Nonlinear Model Predictive Control of End-Use Properties in Batch Reactors

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Significant economic benefits can be derived from reducing the variability of product properties around their specification targets in batch reactors. End-use properties in batch reactors using dynamic models that relate the operating conditions to the end-use properties are related through a static nonlinear model to the molecular properties of the product and are controlled directly. Their values are required to lie in a desired target region in the property space. The control of end-use product properties is formulated as a nonlinear model predictive control problem, and an efficient numerical technique using successive linearizations is utilized for the solution. A parameter adaptive extended Kalman filter is used for state estimation of the molecular properties that are not measured directly. The emulsion polymerization of styrene is chosen as a case study, and an approximate mathematical model is developed. The model is detailed enough to predict the end-use properties, which depend on both the molecular weight and particle-size distributions of the product. The end-use product properties, such as tensile strength and melt index, are controlled in desired target regions by adding a monomer and a chain transfer agent, as well as the flow rate of the coolant, as the manipulated variables.

Introduction

There is a strong incentive in controlling the end of the batch product property values so as to minimize the variability in product quality. This is especially true for products where there is no possibility of downstream blending to achieve the desired specifications. The manipulation of product properties by post processing is an impossible or extremely expensive task. This motivates the use of model-based control techniques that aim to control the end-use properties explicitly during the batch. Nonlinear model predictive control (NLMPC) provides an attractive framework to handle the time varying and highly nonlinear batch processes, where the control actions need to be taken early on the batch to be effective. Furthermore, batch processes are characterized by significant model uncertainties and infrequent and limited measurements, which prompts the use of extended Kalman filter for state and disturbance estimation.

The existing practice aims to control the molecular properties at the end of the batch, and, thus, control the end-use properties indirectly (Mutha et al., 1997; Tsen et al., 1996; Russel et al., 1998). In the applications to emulsion polymerization processes, the molecular weights, composition and/or particle-size averages can be chosen as the control variables (Dimitratos et al., 1994). By controlling these molecular properties, the related end-use properties are indirectly controlled at their set points. The above approach has certain drawbacks. First and foremost, there may not be enough degrees of freedom to control the specified molecular properties at the desired fixed targets, that is, the target set point may not be reachable. Furthermore, in many cases, it is not necessary to maintain all the molecular or end-use properties at a specific target value for achieving the desired product functionality. Considering these reasons, it is preferable to control the end-use properties explicitly. In the present work, a static relationship between molecular and end-use properties is appended to the fundamental dynamic model of the process (Figure 1). With the use of such a model, a methodology for control of the end-use properties within bounded regions is developed.

The defined on-line control problem consists of finding the future inputs that would bring the process outputs to the de-

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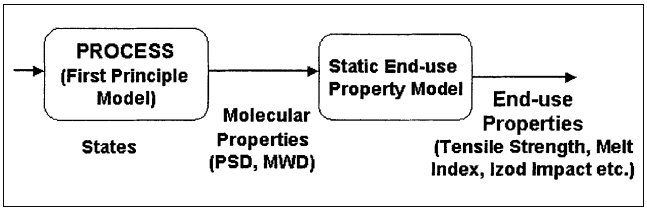

Figure 1. Static end-use property model with first-principles dynamic model for direct control of end-use properties.

sired target region, subject to the constraints on the manipulated variables and their rate of change. This formulation of the end-use property control necessitates the introduction of suitable techniques for the solution of the nonlinear model predictive control problem. The possible options to solve this problem can be broadly classified into two, the primal methods and the penalty methods, of which the former is more preferred due to the reduced computational demands (Elwakeil and Arora, 1995). One primal approach is to formulate this as a min-max problem, which is then solved using sequential quadratic programming (SQP). A more efficient technique uses successive linear approximation of the target region to find the appropriate direction using a quadratic programming subproblem. This method is found to perform better than the min-max method in computational demands and convergence properties.

The calculation of all future control moves at each time is computationally demanding in batch model predictive control. Input parameterization is a suitable procedure to circumvent this difficulty (Goh and Teo, 1988). The inputs for the rest of the batch time are parameterized in terms of a few parameters using Lagrange polynomials. With this approach, the decision variables in the control problem are the reduced number of parameters characterizing the assumed polynomial rather than the input values at each sample interval. Thus, the computational demands are reduced without sacrificing the advantages of a control horizon extending to the end of the batch.

Table 1. Parameters Used for Modeling Emulsion Polymerization of Styrene

Parameter	Value
k_d	$4.5\times10^{16}\exp(-140{,}200/\mathrm{RT})\mathrm{s}^{-1}$
k_{p0}	$2.17\times10^{7}\exp(-3{,}905/T)\mathrm{dm}^3/\mathrm{mol/s}$
υ_{fmc}	0.0383
α	1.85
k_{adj}	1.05×10^{6}
ρ_m	$923.6-0.887T_c$ g/dm^3
ρ_p	$1{,}050.1-0.621T_c$ g/dm^3
k_{tw}	k_{t0}
k_{t0}	$6.2\times10^{9}\exp(-1{,}747/T)$ dm^3/mol/s
f_d	0.7
k_{fm}	2.7×10^{-5} dm^3/mol/s
D_w	5.0×10^{-6}cm^2/s
D_p	5.0×10^{-6}cm^2/s
β	0.3
m_d	1,600
k_{cta}	$17.0\times10^{-4}k_{p0}$

Table 2. Parameters for Discretized PSD of the Seed and Other Process Parameters

No. of Particles	Value	Swollen Dia. of Seed	Value
N_1	20×10^{17}	D_1	90 nm
N_2	80×10^{17}	D_2	150 nm
N_3	20×10^{17}	D_3	250 nm

Batch processes are also characterized by significant model uncertainties and a limited number of noisy measurements. For emulsion polymerization processes of interest here, important molecular properties related to molecular weight averages are not measurable on-line. Furthermore, in most cases, the end-use properties that need to be controlled are not measured *directly* on-line. A state estimator (extended Kalman filter) is useful under these circumstances to estimate the important unmeasured properties using the available measurements. A parameter adaptive extended Kalman filter (PAEKF) is used where the parameters are added as biases to the key states in the system. The process noise covariance matrix for the filter is obtained by a procedure utilizing the known parameter covariance matrix proposed by Valappil and Georgakis (2000). For the time varying and nonlinear batch process like the emulsion polymerization process studied here, the tuning technique significantly reduces the time requirements for the EKF implementation (Valappil and Georgakis, 1999, 2000).

The emulsion polymerization process for styrene is chosen as an example to study the effectiveness of the above methodologies. The main motivation to study this process is that the control of end-use properties is important in emulsion products and these processes are inherently very nonlinear. The use of the population balance approach to modeling emulsion processes involves the solving of integro-partial differential equations to obtain the desired distributions. Using these detailed models on-line for predictive control can be a very computationally demanding task. An alternate approach is used here, and this involves treating a reduced number of representative populations, which are modeled individually. A mathematical model previously developed for bi-disperse population by Liotta (1996) is used for modeling the process. For modeling the molecular weight distribution, moment transformations are utilized. This reduces the model to the scale where they can be used on-line for state estimation and model-based control. Properties like tensile strength and melt index can be related to the molecular properties of styrene. The manipulated variables available for control are the addition rate of the monomer, and the chain transfer agent, as well as the flow rate of coolant. Process-model mismatch in the form of parametric uncertainties and disturbances commonly encountered in these processes are used to test the proposed methodology.

NLMPC of Batch End-use Properties in Target Regions

Many of the features of batch processes such as variability, high nonlinearities and plant-model mismatch, make batch control difficult. For the control actions to be effective, these actions should be taken early in the batch. NLMPC provides

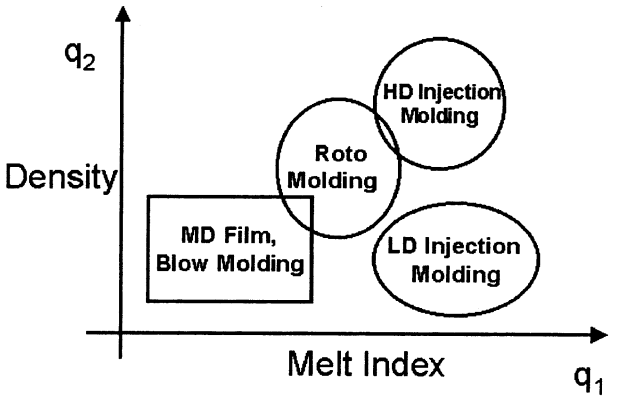

Figure 2. Target regions for different product specifications in polyethylene.

a suitable framework for handling the time varying nature of the batch process, while respecting the process and equipment constraints.

The nonlinear model predictive control problem studied here is different from the previous studies in that the objective is to bring the end-use properties to a desired target region rather than a target point. The formulation introduced uses the shrinking horizon (SHMPC) methodology, but with a different end-of-batch objective. Efficient optimization methods are used, which improve both the computational aspects and the convergence of the controller. The control methodology presented in this article considers the nonlinear nature of a batch process, is multivariable, and can handle constraints on the states, outputs and inputs.

Target regions for end-use property control

The desired values of the end-use properties are determined by the intended use of the product. In many practical situations, the interest is in ensuring that the particular product properties lie in a certain range of specifications. An example is the case of polyethylene where the desired properties, melt index and the density, need to be maintained within certain regions to produce a product with the desired properties (Figure 2). A more general approach to the problem is presented here, whereby the end-use properties are required to lie in a target range. There are other relevant reasons to consider the target region for end-use properties. The alternative to considering the target region is to select a point within the specification region and try to control the properties at this set point. This point would prerferably be at the center of the region to reduce the effect of disturbances or model error. However, the drawback to this approach is that the selected set point may not be reachable with the available manipulated variables. The reachable regions are not known *a priori* (especially for new products), and, thus, the reachability of the desired set point cannot be quickly ascertained without extensive simulations. Furthermore, the reachable regions change with the character and the magnitude of the disturbance presently affecting the process. In the case that the desired set point is not reachable, the nonlinear model predictive controller would guide the process so that the product end-use properties are closest to the set point. This does not necessarily satisfy the product specification constraints, thus resulting in a wasted batch.

