Receding horizon control for polymerization processes

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Abstract

Receding horizon control is used to maximize the productivity of emulsion polymerization processes. The control strategy is adapted for controlling multivariable systems. Two process inputs, the monomer flow rate and the reactor temperature, were manipulated in order to optimize process productivity. The controller performance was validated in the presence of modeling errors.

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1. Introduction

Online control of chemical processes is an important issue for ensuring process security and good product quality. Polymerization processes can be exothermic, rapid and sensitive to impurities. For this reason, modeling, monitoring, and control of these processes is of high importance for optimizing the process productivity. Actually, modeling gives a good comprehension of the process and allows us to relate the process inputs (or manipulating variables) to the process outputs (or the process measurements). Online monitoring is important for obtaining real information on the evolution of the process and for detecting any deviation in its course. Online control allows then the correction of this deviation by manipulating the process variables.

A classical problem in emulsion polymerization processes is optimizing the productivity by increasing the reaction rate under specific constraints. Maximizing the process productivity allows one to minimize the process time while ensuring its security. The process constraints are usually due to physical limitations such as the jacket capacity to evacuate the heat and the limited concentration of monomer in the polymer particles.

The process productivity is a function of the reaction operating conditions, temperature, and flow rates of monomer, initiator and surfactant. In the literature, the process productivity was controlled by manipulating the flow rate of monomer (see for example [2,7,4,11,14,13,12]). In this work, the maximum number of operating conditions of the process is used in the control scheme in order to optimize the process productivity. Therefore, the monomer flow rate and the reactor temperature are manipulated to control the reaction rate. Actually, the reaction temperature affects the rate of decomposition of the initiator and the rate of propagation of the monomer, and can therefore be used to maximize the reaction rate.

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Nomenclature

\[ \begin{align*}
A & \quad \text{Heat transfer area (m}^2\text{)} \\
C_P, C_P^{\text{feed}} & \quad \text{Heat capacity of the reaction medium and of the feed (J/kg/deg)} \\
EA & \quad \text{Activation energy (J/mol)} \\
F, F_{\text{feed}} & \quad \text{Monomer flow rate (mol/s) and total reactor feed rate (kg/s)} \\
k_p & \quad \text{Propagation rate (cm}^3\text{/mol/s)} \\
k_p^0 & \quad \text{Pre-exponential factor (cm}^3\text{/mol/s)} \\
M & \quad \text{Mass of the reaction medium (kg)} \\
M_P & \quad \text{Concentration of monomer in the polymer particles (mol/cm}^3\text{)} \\
\bar{n} & \quad \text{Average number of radicals per particle} \\
N, N_T & \quad \text{Residual and total number of moles of monomer (mol)} \\
N_P & \quad \text{Total number of particles} \\
N_A & \quad \text{Avogadro’s number (mol}^{-1}\text{)} \\
Q, Q^{\text{loss}}, Q_{\text{feed}} & \quad \text{Heat of the reaction, heat loss and heat exchanged with the feed (W)} \\
Q_{\text{max}} & \quad \text{Maximum allowable heat of the reaction (W)} \\
R & \quad \text{Universal gas constant (J/mol/K)} \\
R_P & \quad \text{Reaction rate (mol/s)} \\
T, T_{\text{feed}}, T_j & \quad \text{Reactor, feed and jacket temperatures (°C)} \\
U & \quad \text{Heat transfer coefficient (W/°C/m}^2\text{)} \\
V & \quad \text{Reaction medium volume (cm}^3\text{)} \\
\Delta H & \quad \text{Reaction enthalpy (J/mol)} \\
\mu & \quad \text{Total number of radicals per particle} \\
\phi_p & \quad \text{Fraction of polymer in the polymer particles} \\
\rho_m, \rho_p & \quad \text{Monomer and polymer densities (g/cm}^3\text{)} \\
\end{align*} \]

In order to do this, a multivariable control technique should be employed. The receding horizon control is known to be efficient for the control of multivariable linear systems [6,9,10,1,3]. The receding horizon control also takes into account the constraints on the input and on the states comprising constraints on the amplitude and on the rate of variation. It anticipates changes in the process and in the inputs since it is based on the process model.

In the first part of the paper, the process model is presented and the process inputs and outputs are outlined. Then, the process controllability is verified and the controller is developed, taking into account the process constraints. Finally, the controller is validated during the emulsion polymerization of styrene.

