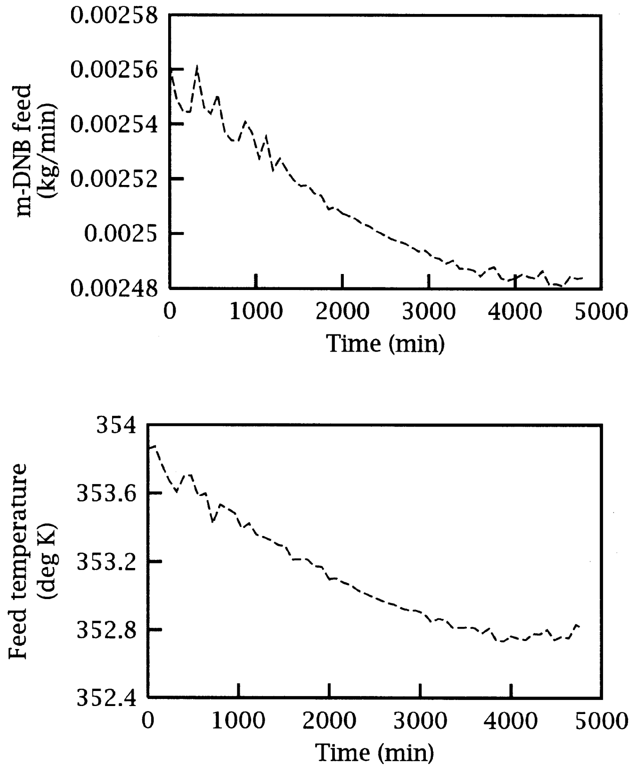


Figure 4. Measured disturbances.

Table 5. Product Grades for MMA-VA Copolymerization

Product Grade	Polymer Production Rate (kg/min)	Mole Fraction of MMA in Copolymer	Copolymer Viscosity (10^{-6} m^3/kg)
A	0.25	0.75	37,000
B	0.30	0.60	35,000
C	0.30	0.65	36,000

Table 6. Feedback Controller Performance for MMA-VA Copolymerization

Case Number	Plant Operation	Feedback Controller	Controller Performance
1	Startup (No polymer to Grade A)	LMPC TLMPC	Fails Succeeds
2	Startup (No polymer to Grade B)	LMPC TLMPC	Succeeds Succeeds
3	Grade change (Grade B to Grade A)	LMPC TLMPC	Fails Succeeds
4	Grade change (Grade B to Grade C)	LMPC TLMPC	Succeeds Succeeds
5	Regulation (Grade A)	LMPC TLMPC	Fails Succeeds
6	Regulation (Grade B)	LMPC TLMPC	Succeeds Succeeds

$$S = S_o \frac{1}{(u_{\max} - u_{\min})^2} \tag{69}$$

The rate of change penalty on inputs is then set by choosing appropriate value of S_o (Table 4). The output disturbance model formulation is employed with filter gain ($L = [0, I]$) to account for the bias between model predictions and plant data. The augmented disturbance model provides the integral action to the controller, thereby resulting in zero steady-state tracking error.

The process objective is to make copolymer grades characterized by composition and viscosity at the required production rates. The copolymer grades specified by their desired targets that are chosen for the feedback controller simulations are listed (Table 5). The main characteristics and conclusions of the feedback controller simulations can be tabulated as follows (Table 6).

Choice of Model in Controller. We first establish the fact that changing polymer product grades is a *nonlinearity* and a linearized model valid at one grade cannot be used for control when the process operates at a different grade. In Table 6 the single, fixed LMPC model is chosen as the linearization of the nonlinear model at the steady state corresponding to *grade B.* We then see success in each case (2, 6) where the transition is to grade B as final product. We see failure when trying to reach grade A, either in a reactor startup, or a grade transition (cases 1, 3, 5). The transition from grade B to C (case 4) is accomplished using LMPC based on model B, showing that not all transitions require the final grade's linearized model to be successful. The target linearization MPC controller, on the other hand, linearizes the nonlinear model based on the current state estimate. Updating the linearized model in this fashion enables the TLMPC controller to make all of the startups and the transitions in either direction.