The end-use properties can be related to the molecular properties of the product at the end of the batch using the available static model. These models could be obtained from fundamental knowledge or could be data-driven. Data-driven techniques like partial least squares (PLS) and artificial neural networks (ANN) can be used to fit a nonlinear model between the two sets of variables. Also, group contribution methods have found an application in relating to the properties of polymer products (Krevlen, 1990). The molecular properties are functions of states at the end of the batch. Thus, a nonlinear functional relationship between the end-use product properties and the state values can be obtained as given below

$$z_f = \boldsymbol{q}(\boldsymbol{x}_f) \tag{1}$$

where z_f is a vector of quality variables, $\boldsymbol{x}_f$ are the states of the system at the end of the batch, and $\boldsymbol{q}$ denotes the functional relationship between the states and the properties.

The requirement that the end-use properties lie in the target region Q can then be transformed into a set of quality constraints in the form of nonlinear equalities and inequalities. A target region Q, of arbitrary shape, can be represented this way

$$\boldsymbol{q}(\boldsymbol{x}_f) \subseteq Q \Leftrightarrow \{\boldsymbol{q}_1(\boldsymbol{x}_f) \le \boldsymbol{0};\ \boldsymbol{q}_2(\boldsymbol{x}_f) = \boldsymbol{0}\} \tag{2}$$

This mixed sex of equalities and inequalities can be converted to a set of inequalities using the absolute values of equalities

$$\{\boldsymbol{q}_1(\boldsymbol{x}_f) \le \boldsymbol{0},\ |\boldsymbol{q}_2(\boldsymbol{x}_f)| \le \boldsymbol{0}\} \tag{3}$$

The representation of a target region in terms of states can be given as

$$\{\boldsymbol{r}(\boldsymbol{x}_f) \le \boldsymbol{0}\} \tag{4}$$

Here, $\boldsymbol{r}(\boldsymbol{x}_f)$ denotes the target region constraints in terms of the end of the batch states.

It needs to be noted that different types of end-use properties can be represented this way. For example, in the simplest case of interval constraints, each constraint would be represented by

$$q_i(\boldsymbol{x}_f) - q_u \le 0;\ q_l - q_i(\boldsymbol{x}_f) \le 0 \tag{5}$$

where q_l and q_u represent the lower and upper bounds on the quality variable, q_i. An example of the target regions for control purposes is shown in Figure 2.

Reachable Regions for End-use Properties. The concept of reachability is important in the framework of end-use property control, since the interest is in attaining a target set for the properties at the end of the batch. The ability of the system to reach this target region with the available inputs has

implications on the success of the control strategy. For a nonlinear continuous time system, a reachable region is defined as the envelope of states, $\boldsymbol{x}(t_f)$ that can be reached from $\boldsymbol{x}(t_0)$ through the use of all available control actions. For end-use properties, the reachable region can be defined as the value of the end-use properties that can be attained starting from a certain initial condition, and by considering the constraints on the input variables that define a set of recipes that can be followed. The reachable region is difficult to determine analytically and tends to be quite nonconvex, as the system is nonlinear and the desired product properties are calculated at the end of the batch. Furthermore, the reachable regions are also affected by the possible disturbances that could enter during the batch.

For the cases where the target region of the end-use properties is not completely reachable, the approach proposed in this article is especially valuable. In these cases, selecting a specific set point within the reachable region for the control of the end-use properties is very difficult, since the reachable regions are not known *a priori*, and can change. Consideration of the target region itself in a control calculation can improve the convergence properties and reduce the computational demands of the controller.

Nonlinear model predictive control formulation

Consider the nonlinear batch process in the state space form as shown below

$$\frac{d\boldsymbol{x}}{dt} = \boldsymbol{f}(\boldsymbol{x}, \boldsymbol{u}, \boldsymbol{p}, \boldsymbol{d}, t) \tag{6}$$

Here $\boldsymbol{x}$ denotes the system states, $\boldsymbol{u}$ the manipulated variables, $\boldsymbol{p}$ the model parameters, and $\boldsymbol{d}$ the disturbances entering the system. The covariance matrix for the parameters is also assumed to be available from the parameter estimation step. The model parameters are assumed to be normally distributed with the mean and covariance as given below.

$$E[\boldsymbol{p}] = \boldsymbol{p}_{\text{nom}}$$
$$E\left[(\boldsymbol{p} - \boldsymbol{p}_{\text{nom}})(\boldsymbol{p} - \boldsymbol{p}_{\text{nom}})^T\right] = \boldsymbol{C}_p \tag{7}$$

where $\boldsymbol{p}_{\text{nom}}$ denotes the nominal parameter values and $\boldsymbol{C}_p$ is the parameter covariance matrix.

At each controller execution, the objective is to determine the current and future moves of the manipulated variable that would bring the states at the end of the batch to the desired target region. The prediction horizon is, thus, varying with time and becomes smaller as the batch progresses. This is termed as shrinking horizon model predictive control, and has been used widely for batch model predictive control (Thomas et al., 1997; Liotta, 1996; Liotta et al., 1997b). In the commonly accepted formulation of SHMPC problem, the minimization of an objective function that represents the deviation from the end of the batch set point is considered. Nonlinear programming methods that minimize this objective function are used to calculate the control actions. This formulation can be given as

$$\begin{aligned} \min_{\boldsymbol{u}} \quad & \| z_f - z_f^{sp} \| \\ \text{subject to } & \frac{d\boldsymbol{x}}{dt} = \boldsymbol{f}(\boldsymbol{x}, \boldsymbol{u}, \boldsymbol{p}, \boldsymbol{d}, t) \\ & \boldsymbol{x}(t=0) = \boldsymbol{x}_0 \\ & \boldsymbol{h}(\boldsymbol{x}(\tau)) \le \boldsymbol{0};\ t \le \tau \le t_f \\ & \boldsymbol{A}\boldsymbol{u}(\tau) \le \boldsymbol{b};\ t \le \tau \le t_f \end{aligned} \tag{8}$$

where z_f represents the properties of interest and z^{sp} is the set point for these properties. In some cases, a weighted objective function is utilized to give more importance to one property than the other. The term $\boldsymbol{h}(\boldsymbol{x})$ represents the nonlinear constraints on the states. The last term denotes the constraints on the manipulated variables and their rates of change, which can be represented as linear inequalities in the manipulated variables $\boldsymbol{u}$.

The above formation is suited for a case where there is a target set point at the end of the batch. For the reasons stated previously, the interest here is for the end-use properties to lie inside a region at the end of the batch. The control problem at each sampling time is therefore formulated as finding inputs such that the process states at the end of the batch satisfy the constraints defining the desired region

$$\begin{aligned} & \{\text{Find } \boldsymbol{u} \ni \boldsymbol{r}(\boldsymbol{x}_f) \le \boldsymbol{0}\} \\ & \frac{d\boldsymbol{x}}{dt} = \boldsymbol{f}(\boldsymbol{x}, \boldsymbol{u}, \boldsymbol{p}, \boldsymbol{d}, t) \\ & \boldsymbol{x}(t=0) = \boldsymbol{x}_0 \\ & \boldsymbol{h}(\boldsymbol{x}(\tau)) \le \boldsymbol{0};\ t \le \tau \le t_f \\ & \boldsymbol{A}\boldsymbol{u}(\tau) \le \boldsymbol{b};\ t \le \tau \le t_f \end{aligned} \tag{9}$$

Since the states are determined by the control actions during the batch, the end-point inequalities can be represented in terms of the inputs, $\boldsymbol{u}$ as shown below

$$\{\boldsymbol{r}(\boldsymbol{x}_f) \le \boldsymbol{0}\} \Leftrightarrow \{\boldsymbol{g}(\boldsymbol{u}) \le 0\} \tag{10}$$

The nonlinear model predictive control problem can, thus, be formulated as solving these set of nonlinear inequalities subject to the constraints

$$\begin{aligned} & \{\text{Find } \boldsymbol{u} \ni \boldsymbol{g}(\boldsymbol{u}) \le \boldsymbol{0}\} \\ & \frac{d\boldsymbol{x}}{dt} = \boldsymbol{f}(\boldsymbol{x}, \boldsymbol{u}, \boldsymbol{p}, \boldsymbol{d}, t) \\ & \boldsymbol{x}(t=0) = \boldsymbol{x}_0 \\ & \boldsymbol{h}[\boldsymbol{x}(\tau)] \le \boldsymbol{0};\ t \le \tau \le t_f \\ & \boldsymbol{A}\boldsymbol{u}(\tau) \le \boldsymbol{b};\ t \le \tau \le t_f \end{aligned} \tag{11}$$

A few things need to be noted about the above formulation for end-use property control. The inequalities that represent

the target region are nonlinear functions of the inputs. Also, in most cases, the inequalities tend to be nonconvex functions of the inputs. Thus, the solution of the above problem is very difficult, and the existence of a solution cannot be guaranteed. The above formulation considers the constraints on the inputs to be linear. This is quite realistic as these are mostly hard bounds on the absolute values of the inputs or their rate of change. The constraints on the states of the system or other variables during the batch (path constraints) can also become important in certain applications. An example is the reactor temperature, which might often need to be maintained within certain limits during the batch. These path constraints are also functions of input values and are handled by augmenting them along with the target region constraints for the solution. They are not explicitly considered separate in the discussion of solution methods.

Input Parameterization in Model Predictive Control. The implementation of the NLMPC involves the calculation of the necessary manipulated variables into the future. It is computationally demanding to determine all the future control moves using nonlinear optimization. First, the dimensions of the problem would be very large. Furthermore, the convergence properties suffer with the increase in the size of the problem. The solution is to parameterize the inputs with the appropriate, reduced number of parameters. Parameterization is implicitly used in many commercial model predictive control packages, in the form of input blocking or other forms. In batch optimal control, the method of orthogonal collocations has also been used for performing input parameterization.