2. Process model

The process model consists of the material balance of monomer and heat balance in a semi-continuous reactor as shown by the following system:

\[ \begin{align*}
\dot{N} & = F - R_P \\
MC_P T & = Q_R + UA(T_j - T) + \frac{F_{\text{feed}}C_P^{\text{feed}}(T_{\text{feed}} - T) - Q_{\text{loss}}}{Q_{\text{feed}}} \\
\end{align*} \] (1)

where \( N \) is the number of moles of free monomer, \( T \) the reaction temperature, \( F \) the monomer flow rate, \( M \) the reaction medium mass, \( C_P \) the heat capacity of the reaction medium, \( U \) the heat transfer coefficient, \( A \) the heat transfer area, \( T_j \) the jacket temperature, \( F_{\text{feed}} \) the input flow rate, \( T_{\text{feed}} \) the input temperature, \( C_P^{\text{feed}} \) the heat capacity of the feed, \( Q_{\text{loss}} \) the heat loss to the environment due to the non-surrounded regions of the reactor and to the condenser and \( R_P \) is the reaction rate. The heat produced by the reaction (\( Q_R \)) is related to the reaction rate by the following equation:

\[ Q_{\text{R}}^{\text{max}} = (-\Delta H) R_P \] (2)
The reaction rate is related to the concentrations of monomer and radicals in the polymer particles \([M^P] \) and \(\mu\) as given by the following equation:

\[
R_P = k_P(T) \overline{\tilde{n} N_P} [M^P]V
\]

where \(V\) is the reaction medium volume and \(k_P\) is the propagation rate coefficient that is related to the reaction temperature \((T)\) by the following relationship:

\[
k_P(T) = k_{p0} \exp \left( -\frac{E_A}{RT} \right)
\]

where \(E_A\) is the activation energy, \(R\) the universal gas constant and \(k_{p0}\) the pre-exponential factor.

The concentration of monomer in the particles is constant during interval II where the polymer particles are saturated in monomer and monomer droplets exist. During interval III, all the monomer is assumed to be in the polymer particles as given by the following equation:

\[
[M^P] = \begin{cases} 
\frac{(1 - \phi_P^P)\rho_m}{MW_m} & \text{if } \frac{MW_m N}{\rho_m} - 1 - \phi_P^P \left( \frac{MW_m}{\rho_m} \right) \left( N^T - N \right) \geq 0 \\
\frac{MW_m (N^T - N)}{\rho_m} + \frac{N}{\rho_m} & \text{else}
\end{cases}
\]

where \(\phi_P^P\) is the volume fraction of polymer in the particles under saturation, \(MW_m\) the monomer molecular weight and \(\rho_m\) and \(\rho_p\) the monomer and polymer densities respectively. These parameters are constant with respect to time. \(N^T\) is the total amount of monomer introduced into the reactor:

\[
N^T = \int_0^t F dt.
\]

Due to the exothermic nature of the reaction, the process is usually monitored by calorimetry. The reactor temperature and that of the inlet and the outlet of the jacket are measured online. The unknown terms in the heat balance, \(Q_{\text{loss}}\) and \(U\), can be estimated either by introducing discrete measurements of the monomer conversion or by interpolating \(U\) as a function of the solid contents \([4,5]\). The heat produced by the reaction can therefore be estimated from the heat balance, which allows us to estimate the reaction rate and the number of moles of monomer in the reactor. With these measurements, the parameter \(\mu\) can be estimated by applying an observer in order to avoid oscillations due to measurement noise \([12]\).

3. Process control

Eq. (3) shows that the reaction rate can be controlled by manipulating either the concentration of monomer in the polymer particles or the reaction temperature. Controlling the concentration of monomer, one needs to manipulate the monomer flow rate and controlling the reaction temperature can be done by manipulating the jacket temperature. Process control has to be adapted to nonlinear systems and has to be done by taking into account the constraints that are the saturation of polymer particles and the maximum allowable heat produced by the reaction.