Figures 5–6 show the regulated outputs and the manipulated inputs for case 1, a typical failure; Figures 7–8 show the regulated outputs and manipulated inputs for case 2, a typical success. LMPC is a failure for case 1 (Figure 5) because the outputs oscillate and never settle at their set points, as compared to TLMPC. The outputs for TLMPC settle after 17 h for the reactor startup, whereas the outputs for LMPC have not settled even after 50 h. The polymer viscosity is aroung 35,000, and is 2,000 different from the specification of 37,000. Based on our experience with industrial practitioners, that much difference is a different polymer grade and clearly an off-specification product. Detailed simulations for all six cases are available in the first author's PhD Thesis (Bindlish, 1999, Figures 6.6–6.41).

Table 4. Input Constraints and Tuning for MMA-VA Copolymerization

	S_o (LMPC)	S_o (TLMPC)	$u_{\max}$	$u_{\min}$
Vinyl acetate feed ($G_{b,f}$)	100	10	3.0 kg/min	0.15 kg/min
Acetaldehyde feed ($G_{t,f}$)	100	100	0.25 kg/min	0.005 kg/min
Coolant temperature (T_j)	100	1,000	373°K	298°K

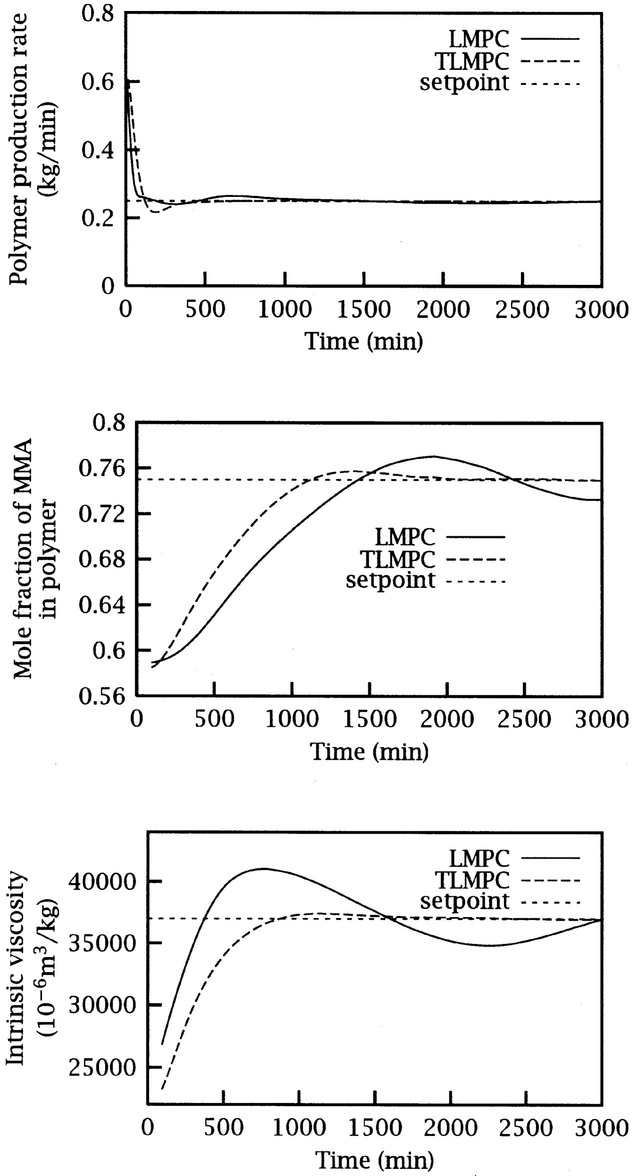


Figure 5. Case 1: Closed-loop outputs during plant startup.