A Lagrange polynomial approximation approach is used here for input parameterization, whereby the parameters are the values of the inputs at certain future time intervals. Manipulated variables at other time instants can be represented in terms of the parameters as

$$u(t) = \sum_{i=1}^{n_{\text{par}}} \upsilon_i \frac{\prod (t - t_j)}{\prod (t_i - t_j)} \quad j = 1, 2, \ldots, n_{\text{par}} \quad j \neq i \quad (12)$$

where n_{par} is the number of parameters. This gives the value of input u at any time t in terms of the parameters υ_i that are the values of $u(t)$ at time t_i. For a discrete controller implementation, a constant value for $u(t)$ during the sample interval is obtained by averaging the $u(t)$ values. The vector of averaged values for future instants $\boldsymbol{u}$ can be given as linear combination of the parameters υ_i

$$\boldsymbol{u} = \boldsymbol{\Phi}\boldsymbol{v} \quad (13)$$

where $\boldsymbol{u}$ denotes the input values at the future sample intervals, $\boldsymbol{v}$ represents the parameters characterizing the input profile and $\boldsymbol{\Phi}$ is a matrix of appropriate size. Here, the matrix $\boldsymbol{\Phi}$ remains unaltered while the calculations at a particular time instant are performed. The controller determines the values of the parameter vector υ, which is the used in Eq. 13 to find the future input values.

State estimation

The successful application of a model based control is very dependent on the proper estimation of the present and future states. This is especially true for the control of the end-of-batch properties, as the properties are not measured on-line and the prediction horizon is substantially long, especially at the beginning of the batch. This prompts the use of state estimators to estimate on-line the unmeasured states and disturbances while accounting for the process-model mismatch. The recursive nature of the EKF and its ability to account for the model uncertainties in a transparent way makes the EKF a suitable choice for this application.

The performance of the EKF depends on the validity of the model. If the model predictions are highly biased, the EKF would perform poorly, giving estimates that are biased or highly uncertain. A solution to this is the use of parameter adaptive extended Kalman filter, where certain fictitious parameters are estimated along with the states using the available measurements. In the PAEKF, the system states are augmented with adapted parameters θ to form the augmented state space system as shown below

$$\frac{d}{dt}\begin{pmatrix} \boldsymbol{x} \\ \theta \end{pmatrix} = \begin{pmatrix} \boldsymbol{f} \\ \boldsymbol{0} \end{pmatrix} + \begin{pmatrix} \boldsymbol{w} \\ \boldsymbol{w}_\theta \end{pmatrix} \quad (14)$$

The covariance matrix for the augmented system and the augmented state space matrix are appropriately given

$$\boldsymbol{P}_a = \begin{bmatrix} \boldsymbol{P}_x & \boldsymbol{P}_{x\theta} \\ \boldsymbol{P}_{\theta x} & \boldsymbol{P}_\theta \end{bmatrix}; \quad \boldsymbol{A}_a = \begin{bmatrix} \dfrac{\delta \boldsymbol{f}}{\delta x} & \dfrac{\delta \boldsymbol{f}}{\delta \theta} \\ \boldsymbol{0} & \boldsymbol{0} \end{bmatrix} \quad (15)$$

The successful application of the EKF is dependent on the specification of values of the process noise covariance matrix $\boldsymbol{Q}$. This value determines the confidence the filter places on the model. A methodology for systematically specifying $\boldsymbol{Q}$ proposed in Valappil and Georgakis (2000) is used here. This technique uses a linearization approach to translate the information about model uncertainty to $\boldsymbol{Q}$. For this, the parameters in the model are assumed to be normally distributed with the mean as the nominal value of the parameter and the covariance from the parameter covariance matrix. The process noise convariance matrix is time varying and calculated on-line as

$$\boldsymbol{Q} = \boldsymbol{J}_p(t)^T \boldsymbol{C}_p \boldsymbol{J}_p(t) \quad (16)$$

where $\boldsymbol{J}_p$ is the sensitivity matrix with respect to the parameters. The value $\boldsymbol{Q}$ obtained above is used in the EKF equation for state covariance propagation.

Solution Methods for the NLMPC Problem

The nonlinear model predictive control of end-use properties needs to be solved on-line at each time step with the estimated states from EKF. Hence, the computational efficiency and the convergence properties of the numerical methods that are used for the solution becomes important. The existing formulations of batch MPC use the approach of the minimization of a weighted objective function. For the control of end-use properties, the target region constraints are of primary importance. The use of optimization methods

tailored to the problem under investigation is important for the successful application of NLMPC. Numerical algorithms to find the initial feasible points for constrained optimization problems, can be utilized for the solution of the end-use property control problem presented in the subsection titled "Nonlinear model predictive control formulation." A comparison of a few methods for finding initial feasible points in nonlinear programming is given in Elwakeil and Arora (1995). It is recognized that this problem is as hard as solving the nonlinear optimization problem. A broad classification of this problem can be done in two groups, penalty and primal methods. The penalty methods use a quadratic penalty with all the constraints together and use it as an objective function for unconstrained minimization. Their performance is dependent on the penalty parameters and the strategy for updating the parameters. Primal methods consider the constraints directly in the algorithm and have been shown to be computationally superior to the penalty methods (Elwakeil and Arora, 1995).

Two different solution methods that belong to the primal group of algorithms and suitable for the end-use property control are studied here. An accepted approach, used frequently for such problems, is to use to min-max problem formulation. This involves the use of conventional nonlinear programming techniques for the solution, and is studied first. Newton type approaches have also been used for solving these types of problems (Mayne and Sahba, 1985; Mayne et al., 1981; Chen and Kostreva, 1999). The second method utilized here is a Newton type based on the successive linearization of the feasible ratio with a modification to improve the convergence properties and the speed of the algorithm. These methods are described in the following sections.

Min-max method

The NLMPC problem can be formulated as a min-max problem by treating the target region constraints as objective functions to reduce their values until the feasible solution is obtained. The constraint with a maximum value is treated as the objective to be minimized. This min-max formulation is given by

$$\begin{aligned} &\min_{\boldsymbol{u}} - \max_{i} \left\{ r_i(\boldsymbol{x}_f),\ i = 1, 2, \ldots, m \right\} \\ &\frac{d\boldsymbol{x}}{dt} = f(\boldsymbol{x}, \boldsymbol{u}, \boldsymbol{p}, \boldsymbol{d}, t) \\ &\boldsymbol{x}(t = 0) = \boldsymbol{x}_0 \\ &\boldsymbol{A}\boldsymbol{u}(\tau) \le \boldsymbol{b};\ t \le \tau \le t_f \end{aligned} \tag{17}$$

where m denotes the number of inequalities defining the end-point constraints (Eq. 4). Different methodologies exist in the literature to solve min-max problems. A very efficient solution technique is the sequential quadratic programming (SQP) method. Here, the inequalities are treated as objective functions and their values and their gradients are used in SQP. Many of the numerical routines that solve min-max problems utilize this approach, which is also used for the min-max solution in this work.

The solution to this problem involves a quadratic approximation to the objective function with a quasi-Newton Hessian update. This increases the computational demands without much added benefit, as we are interested in getting a solution that satisfies the target region constraints. Also, the min-max formulation results in solving a nondifferentiable optimization problem, which is more difficult than a differentiable optimization problem. This prompts the consideration of alternate methods that are more tailored to the problem of controlling the end-use properties within a region.

Successive target region linearization method

Newton type methods are widely used for the solution of nonlinear equations and inequalities. These methods are characterized by good computational speed and convergence properties. A Newton type approach for the solution of nonlinear inequalities that successively linearizes the nonlinear problem was presented by Mayne and Sahba (1985). The method was shown to be quite efficient in solving nonlinear inequalities, and the convergence properties are well established (Sahba, 1987). This approach with suitable modification is utilized here for calculation of the controller moves.

As presented in the subsection titled "Nonlinear model predictive control formulation," the target region constraints can be represented as nonlinear inequalities in the inputs for the rest of the batch. The maximum of this set of inequalities is defined as

$$\psi(\boldsymbol{u}) = \max\left\{ g_j(\boldsymbol{u}) / j = 1, 2, \ldots, m \right\} \tag{18}$$

and the first-order approximation to the above function given by $\hat{\psi}(\boldsymbol{u})$

$$\hat{\psi}(\boldsymbol{u}) = \max\left\{ g_j(\boldsymbol{u}) + \nabla g_j(\boldsymbol{u}) \Delta \boldsymbol{u} / j = 1, 2, \ldots, m \right\} \tag{19}$$

where $\Delta \boldsymbol{u}$ denotes the change in inputs. The Newton type approach is based on linearizing the target region of interest successively until a solution that satisfies all the inequalities is obtained. At each iteration, the values of the target region constraints and their gradients are used to define a quadratic program (QP) that finds a Newton step towards the target region. Utilizing the above Newton step, the decision variables can be updated until all the target region constraints are satisfied.

The conventional Newton type approach based on successive linear approximation fails when the linearization makes the problem infeasible. This can happen if the initial guess is far from the solution or when the target region constraints are very stringent, thus resulting in an infeasible quadratic program. A modification to the above procedure was reported in Mayne and Sahba (1985) and is suitable to handle this drawback. Another motivation for the same modification is the possible improvement in the efficiency of the algorithm. The modification is such that the new vector $\boldsymbol{u} + \Delta \boldsymbol{u}$ lies in the interior of the linearized feasible region, if there is a feasible solution to the same. Otherwise, the linearized constraints are relaxed to accommodate a temporary feasible solution. In this case, the direction vector is obtained as a solution to the quadratic program given as follows:

$$\min_{\boldsymbol{u}} \{\boldsymbol{\Delta u}'\boldsymbol{\Delta u}\}$$
$$\boldsymbol{g}(\boldsymbol{u}) + \nabla \boldsymbol{g}(\boldsymbol{u})\boldsymbol{\Delta u} \le \hat{\psi}_{\epsilon}^{0}(\boldsymbol{u})$$
$$\boldsymbol{A u} + \boldsymbol{A}\boldsymbol{\Delta u} \le \boldsymbol{b} \tag{20}$$

where $\boldsymbol{\Delta u}$ denotes the step, $\nabla \boldsymbol{g}$ denotes the gradient of the target region constraints with respect to the input and the value of $\hat{\psi}_{\epsilon}^{0}(\boldsymbol{u})$ is obtained as

$$\hat{\psi}_{\epsilon}^{0}(\boldsymbol{u}) = \max\left\{\hat{\psi}^{0}(\boldsymbol{u}), -\boldsymbol{\epsilon}\right\} \tag{21}$$

Here, $\boldsymbol{\epsilon}$ is chosen by the user and $\hat{\psi}^{0}(\boldsymbol{u})$ is estimated from a calculation as below

$$\hat{\psi}_0(\boldsymbol{u}) = \min_{\boldsymbol{u}} - \max_{j} \left\{g_j(\boldsymbol{u}) + \nabla g_j(\boldsymbol{u})\boldsymbol{\Delta u}/j = 1, 2, \ldots, m\right\}$$
$$\| \boldsymbol{\Delta u} \|_{\infty} \le L$$
$$\boldsymbol{A u} + \boldsymbol{A}\boldsymbol{\Delta u} \le \boldsymbol{b} \tag{22}$$

where L is a suitably chosen positive number. The norm constraint is utilized to avoid any unbounded solution. The above min-max problem (Eq. 22) is linear and can be solved as a linear programming problem

$$\min_{\boldsymbol{u},c} \{c\}$$
$$\boldsymbol{g}(\boldsymbol{u}) + \nabla g(\boldsymbol{u})\boldsymbol{\Delta u} \le c$$
$$\boldsymbol{A u} + \boldsymbol{A}\boldsymbol{\Delta u} \le \boldsymbol{b}$$
$$\| \boldsymbol{\Delta u} \|_{\infty} \le L \tag{23}$$

where the variable c is a scalar. If $c > 0$, the linearized constraint set is infeasible; hence, this value is used in Eq. 20 for $\hat{\psi}_{\epsilon}^{0}$. Otherwise, the value from Eq. 21 is utilized, which approximately selects the amount to move the linearized constraints (which are feasible) to the interior of the target region.

Once the direction vector $\boldsymbol{\Delta u}$ is selected, an adequate step is taken in this direction to reduce the values of the inequality functions. An Armijo rule is used to select the appropriate move (Mayne and Sahba, 1985). For this, it is required that the step-size be such that the actual reduction in ψ be at least half the estimated reduction. The estimated reduction is based on the linear approximation to the constraints (Eq. 25). The procedure is repeated till no significant improvement in the function ψ is obtained, which can be represented as

$$\| \psi(\boldsymbol{u}_{k+1}) - \psi(\boldsymbol{u}_k) \| \le \eta \tag{24}$$

where η is the tolerance criterion for stopping the algorithm and $\boldsymbol{u}_k$ denotes the inputs at iteration k. The complete algorithm for the control move calculation is given below.

Algorithm for control variable calculation.

(1) Inputs

$\boldsymbol{u}_0$, $\boldsymbol{\epsilon}' \in (0,1)$, $L \ge 1$, $\beta \in (0,1)$ Define set $S = \{1, \beta, \beta^2, \ldots\}$

(2) Set $i = 0$, $\boldsymbol{\epsilon}_0 = \boldsymbol{\epsilon}'\psi(\boldsymbol{u}_0)$

(3) Linear programming step: compute $\hat{\psi}_0(\boldsymbol{u}_i)$ (Eqs. 22 and 21)

(4) Quadratic programming step: compute $\boldsymbol{\Delta u}_i$ (Eq. 20)

(5) Compute $\lambda_i = \lambda_{\epsilon i}(\boldsymbol{u}_i)$, the largest λ in $\boldsymbol{S}$ such that

$$\psi(\boldsymbol{u}_i + \lambda\boldsymbol{\Delta u}_i) - \psi(\boldsymbol{u}_i) \le \lambda\theta(\boldsymbol{u}_i, \boldsymbol{\Delta u}_i)/2 \tag{25}$$

where

$$\theta(\boldsymbol{u}_i, \boldsymbol{\Delta u}_i) = \hat{\psi}(\boldsymbol{u}_i, \boldsymbol{\Delta u}_i) - \psi(\boldsymbol{u}_i) \tag{26}$$

(6) Set $\boldsymbol{u}_{i+1} = \boldsymbol{u}_i + \lambda\Delta\boldsymbol{u}_i$

Go to step 3 if convergence conditions (Eq. 24) are not satisfied.

The above algorithm is very suitable for the problem under consideration. First, the modification in step 3 ensures that the method does not stop if there is no solution to the linearized problem. It is quite possible that linearization of the problem can destroy the feasibility in some cases. As is clear from Eq. 21, the linear constraints on manipulated variables are also considered at each iteration of the algorithm. Thus, once the initial guess of inputs is feasible, all the subsequent values are feasible. This is a desirable property as it ensures that the input values that do not make any sense do not occur during the iteration. An example is the flow rate of a monomer, which cannot be negative. Previous studies on using the iterative calculation of step response coefficients, by perturbing the model, are similar to the above method. The advantages of this approach compared to the previous studies are that it is multivariable, constrained and can calculate multiple moves into the future.

Different conditions can be used for stopping the algorithm. One way is to stop once all the constraints are satisfied, that is, when $\psi(u)$ equals zero. However, such a policy would be susceptible to disturbances and uncertainties that can enter the batch, because the properties are controlled at the boundaries of the target region. The stopping criterion used for this algorithm is based on the minimum of the constraint that is under maximum violation (Eq. 24). The above stopping criterion implies that the controller will move the product properties at the end of the batch to the interior of the target region.

Calculation of Target Region Constraints and Their Gradients. The numerical algorithms described earlier require the values of the target region constraints and their gradients. This is obtained by integrating the nonlinear model for the batch process. Finite difference gradients are not recommended for use in Newton type methods, whose convergence depends on gradient information. The gradients here are obtained by sensitivity analysis, utilizing the equation for the time evolution of the sensitivity of the states with respect to inputs

$$\frac{d}{dt}\frac{\partial \boldsymbol{x}}{\partial \boldsymbol{u}} = \frac{\partial \boldsymbol{f}}{\partial \boldsymbol{x}}\frac{\partial \boldsymbol{x}}{\partial \boldsymbol{u}} + \frac{\partial \boldsymbol{f}}{\partial \boldsymbol{u}}$$
$$\frac{\partial \boldsymbol{x}}{\partial \boldsymbol{u}}_{(t=0)} = \boldsymbol{0} \tag{27}$$

The sensitivity equations are solved simultaneously along with the nonlinear equations using the program ODESSA

(Leis and Kramer, 1988). The nonlinear relationship between the end-use properties and the states is used to then find the gradients of the constraints of the target region $\nabla \boldsymbol{g}$

$$\frac{\partial \boldsymbol{g}}{\partial \boldsymbol{u}} = \frac{\partial \boldsymbol{r}}{\partial \boldsymbol{x}_f} \frac{\partial \boldsymbol{x}_f}{\partial \boldsymbol{u}} \tag{28}$$

Case Study: Emulsion Polymerization of Styrene

There is a strong interest in the control of emulsion polymerization reactors, as it provides the capability of producing polymers with special properties and performance. Emulsion processes are widely used for the manufacture of latex paints, adhesives, coatings and textile products. The importance of controlling end-use properties in these processes have been recognized (Dimitratos et al., 1994). The lack of adequate process and end-use property models and on-line measurements has been cited as the main problems for model-based control of emulsion polymerization processes.

The emulsion polymerization process represents an ideal choice for demonstrating the methodologies in this article. The dynamics of the process is very challenging due to the gel-effect and the inherent nonlinear nature. Secondly, many of the products from emulsion processes are value added, and the control of end-use properties is very important.

Mathematical model for emulsion polymerization of styrene

Different approaches can be used for developing a mathematical model for emulsion polymerization. One approach is to take the fundamental path and postulate the detailed equations for the polymerization process. A very comprehensive work on modeling emulsion polymerization using population balance approach was reported by Min (1976). Such an approach has the advantage that it considers all the necessary phenomena. However, using these models involves solving integro-partial differential equations for the evolution of particle sizes and molecular weights during the batch. On the other hand, semi-empirical equations have been proposed for modeling different phenomena in emulsion polymerization. An intermediate approach between a fundamental and a semi-empirical model is taken here in modeling the styrene polymerization system. This is based on a model previously developed by Liotta (1996) for bi-disperse particle size populations.

The particle-size distribution (PSD) of a latex product is an important characteristic due to its influence on the end-use properties. When the polymer latex is employed as paint, many of the end-use properties are directly related to the PSD of the polymer. This includes properties like scrub resistance, covering capacity and film forming ability. In other uses, the end-use properties are indirectly related to the PSD. In these cases, the molecular-weight distribution influences the end-use properties to a great extent and their final values are determined by the development of molecular architecture during the batch. The molecular-weight and particle-size distribution are dependent on each other due to the compartmentalized nature of the system.

A practical approach to modeling the emulsion polymerization system is pursued here. The PSD is discretized into a reduced number of populations, chosen based on statistical and physical arguments. The growth of these populations are modeled individually. The molecular weight part is handled by using moment transformations and retaining the first three moments of MWD. The moments for each particle-size population are modeled and are used to calculate the moments of the MWD for the overall polymer product. This is sufficient to predict the number and weight average molecular weights, and, hence, the end-use properties that depend on these average molecular weights.

Particle size evolution model. The particle-size distribution model is obtained from the study reported in Liotta (1996), where a bi-disperse model was presented. This was extended to the more general case with an appropriately chosen number of populations. The model involves macroscopic balances for the number of particles in each population and heat balance for the reactor. The rate of polymerization is modeled with the gel effect correlations reported in Liotta (1996).

The average number of radicals per particle are some of the most important and most difficult to predict states in the system. These states are decided by a combination of complex phenomena including radical entry into the particles, radical desorption, and radical termination in the particles. Traditionally, Smith Ewart recursion equations have been used to predict them. A different approach proposed in Li and Brooks (1993) is used here. The balance for the average number of radicals per particle $\bar{n}_i$ for each population can be given as

$$\frac{d\bar{n}_i}{dt} = \sigma_i - k_i \bar{n}_i - \frac{f_i k_t}{V_{si} N_a} \bar{n}_i \tag{29}$$

where σ_i is the radical absorption rate, k_i is the desorption rate constant for the population number, i and k_t is the radical termination coefficient, and V_{si} is the swollen volume. The aqueous phase radical balance, radical absorption, desorption and termination in the particles are obtained using correlations reported in the literature and explained in more detail in Valappil (2001).