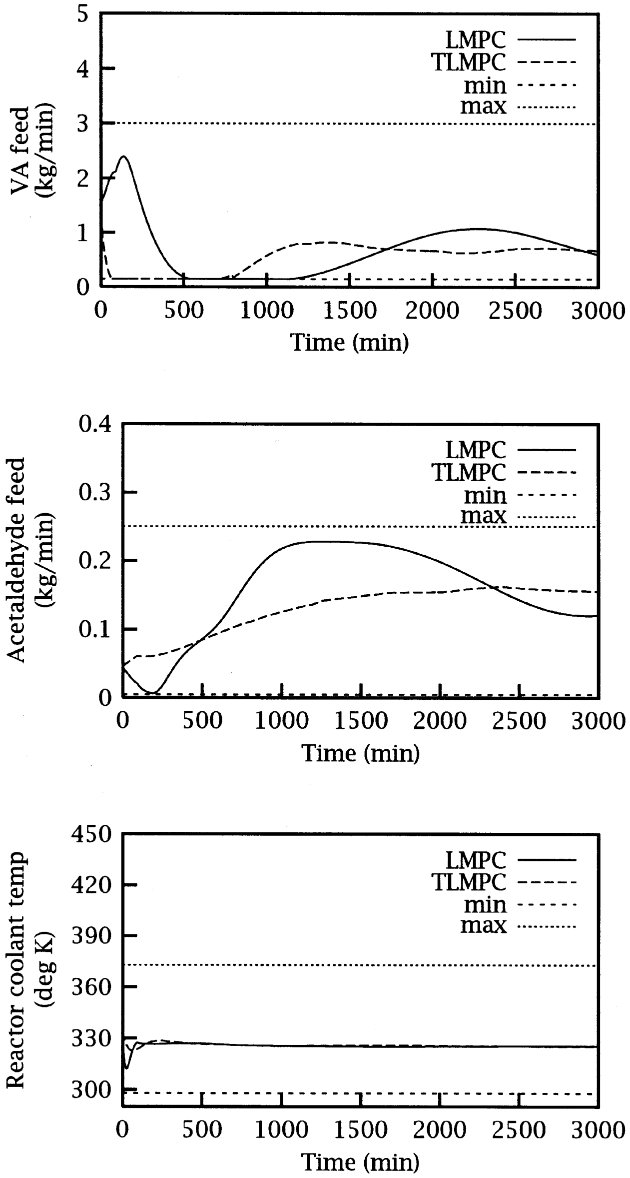


Figure 6. Case 1: Closed-loop inputs during plant startup.

The performance of the TLMPC controller is comparable to a complete NMPC controller for case 3 based on the TLMPC results available in the first author's PhD Thesis (Bindlish, 1999, Figures 6.18–6.23) and NMPC results for the same copolymerization process (Tenny et al., 2001; Rawlings et al., 2002). The performance of the TLMPC controller is very similar to the NMPC controller; even though the NMPC controller assumes that copolymer composition and viscosity are on-line measurements without delay. The TLMPC and NMPC results for the grade transition show that the use of complete nonlinear model for dynamic regulation does not significantly improve the performance of the feedback controller.

Next consider the issue of controller tuning. It is certainly possible to improve performance by an exhaustive search of the LMPC controller's tuning parameters (Q, R, S) and scheduling the tuning parameters based on the final grade. This approach is counterproductive for a model based controller, because we do not have the structure of nonlinearity in place.

Measured Disturbances and Scheduling Linear Models. A simple and logical alternative to the TLMPC approach therefore is to schedule the linear models themselves and select the model based on the final product grade. If we schedule the models for the cases in Table 6 based on the final product grade, the linear MPC controllers succeed in all cases. Scheduling linear models is problematic, however, when we consider the *unregulated inputs* to the process. Achieving a polymer grade is not determined solely by the manipulated inputs u. As shown in Eqs. 44–48, the measured disturbances d play a large role. Consider the case in which we linearize the model for the final grade given one set of measured disturbances, but the measured disturbances vary significantly.