Model for Molecular Weight Averages: Rate Processes Influencing the MWD. For accurate prediction of the MWD, all the important kinetic events that occur during the polymerization must be taken into account. Different rate processes such as radical entry to the particles, propagation, radical exit, termination and chain transfer influence the molecular weight of the final product. Of these, chain transfer and termination are the most important in deciding the MWD. In emulsion polymerization systems, the MWD and PSD depends on each other and cannot be treated independently. Exceptions occur when the chain transfer reactions dominate the other reactions, in which case, the entry or exit of radicals have little effect on MWD. Although a chain transfer agent is used here, we do not necessarily assume operation in a chain transfer dominant regime. An approach that is compatible with the method of discretization of the PSD is used here. The radicals balance for each population is used and the evolution of the average molecular weight for each population is modeled separately. The method of moments is utilized to find the first three moments of MWD for each selected population.

Finally, the combined effect of all three populations is utilized to find the average molecular weight.

A balance of live polymer chains for each population can be written by considering the rate processes influencing the MWD

$$\frac{dR_i^n}{dt} = \left(\frac{\sigma_i - k_i\bar{n}_i}{N_A}\right)\delta(n-1) + k_p[M]_p\left(R_i^{n-1} - R_i^n\right) + \left\{k_{fm}[M]_p + k_{cta}[X]\right\}\left(\sum_{j=1}^{\infty} R_i^j\delta(n-1) - R_i^n\right) - \frac{fk_t}{V_{si}}R_i^n\left(\frac{\bar{n}_i}{N_A}\right) \quad (30)$$

where R_i^n denotes the amount (in grmoles) of live polymer chains with n radicals in the ith population. The first term accounts for the entry and exit of the radicals, which have a single monomer unit. Hence, the term $\delta(n-1)$ in the above equation. The second term accounts for the propagation in the particle, where the radical with chain length $n-1$ grows to chain length n. The third term accounts for the chain transfer events to both monomer and the chain transfer agent. Here, k_{cta} is the rate constant for the chain transfer to the chain transfer agent, and $[X]$ is the concentration of the chain transfer agent. The final term accounts for the radical termination where the radical with chain length n terminates with all possible radicals.

The zeroth moment of the live polymer chain is related to the average number of radicals per particle and is given by $\bar{n}_i/N_A$. The moment transformation applied to the above equation yields the first moments for the live polymer chain distribution

$$\frac{d\lambda_i^1}{dt} = \left(\frac{\sigma_i - k_i\bar{n}_i}{N_A}\right) + k_p[M]_p\lambda_i^0 + \left\{k_{fm}[M]_p + k_{cta}[X]\right\}\left(\lambda_i^0 - \lambda_i^1\right) - \frac{fk_t}{V_{si}}\lambda_i^0\lambda_i^1 \quad (31)$$

The second moments for the live polymer chain distribution of population i are given by

$$\frac{d\lambda_i^2}{dt} = \left(\frac{\sigma_i - k_i\bar{n}_i}{N_A}\right) + k_p[M]_p\left(\lambda_i^0 + 2\lambda_i^1\right) + \left\{k_{fm}[M]_p + k_{cta}[X]\right\}\left(\lambda_i^0 - \lambda_i^2\right) - \frac{fk_t}{V_{si}}\lambda_i^0\lambda_i^2 \quad (32)$$

A balance for the dead polymer chain can similarly be performed as above. Here, the main events that lead to the formation of dead polymer are the chain transfer to the polymer and the radical termination in the particles. The amount of dead polymer chains P_i^n is given by

$$\frac{dP_i^n}{dt} = \left\{k_{fm}[M]_p + k_{cta}[X]\right\}R_i^n + \frac{fk_t}{V_{si}}\sum_{j=0}^{n} R_i^j R_i^{n-j} \quad (33)$$

The equations for the first, second, and third moments for the dead polymer chain distribution can be obtained as

$$\frac{d\mu_i^0}{dt} = \left\{k_{fm}[M]_p + k_{cta}[X]\right\}\lambda_i^0 + \frac{fk_t}{V_{si}}\left(\lambda_i^0\right)^2 \quad (34)$$

$$\frac{d\mu_i^1}{dt} = \left\{k_{fm}[M]_p + k_{cta}[X]\right\}\lambda_i^1 + \frac{fk_t}{V_{si}}\lambda_i^0\lambda_i^1 \quad (35)$$

$$\frac{d\mu_i^2}{dt} = \left\{k_{fm}[M]_p + k_{cta}[X]\right\}\lambda_i^2 + \frac{fk_t}{V_{si}}\left\{\lambda_i^0\lambda_i^2 + \left(\lambda_i^0\right)^2\right\} \quad (36)$$

The molecular weight averages can be obtained from the moments of distribution. The number average ($\overline{MW}_n$) and weight average ($\overline{MW}_w$) molecular weights are obtained as

$$\overline{MW}_n = \frac{\sum_{i=1}^{n_p}\lambda_i^1 + \sum_{i=1}^{n_p}\mu_i^1}{\sum_{i=1}^{n_p}\lambda_i^0 + \sum_{i=1}^{n_p}\mu_i^0} \qquad \overline{MW}_w = \frac{\sum_{i=1}^{n_p}\lambda_i^2 + \sum_{i=1}^{n_p}\mu_i^2}{\sum_{i=1}^{n_p}\lambda_i^1 + \sum_{i=1}^{n_p}\mu_i^1} \quad (37)$$

The polydispersity of the polymer is obtained as the ratio of weight average to number average molecular weight

$$PD = \frac{\overline{MW}_w}{\overline{MW}_n} \quad (38)$$

The number of states in the model developed with the discretization of PSD noted earlier depends on the number of populations. For the case with three populations, 26 states are to be considered. Some of the parameters that are used for the above model were validated at 50°C. Thus, the control studies are done by maintaining the temperature in the vicinity of 50°C. However, the temperature dependence of the important parameters (including initiation, propagation and termination) are considered in this model.

On-line measurements. The availability of reliable on-line measurements is very important for the successful application of the nonlinear MPC. The density measurement is usually available quite readily in emulsion polymerization. A PSD measurement is less commonly employed on-line, but is considered in this study. The measurements available in this process are listed below.

- *Density of reaction mixture*: Density can be related to the conversion of the instantaneous reaction mixture as

$$\rho_l = \frac{Mass_0 + M_{\text{add}}}{Vol_0 + M_{\text{add}}/\rho_m - X(M_0 + M_{\text{add}})(1/\rho_m - 1/\rho_p)} \quad (39)$$

- *Reactor and jacket temperature*
- *Infrequent measurement of PSD*: This is an important measurement in the system. Capillary hydrodynamic fractionation (CHDF) measurement of an unswollen particle size is considered in this study. The sampling is not very frequent as the measurement values are available with a delay due to the complexity of the method. Both the frequency of sampling and the measurement delay are assumed to be 10 min.

End-Use properties..

• *Melt Flow Index:* This property is given by a correlation with the weight average molecular weight (Bremner and Rudin, 1990) as below

$$MI = \frac{30}{\left(\overline{MW}_w^{3.4} \times 10^{-18} - 0.2\right)} \tag{40}$$

• *Tensile Strength:* The dependence of the tensile strength on the number average molecular weight reported in Bernsted and Anderson (1990) is used

$$\sigma = 7390 - 4.51 \times 10^8 \left(\frac{1}{\overline{MW}_n}\right) \tag{41}$$

• *Weight Average Particle Diameter:* Some of the end-use properties of commercial polymers are dependent on the particle size distribution of the polymer. The weight average particle diameter ($\overline{D}_W$) of the production is considered as a property, and controlled. This value is calculated from the particle sizes of the individual populations as

$$\overline{D}_w = \frac{\Sigma_{i=1} D_i W_i}{\Sigma_{i=1} W_i} \tag{42}$$

where D_i is the diameter of the ith population and W_i denotes the weight fraction of each population in the total polymer.

State Estimation and NLMPC of Styrene Polymerization

Parameter adaptive extended Kalman filter

The emulsion polymerization process under study here is characterized by infrequent PSD measurements that are noisy. Also, unmeasured disturbances and the process model mismatch enter during the batch. As the solid content in the reactor increases during the batch, the heat-transfer efficiency is reduced, which acts as an unmeasured disturbance. A PAEKF is very useful in handling these cases. The selection of the estimated parameters is very critical in the performance of the EKF. Usually, the parameters or states with the greatest uncertainties are selected for adaptation. In the case of emulsion polymerization, the average number of radicals per particle is the important variable that affects the evolution of both particle size and molecular weight (Liotta, 1996). Thus, the augmented parameters are biases added to the three equations for the average number of radical per particle

$$\frac{d\bar{n}_i}{dt} = \sigma_i - k_i \bar{n}_i - \frac{f_i k_t}{V_{si} N_a} \bar{n}_i + \theta_i, \quad i = 1, 2, 3 \tag{43}$$

The adapted parameters need to be selected such that the augmented state space system is observable with the available measurements. For this, the number of adapted parameters should not be greater than the number of measurements. These parameters are estimated only when the PSD measurements become available every 10 min. A two-time scale filter is used in such a situation and a number of such studies are reported in the literature. When only the frequent measurements are available, the filter performs a closed-loop estimation with only the observable states, while the states related to molecular-weight distribution are estimated in open-loop. When the delayed measurements of these states are available, a full order filter is used to re-update all the states at the earlier measurement time. This is then used along with the most recent frequent measurements to update the state values to the present. The additional details on the two-time scale filter are available in Liotta (1996) and Liotta et al. (1997a). The process noise covariance for the filter is specified using the approach developed in Valappil and Georgakis (2000). Valappil and Georgakis (2000). The covariance for the adapted parameters are obtained by trial with different values.

Implementation of nonlinear model predictive control

The nonlinear model predictive formulation discussed in the subsection titled "Nonlinear model predictive control formulations," is used for control of the different end-use properties of polystyrene. Three manipulated variables are used in this study: the monomer addition rate, the CTA addition rate, and the coolant flow rate. The coolant flow rate is

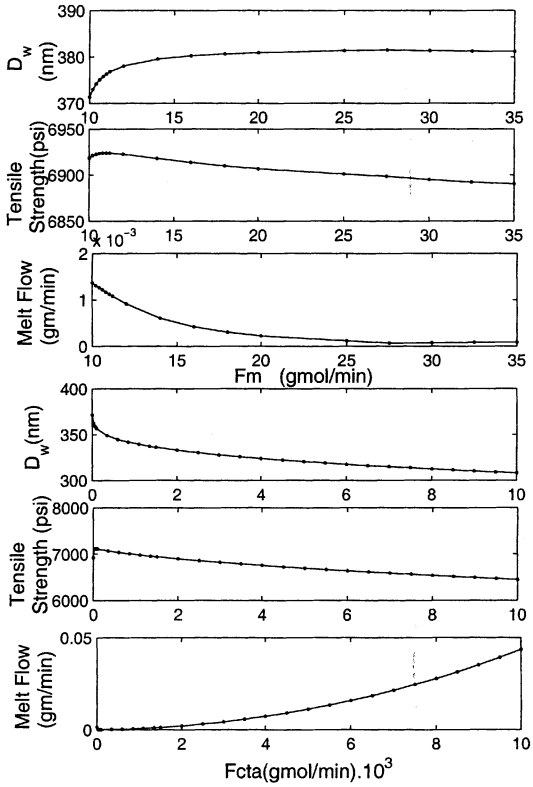

Figure 3. Effect of manipulated variables (monomer and CTA addition) on the three end-use properties of polystyrene.

predominantly used to control the temperature at around 50°C. This is achieved by considering the path constraints on the temperature, and using these constraints in the NLMPC algorithm. Temperature values five steps into the future are controlled this way. The total amount of monomer to be added during the batch is constrained and is pre-calculated based on the recipe and the desired final PSD. Thus, the total amount of monomer to be added during the batch is constrained. This constraint is considered along with the target region constraint and the manipulated variable constraints in the LP and QP steps of the control algorithms.

In the application of Lagrange polynomial method for the input parameterization of the future values of the input variables, two different types of manipulated variables are considered.

• *Inputs constrained by a pre-specified total amount (for example, total monomer addition allowed).* In this case, the inputs at equi-spaced points in time and the total time of addition are chosen as the parameters. This introduces a new constraint in the problem in the form of the total amount of a specific reactant to be added during the batch.

• *Inputs not constrained by the amount to be added.* In this case, the parameters are the inputs at equi-spaced points between the current time and the end of the batch. For the addition of the chain transfer agent (CTA), four points are considered in its parameterization.

The effectiveness of the control strategy is tested under different disturbances and model uncertainties. These include the following:

• Mismatch in the concentration of the initiator in the recipe.

• Mismatch in the parameter values of k_{adj} and k_t.

• Heat transfer fouling during the batch that reduces the heat-transfer coefficient.

The above disturbances entering during the batch are all unmeasured, and, hence, the NLMPC controller would rely on the information from the state estimator for finding the control actions. The interest here is to study the effectiveness of the target region control approach in handling these disturbances. Also, the advantages of using the target region compared to a fixed set point needs to be examined. Apart from these, the comparison of the min-max and the successive linearization method for the solution of the problem is of interest.

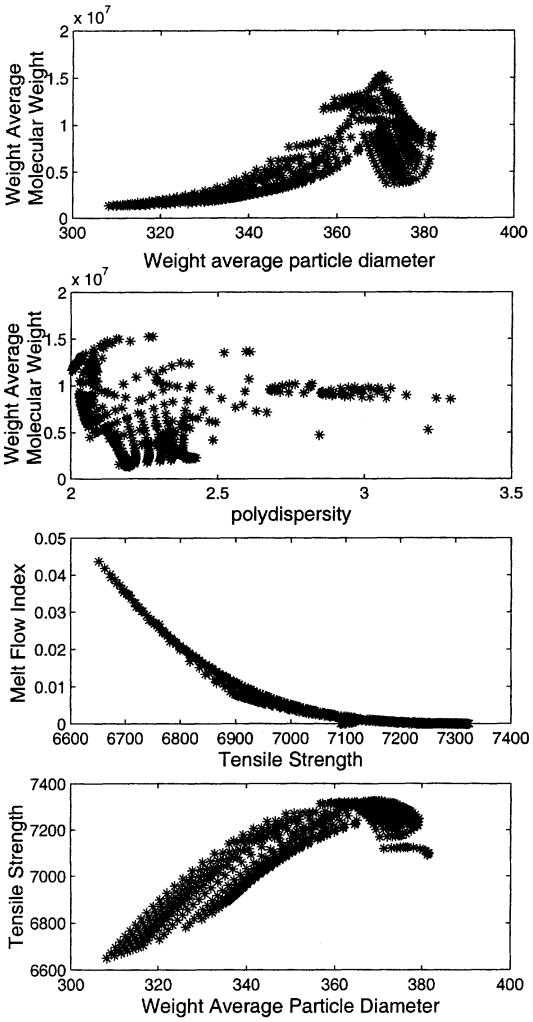

Figure 4. Reachable regions at the end of the batch attained by varying the constant monomer addition rate and the CTA addition rate within their acceptable range.

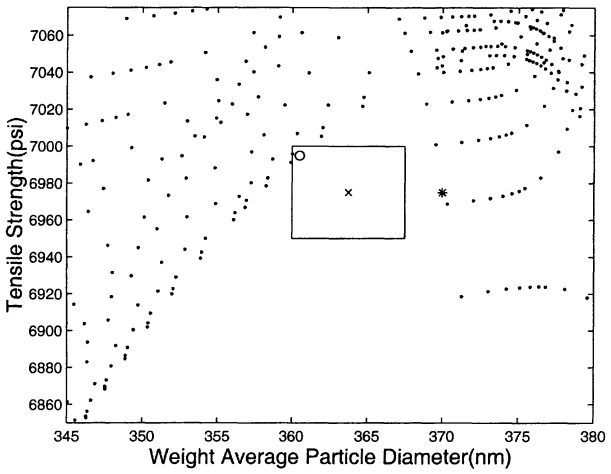

Figure 5. Comparison of traditional and target region approaches in controlling two end-use properties.

The asterisk shows the end of the batch properties attained by considering a control strategy that aims at a set point x at the center of the region. Point o shows the end-use properties attained with the control strategy based on the target region depicted by the parallelogram. Dotted points represent the reachable region calculated with the assumption of a constant flow rate.

Results

The effect of the two available manipulated variables, monomer and CTA, on the three end-use properties of the final polymer product are shown in the Figure 3. This plot can help identify the manipulated variable that can change a property in a desired direction. For the control of melt index

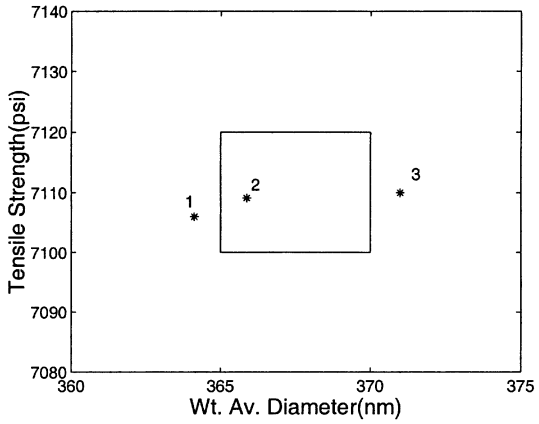

Figure 6. End of the batch properties for control of tensile strength and weight average particle diameter.

Point 1 shows the point with open-loop profile in the presence of disturbance, 2 shows the point with target region approach, and 3 shows the point with conventional minimization approach.

and tensile strength, the CTA addition needs to be manipulated, as this has a significant effect on these properties. However, it affects the conversion adversely, and thus the addition of CTA is constrained with an upper limit, which is .001 grmol/min. The monomer addition rate increases both the average molecular weight and the average particle size at the end of the batch, although the effect on molecular weight is not as pronounced as it is on the particle size.

The reachable regions using a constant addition rate of the monomer and chain transfer agent are shown in Figure 4. The addition rates for a monomer vary from 10 grmole/min to 35 grmole/min, and for the CTA varies from 0 grmol/min to 1.0 grmol/min. Two important things are to be noted about these reachable regions. First, the end-use properties appear to be very correlated, as they are all affected by the same kinetic mechanism. Also, the reachable regions tend to be substantially nonconvex. The above regions are seldom available *a priori* due to the computational effort involved in obtaining them and the fact that they are modified by the presence of nonstationary disturbances. It needs to be noted that the reachable regions depicted in Figure 4 were determined assuming the constant flow rate with the addition of a monomer and CTA. They could be denoted as constant flow reachable regions (CFRR). They only represent a subset of the reachable regions that can be obtained if all possible flow rate profiles in time are allowed.

The advantages of using a controller that aims at a target region over a controller that aims at a single set point is demonstrated in Figure 5. Here, the CFRR, denoted by the dotted points, is quite nonconvex and intersects the target region at one corner. The set point chosen for this comparative study is at the center of the target region, which is inside the CFRR. It is not known *a priori* whether it is reachable with a more complex flow profile. A weighted minimization objective is used for finding the control moves using sequential quadratic programming. This approach results in the controller finding a local minimum at some boundary point of the reachable region. Thus, the controller cannot ensure that it achieves a point within the target set. This can happen whenever the desired set point is not within the reachable region. This demonstrates that considering the desired end-use property region, the NLMPC can reduce the possibility to failing to meet product specifications.