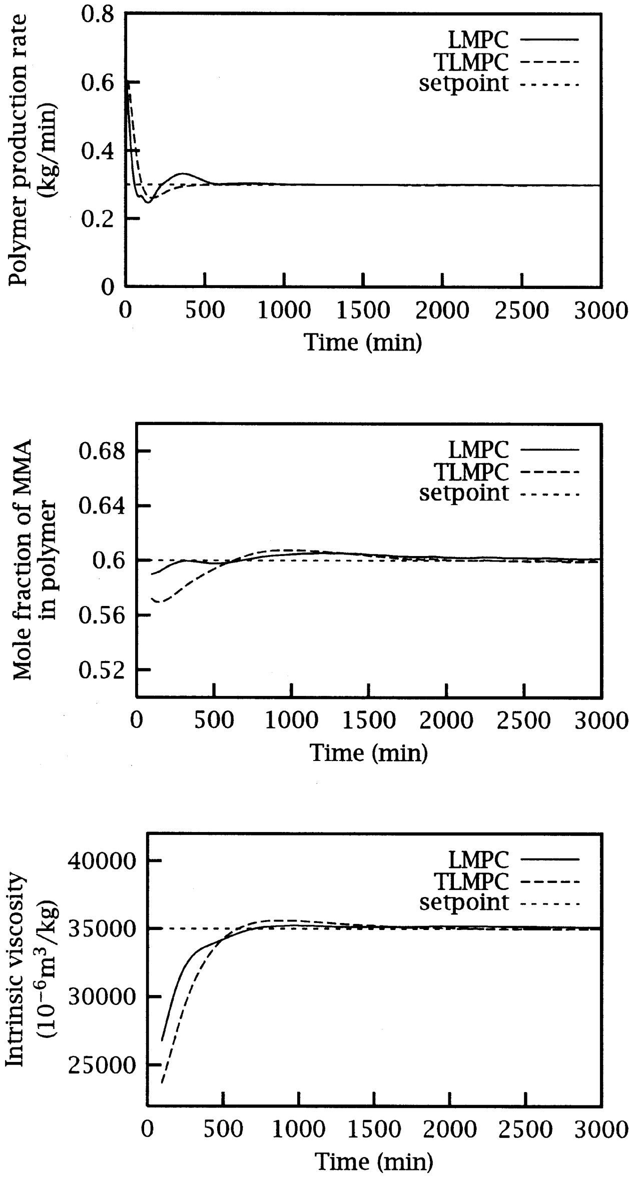


Figure 7. Case 2: Closed-loop outputs during plant startup.

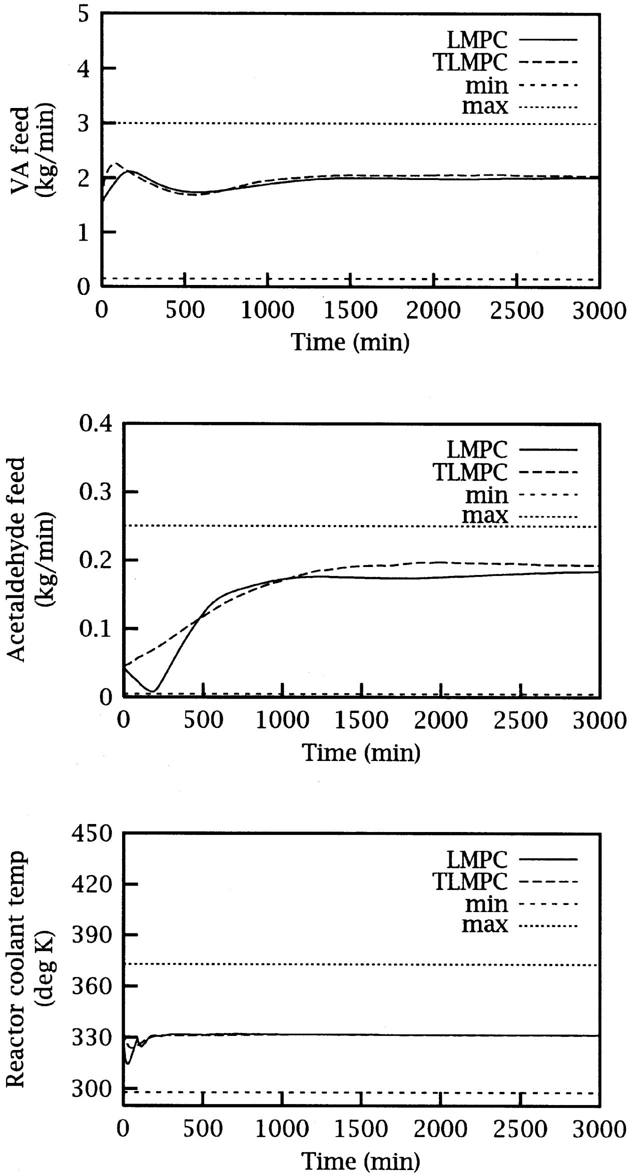


Figure 8. Case 2: Closed-loop inputs during plant startup.