The control of two end-use properties of interest, weight-average-particle-diameter and tensile strength using the monomer flow and the coolant flow rate, is shown in Figure 6. The CTA flow is fixed at 5×10^{-5} mol/min and an initial W_p value of 0.66 is used. The mismatch in the parameter k_{adj} is considered, whereby the model value of this parameter is 25% higher than in the actual process. As a result, the actual end of the batch properties attained without any compensating control action (Point No. 1 in Figure 6) is outside the specifications for the weight average diameter. Point No. 2 in the same figure is attained with the target region controller. The monomer addition rate is increased to appropriately account for the mismatch and the product specifications are

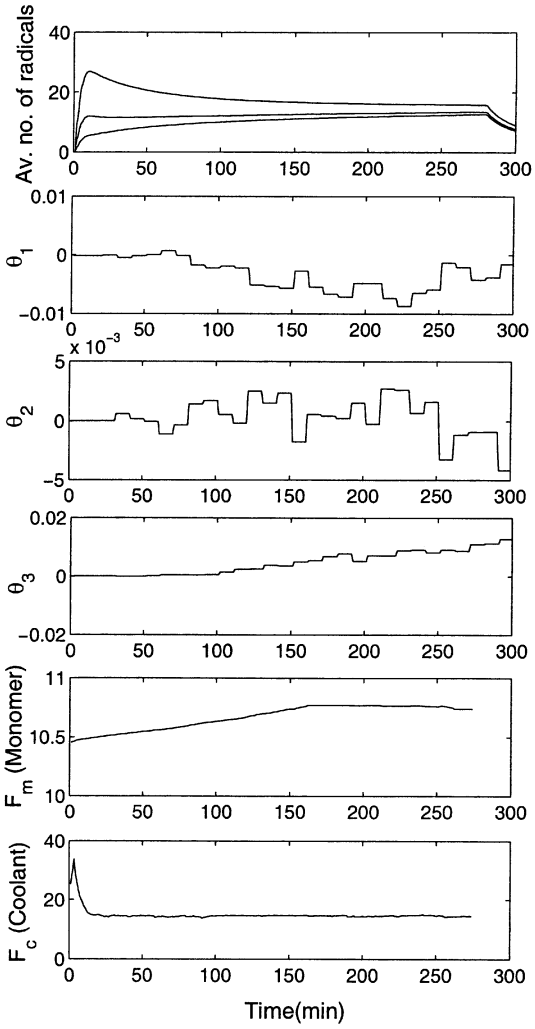

Figure 7. Time profiles corresponding to Point 2 of Figure 6.

The three adapted parameters are shown in the plots on the left. The monomer flow and coolant flow are given in plots on the right along with the average number of radicals per particle.

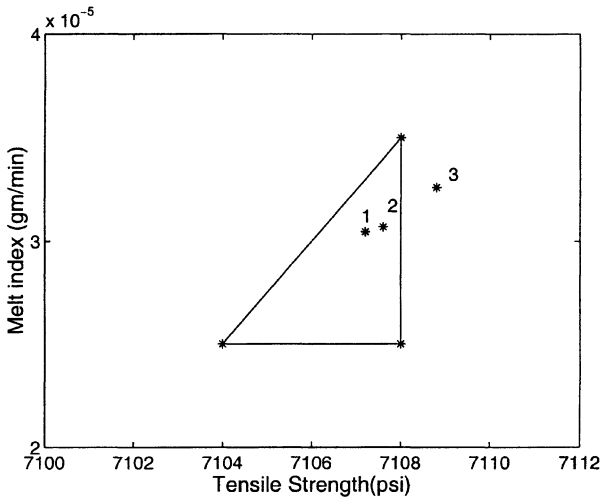

Figure 8. End of the batch properties attained using CTA flow rate as the manipulated variable.

Point 1 shows the properties that the controller predicted would be achieved, Point 2 shows the point that was achieved with the target region controller and Point 3, the properties with open loop policy.

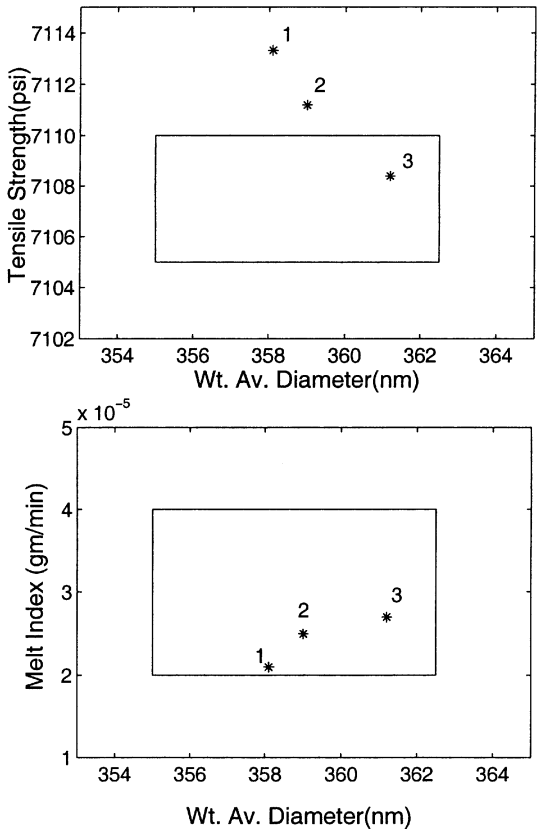

Figure 9. Control of the properties in a region using CTA as the manipulated variable.

Point 1, open-loop policy, Point 2, using set point approach, and Point 3, using proposed target region controller. Mismatch is in the initial initiator concentration.

achieved (Figure 7). The traditional MPC controller using the center of the target region as the desired set point could not succeed in making the end-use properties be inside the target region (point No. 3 in Figure 6). The monomer flow rate in this case is depleted towards the end of the batch, and, thus, the average number of radicals per particle drop at that time. The coolant flow rate associated with this batch run is also given in the Figure 7. The controller successfully maintained the batch temperature within the constraints demanded, which is 50 ± 0.5°C.

Another case study for control of the melt index and tensile strength is shown in Figure 8. Here, the CTA addition flow rate is primarily used to control the properties, and a fixed monomer addition of 10 grmol/min is used throughout the batch. A triangular target region is considered as the control target. The disturbance in this case is a mismatch in the initial concentration of the initiator. This mismatch is such that the model assumes that the initiator charged in the recipe is 30% more than what is actually present in the reactor. The effect of this mismatch is an increase in the tensile strength of the actual product, resulting in the end-of-batch properties shown in point 3 of Figure 8, outside the target. With the target region controller, the point 2 is achieved, which is well within the specifications. Note that the controller actually intended to attain the two properties at point 1, but the disturbances moved it to the point 2 instead. This point is closer to the boundary of the region, but is still within the product specifications.

The attempt to control three properties (tensile strength, melt index, and weight-average-particle-diameter) at the same time using the CTA addition as the sole manipulated variable is demonstrated in Figure 9. Here, the reachable region is a 1-D line that passes through the target region. A mismatch in the initial concentration of the initiator is introduced. The initiator concentration used in the model is 30% less than that in the process, and this causes the tensile strength specification to be violated (Point 1 in Figure 9). The adapted parameters in EKF are shown in Figure 10. They assume positive values to appropriately account for the decrease in the values of an average number of radicals per particle. The controller decreases the CTA addition rate until the zero flow constraint is reached at around 260 min. This way, the end of the batch properties are controlled within the target region (Point 3 of Figure 9). The end-use properties that are attained using the conventional approach set point target are shown in point 2 of Figure 9. Again, the center of the target region is chosen as the set point for the minimization objective. The tensile strength specification is not met in this case, due to the nonconvex nature of the reachable region.

The effect of an additional disturbance entering during the batch, on the control of three properties is shown in Figure 11. Here, the disturbance considered is reactor fouling, which occurs at $t = 100$ min. This is in addition to a mismatch in the initiator charge previously considered. This reduces the heat-transfer coefficient and results in an open-loop value of properties that are characterized by point 1 in Figure 12. The values of end-use properties that are attained using the target region controller is point 2, which is outside the target region. The CTA addition reaches the lower bound of zero, and remains there. Hence, the controller is not able to reach

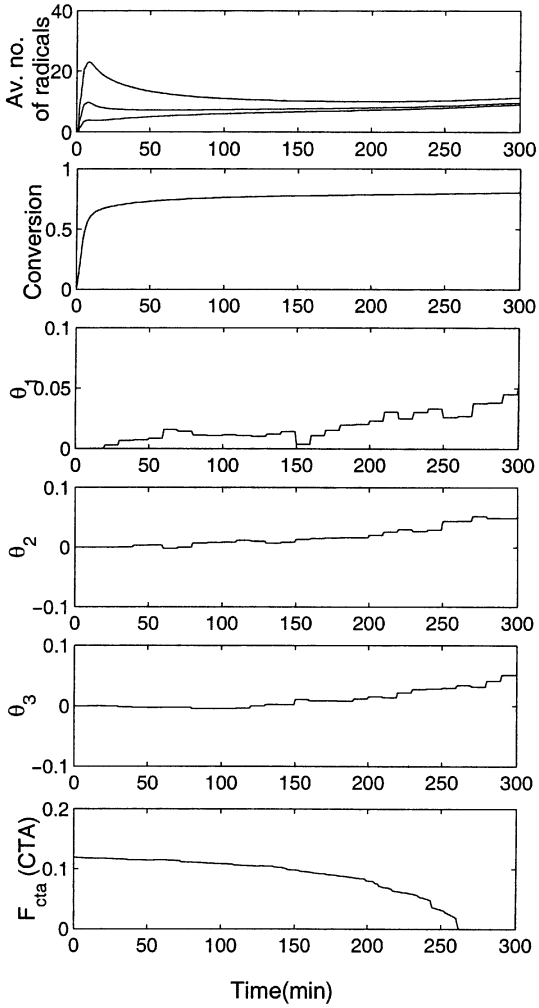

Figure 10. Three adapted parameters from EKF on the left.

CTA addition used for control is shown on the right, along with conversion and average number of radicals per particle plots.

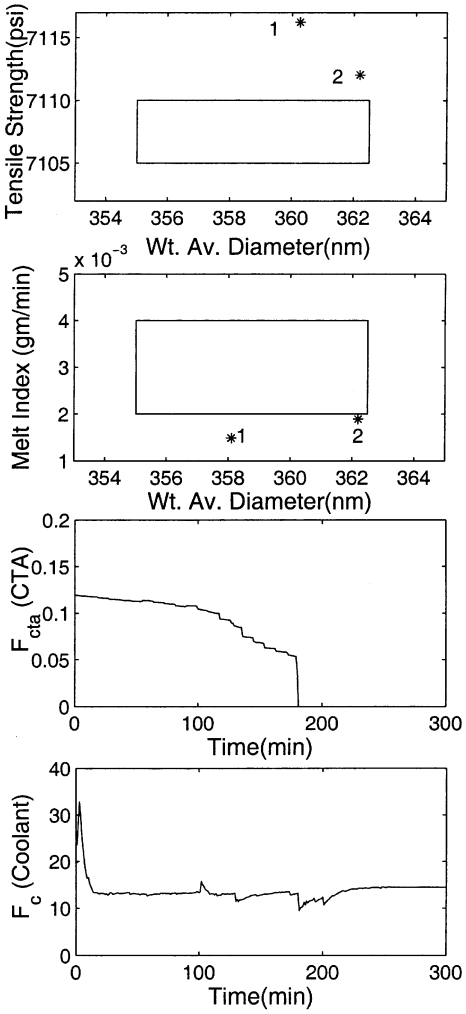

Figure 11. Control of 3 end-use properties with an additional disturbance, heat-transfer fouling.