The measured disturbances are moved from their initial values to see their effect on the feedback controllers (Figure 9). The feedrate of MMA is maintained at 0.315 kg/min instead of 0.3 kg/min. The reactor is initially at the set point for the polymer grade. The process objective to keep making the particular grade (Grade A) of copolymer characterized by composition and viscosity, at a certain production rate as disturbances enter the process. The LMPC feedback controller is based on a linear model obtained around the conditions for grade A with different measured disturbances (Figure 4). The LMPC feedback fails whereas a TLMPC feedback succeeds in reaching the process goals for a different set of measured disturbances. The polymer production rate and composition reach their set points for both controllers (Figure 10). The polymer viscosity reaches its set point for TLMPC whereas it oscillates around the set point for LMPC as depicted in Figure 10. The closed-loop inputs corresponding to LMPC and TLMPC are also shown in Figure 11. Hence, the target linearization model predictive controller is able to handle different measured disturbances in comparison to a fixed linear model predictive controller, even if it is scheduled according to the polymer grade.

Finally, one might attempt to schedule the linear models based on both the product grade and the values of the measured disturbances, but the bookkeeping in this case becomes significant. One does not have a finite, well characterized set of measured disturbances as is the case for the polymer grade. The advantage of the TLMPC controller is that it performs this model bookeeping automatically depending on both the grade and the measured disturbances. The price to be paid for this performance is that an optimization problem must be solved for the steady-state targets at each sample time (Eqs.

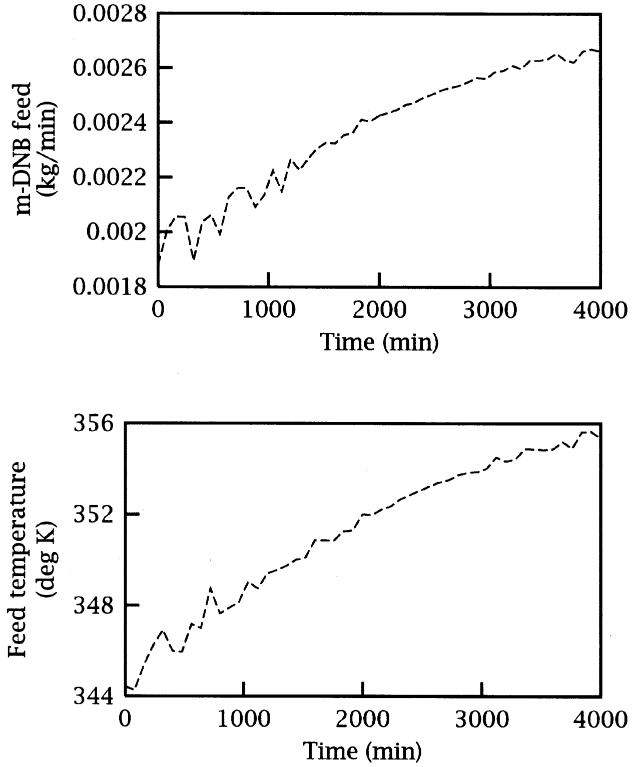


Figure 9. Measured disturbances to examine controller robustness.

53–56). However, it is not necessary to solve the full dynamic nonlinear MPC regulation problem (Bindlish, 1999; Tenny et al., 2001; Rawlings et al., 2002). If the *targets* are updated with the *nonlinear model*, the corresponding *linear model* suffices for the *dynamic regulation* problem in all cases studied for polymer control.

Robustness to Model Error. The performance of the model predictive controllers is also examined for different kinetic parameters and initial states for the model that is used in the controller in comparison to the plant for the first regulation case (case 5). In this case, the filter gain ($L=[L_x, L_p]$) is tuned based on the covariances of state noise (w_k), noise in sensor measurement (v_k), and noise in output disturbance (ξ_k) resulting in an extended Kalman filter for the nonlinear system to evaluate estimates of the states ($\hat{x}$) and the output disturbances ($\hat{p}$). The kinetic parameters are varied based on parameter uncertainty obtained for an industrial copolymerization process (Bindlish, 1999; Bindlish et al., 2003). The pre-exponential factors for the propagation rate constants are varied for the model that is used in the controller, from their nominal values corresponding to the plant simulations (Table 7). Modeling discrepancy is also introduced for the initial state estimates ($\hat{x}_0$) compared to the initial plant states (x_0)

$$\hat{x}_0 = f \cdot x_0 \tag{70}$$

The inputs are tuned by setting $S_o = [100, 1{,}000, 10{,}000]$ to make the controller less aggressive in the presence of plant-model mismatch for kinetic parameters and initial states. The closed-loop response for the regulated outputs (Figure 12) shows the performance in the presence of disturbances, plant-model mismatch, and modeling errors. The performance of TLMPC is better for $f = 1.05$ and $f = 1.1$ that correspond to 5% and 10% relative error, in comparison to $f = 1.5$ and $f = 2.0$ that correspond to 50% and 100% relative error. The corresponding inputs for the TLMPC feedback scheme are shown (Figure 13). The closed-loop inputs for $f = 1.05$ and $f = 1.1$ move around less in comparison to those for $f = 1.5$ and $f = 2.0$. The closed inputs and outputs for $f = 2.0$ exhibit a lack of performance by the feedback controller in the presence of extreme uncertainty in the process model. In conclusion, the proposed target linearization model predictive controller attains process objectives in the presence of realistic disturbances, plant-model mismatch and modeling errors.

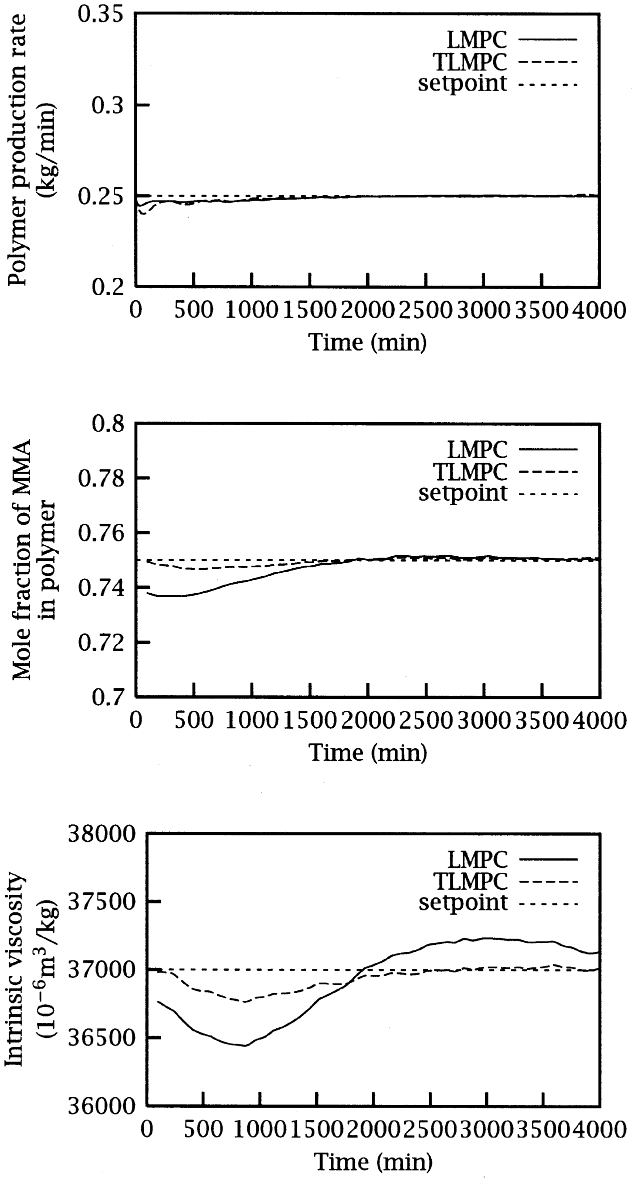


Figure 10. Closed-loop outputs (controller robustness for measured disturbances).