The open-loop properties are shown in point 1 and the properties attained using target region control, at point 2 in the top plot. The CTA addition is given in bottom left plot, the controller does not have enough freedom to bring the properties to the target region.

the target region specifications in this case. The solution to this would be to either enlarge the region or to use a different additional manipulated variable, thus expanding the reachable region so as to interest the target region.

The above case study of controlling the above three properties was repeated with both the monomer addition and the CTA as the manipulated variables. This expands the reachable regions significantly, and the controller is able to bring the properties to the desired region. The end-use properties attained at the end of the batch in this case is shown in point 2 in Figure 12. The three adapted parameters are also shown in Figure 13 These parameters take positive values to increase the average number of radicals. The CTA flow rate is used primarily at the initial stages of the batch for control, but hits the lower constraint in the middle of the batch. The controller then manipulates the monomer addition to bring the properties to the desired target region (Figure 13).

The comparison between the two optimization formulations examined here, the min-max method and the target control with successive linearizations, is shown in Figure 14 and Table 3. Here, two aspects are studied, the computational requirements and the convergence properties. The average time requirement in seconds at each control execution for the methods is shown in Table 3. The min-max method takes significantly longer to converge, as can be seen from these results. Another important feature, the convergence of the algorithm, is sketched in Figure 14. This figure shows the approximate boundaries of the region of convergence, the initial guess from where the algorithm can start and still find the solution to the end-use property control problem. The controller was started with initial guesses for two input variables at the grid points. These boundaries are only approximate, as a rigid demarcation of this property is not possible without a substantial number of trials. As can be seen from Figure 14, the target region linearization method has a larger region of convergence than the min-max method.

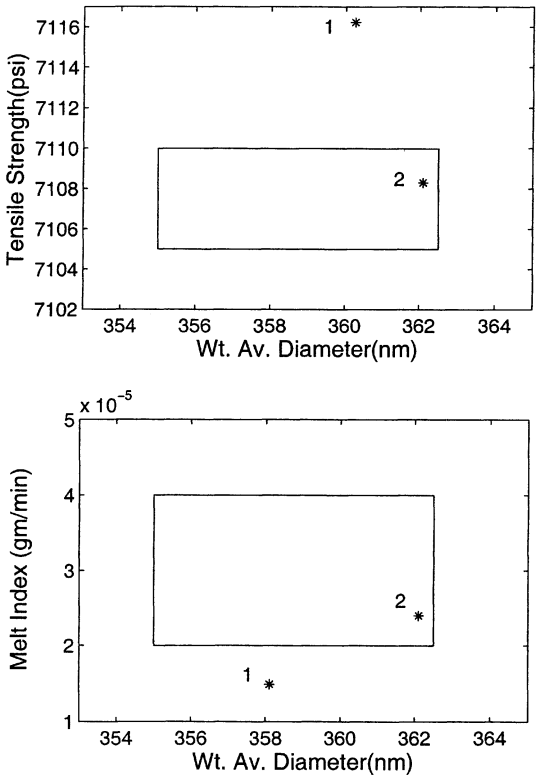

Figure 12. Control of 3 end-use properties with manipulation of both monomer and CTA addition.

The desired target region is attained in this case with closed-loop control, as shown in point 2. The effect of disturbances in this case is to move the end-use properties to point 1 (open-loop case).

Conclusions

In this article we have formulated a nonlinear model predictive controller for the control of end-use properties so that they fall within a target region. The change of attention from molecular to end-use properties used here fits well with the recent emphasis on customer driven manufacturing. The issue of inadequate degrees of freedom to control all the molecular or end-use properties is handled with this approach. This framework requires the use of a static or dynamic model that relates the molecular to the end-use properties.

The formulation of the control problem in terms of a target region, instead of a set point target, was found to be beneficial from several viewpoints. First, the ability of the controller is enhanced, as it handles processes with nonconvex reachable regions. This was demonstrated for the case when the set point is not inside the with nonconvex reachable region. In such a case, the presence of local minima result in the failure of a conventional controller to achieve a product with properties within the desired ranges. A better selection of the set point target is not feasible as the reachable regions are not known *a priori* and depend on the active disturbances. Furthermore, the reachable regions are nonconvex and depend nonlinearly on the operating conditions of the batch process. Even in the absence of disturbances that modify the reachable regions on-line, their calculation is a very intensive computational task.

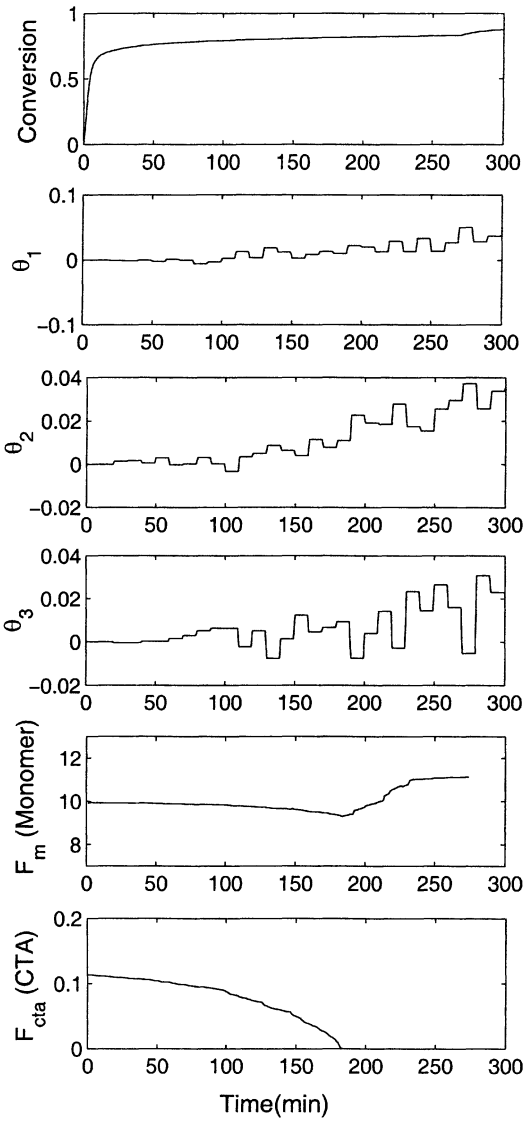

Figure 13. Two manipulated variables on the right.

The CTA Flow hits the lower constraint in the middle of batch. The monomer addition is manipulated later on in the batch to control the properties. The three adapted parameters are shown in the left plots.

The formulation of NLMPC with a target region necessitates the introduction of suitable optimization techniques for this problem. The method proposed is based on the work of Mayne and Sahba (1985) and utilizes successive linearizations to solve the inequality optimization problem. Its advantages over the min-max method were also demonstrated. It is seen that, both from the computational point of view and from convergence considerations, this algorithm performs better for the end-use property control.

This NLMPC for end-use property control within target regions was tested in the presence of unmeasured distur-

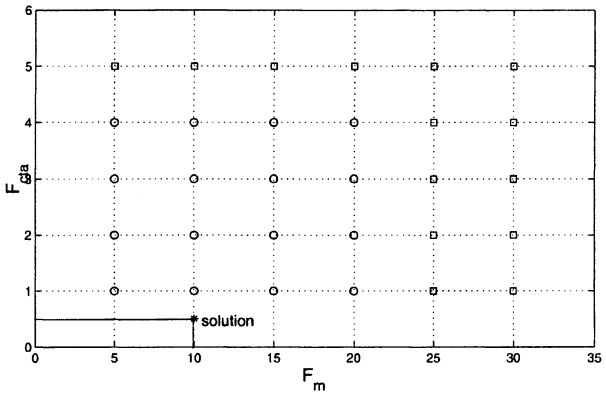

Figure 14. Comparison of convergence properties of the min-max and successive linearization approaches.

Initial guesses marked the circles indicate the convergence of both methods, while those marked with a square indicate convergence of successive linearization approach.

Table 3. Average Computational Time Required to Calculate Control Moves for the Min-Max and Target Region Methods for Different Control Case Studies

Controlled End-Use Properties	Min-Max (s)	Succ. Lineraiz. (s)
Tensile Strength, Avd. Dia.	1.93	0.41
Tensile Strength, Melt Index	2.21	0.56
Tensile Strength, Melt Index, Avg. Dia.	2.87	0.71

bances and model uncertainties. The PAEKF scheme used is found to be very efficient in handling both parametric uncertainties and unmeasured disturbances. The methodology proposed by Valappil and Georgakis (2000) for specifying the state noise covariance matrix for the EKF was also very useful, as the state vector dimension in the example is very large. Trial and error tuning of these values is bound to involve significant efforts.

The end-use properties that are considered are of the same magnitude in the example problem studied here. If the magnitudes of the properties are very different, it would be necessary to scale them appropriately to improve the numerical properties of the algorithm. This can be achieved in different ways, including using the ratio of the end-use property to an average value or a bound.

The objective of the control method presented here was to find the inputs that would bring the end-use properties to the interior of the target region. The concepts and methodologies developed here are very relevant for the application studied, since product quality is of primary importance in emulsion products. The challenging features that characterized the styrene polymerization process include highly nonlinear dynamics, insufficient measurements, unmeasured disturbances, and process-model mismatch. A realistic scenario where the end-use properties depend on both particle size and molecular weight components was studied here. Even though the model used here for the on-line control has 26 states, the execution time for state estimation and NLMPC was found to be very reasonable.

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