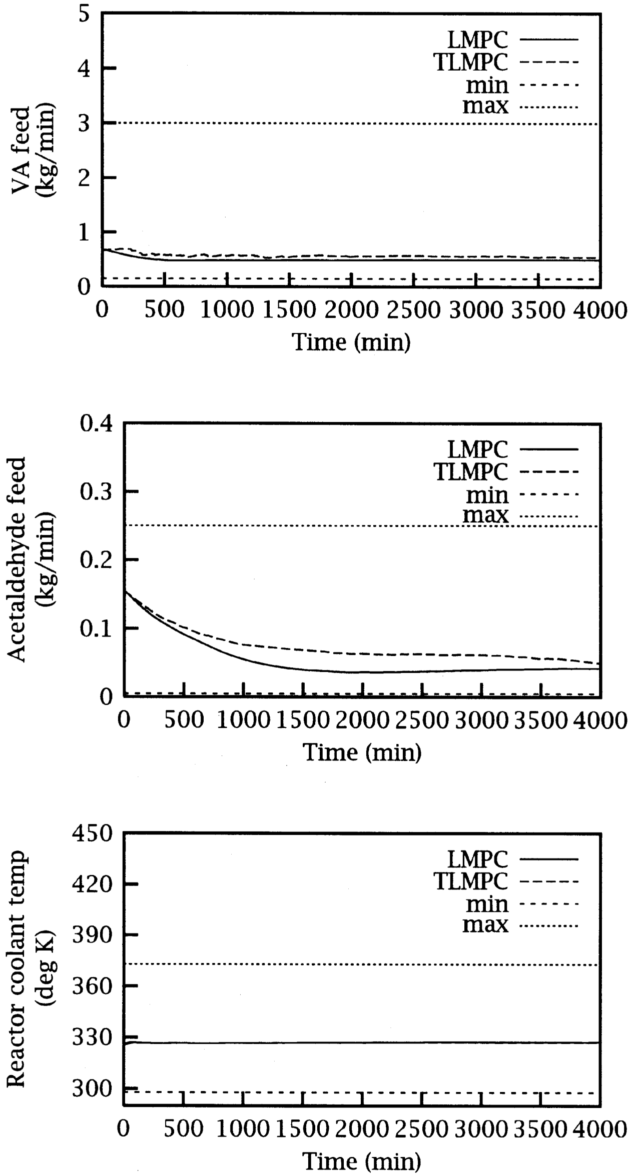


Figure 11. Closed-loop inputs (controller robustness for measured disturbances).

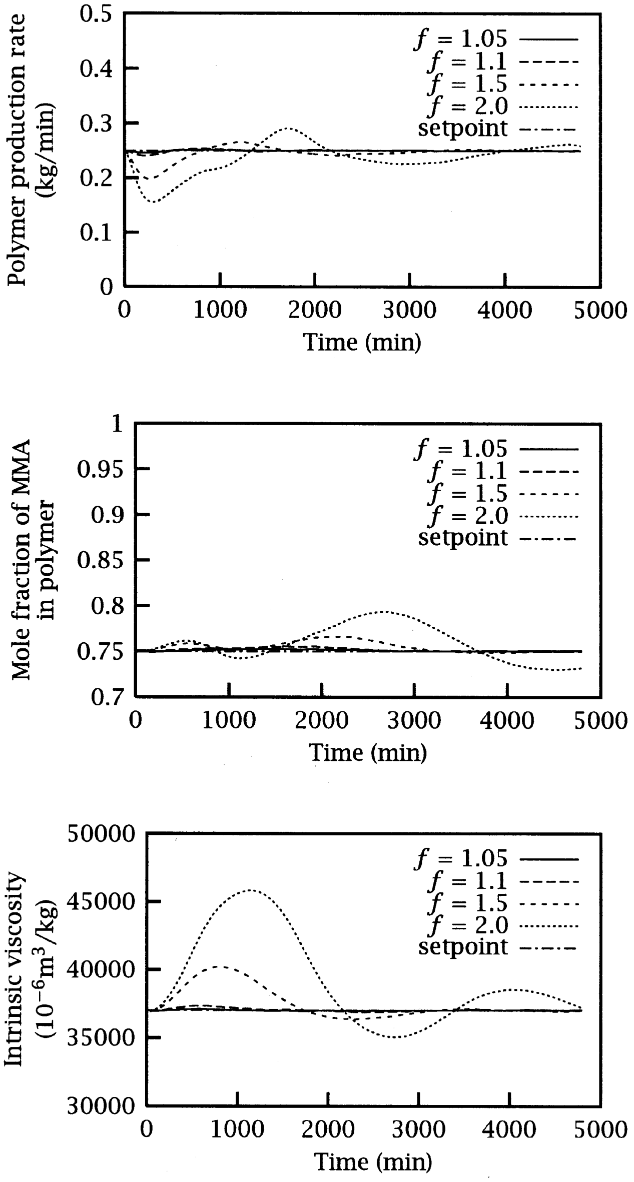


Figure 12. Closed-loop outputs (plant-model mismatch for kinetic parameters and initial states).

Conclusions

The proposed feedback controller (TLMPC) is able to attain varying process objectives for a typical process in the polymer industry without using a complete NMPC. A TLMPC requires the calculation of targets using a nonlinear model, and then uses the targets to evaluate the appropriate linear model. The dynamic regulation problem is solved using the linear model thereby ensuring that it is solved efficiently as compared to evaluating controller moves for nonlinear dynamic regulation A TLMPC performs better than a fixed linear model predictive controller for different operating conditions. The TLMPC controller shows good robustness with respect to kinetic parameter error, incorrect initial state estimates, and different measured disturbances.

The process objectives specified by product grades have started changing more rapidly due to the changing needs of the customer and the desire of the manufacturers to find new uses for the polymers. This market scenario imposes new grade requirements for the product, thereby making it harder to maintain a set of linear models that can attain the required process objectives; maintaining a set of linear models

Table 7. Plant-Model Mismatch for Pre-exponential Factors of Propagation Rate Constants

	Model (m^3/kmol·s)	Plant (m^3/kmol·s)
A_{paa}	$f\cdot 3.207\times 10^6$	3.207×10^6
A_{pab}	$f\cdot 1.233\times 10^5$	1.233×10^5
A_{pba}	$f\cdot 2.103\times 10^8$	2.103×10^8
A_{pbb}	$f\cdot 6.308\times 10^6$	6.308×10^6

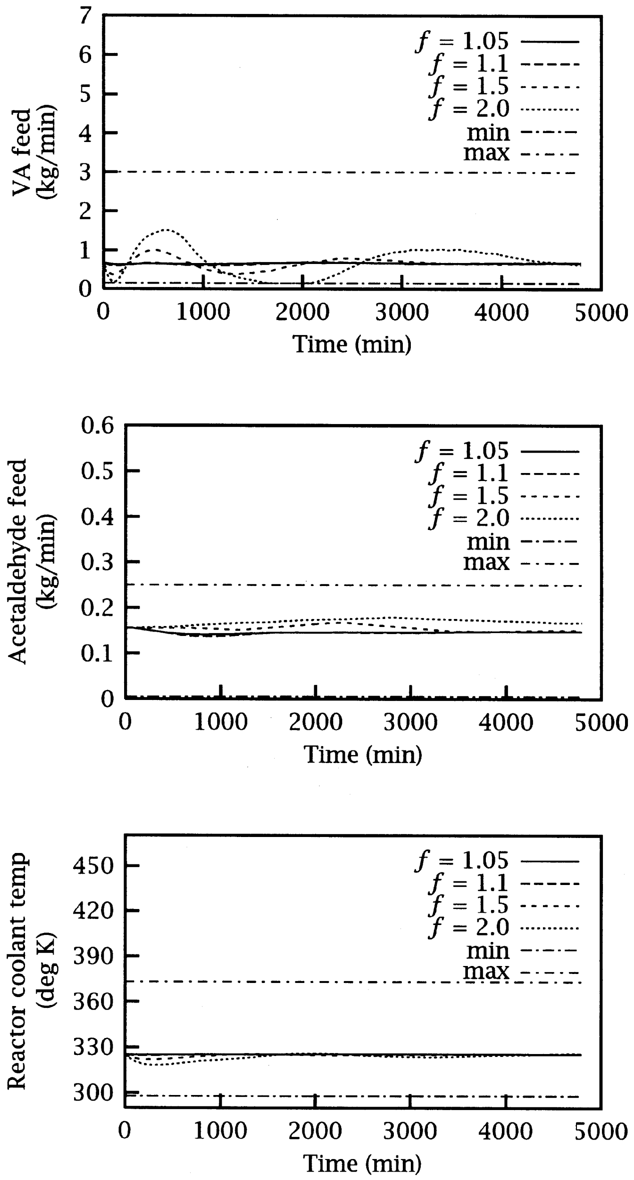


Figure 13. Closed-loop inputs (plant-model mismatch for kinetic parameters and initial states).

becomes particularly difficult for operation with significant measured disturbances.

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