3.1. Process controllability

The process model can be written in the following form:

\[
\begin{bmatrix}
\dot{N} \\
\dot{T}
\end{bmatrix} = \begin{bmatrix}
1 & 0 \\
0 & \frac{UA}{MC_P}
\end{bmatrix} \begin{bmatrix}
F \\
T_j
\end{bmatrix} + \begin{bmatrix}
0 \\
\frac{-R_P}{MC_P}
\end{bmatrix} + \begin{bmatrix}
\frac{Q_{\text{feed}} - Q_{\text{loss}}}{MC_P}
\end{bmatrix} f(x)
\]

\[
y = h(x) = x.
\]
The controllability of the system can be verified by calculating the characteristic matrix \([8]\) that has to be non-singular. The characteristic matrix of the system is calculated as follows:

\[
C(x) = \begin{bmatrix}
L_g L_f^{-1} h_1 & L_g L_f^{-1} h_1 \\
L_g L_f^{-1} h_2 & L_g L_f^{-1} h_2
\end{bmatrix} = \begin{bmatrix}
\frac{\partial h_1}{\partial x_1} g_1 & \frac{\partial h_1}{\partial x_2} g_2 \\
\frac{\partial h_2}{\partial x_1} g_1 & \frac{\partial h_2}{\partial x_2} g_2
\end{bmatrix} = \begin{bmatrix}
1 & 0 \\
0 & UA
\end{bmatrix}
\]

(8)

where \(L_f h\) is the Lie derivative, that is the directional derivative of the function \(h\) in the direction of the vector \(f\). The characteristic matrix is non-singular. The system is therefore controllable.

### 3.2. Input/output linearization

In order to apply the linear receding horizon control, the process needs to be linearized. The system does not need to be linearized around an operating point. An input/output linearization \([8]\) is sufficient for applying a linear controller. The input–output linearization gives the following linear system:

\[
\begin{bmatrix}
\dot{N} \\
\dot{T}
\end{bmatrix} = \begin{bmatrix}
w_1 \\
w_2
\end{bmatrix}.
\]

(9)

The real process input is calculated from this linearization as follows:

\[
u = \begin{bmatrix}
F \\
T_f
\end{bmatrix} = \begin{bmatrix}
MC_p \\
UA
\end{bmatrix} w_2 - \frac{1}{UA} (Q_R + Q_{feed} - Q_{loss}) + T
\]

(10)

where \(w_1\) and \(w_2\) are the new inputs of the linear system.

### 3.3. Receding horizon controller

Linearizing the process allows decoupling the coupled variables \(u_1\) and \(u_2\) and now any linear controller can be used to control the process. A robust controller that takes into account model uncertainties should however be applied since the linear part of the process model might be inaccurate. A sliding horizon controller that is based on the minimization of an objective function is used in this work to accomplish this objective.

The controller applied to the linearized system is summarized in the block diagram (Fig. 1), where \(A\) and \(R\) are two reference models related to the set point and to the regulation respectively. These models ensure the application of real process inputs and errors by filtering hard changes in these variables.

The objective function of the controller is given by the following convex criterion:

\[
J_k = \sum_{i=k+1}^{k+N} (\varepsilon_k^T Q \varepsilon_k + (u_{i-1} - u_{i-2})^T R (u_{i-1} - u_{i-2}))
\]

(11)

where \(k\) is the sampling time, \(N\) the horizon length that is finite, \(Q \geq 0\), \(R > 0\) and

\[
\varepsilon = y_k^d - y_k^M.
\]

(12)

In the first part of the criterion shown in Eq. (11), minimization of the error between the real process output and the process model and between the process output and the desired output is done. In the second part of this criterion, minimization of two successive inputs is done which allows the elimination of static error and smoothing the changes in the input. Therefore, the controller allows eliminating static error in a static state without requiring an integration of the error, which represents an advantage of the controller.

The control algorithm can be summarized in these steps:

- The desired process output is calculated and is considered to be the desired value on the whole horizon.
- Optimization of the cost function (11) allows one to calculate the vector of inputs \(u_k\) necessary for all the horizon. This ensures fulfilling criterion (11).
The first value of the inputs obtained is applied to the process.

This procedure is repeated, which results in a feedback control until the end of the reaction.

This allows predicting variations in the system using the model output on the horizon length. The stability of the controller is ensured for $N \to \infty$. Therefore, the horizon length should be chosen sufficiently long to ensure the process stability and the tracking of the output. However, a long horizon implies a long computation time. The horizon length should be chosen as a compromise between rapidity of computation and stability of the controller.

The matrices of the models of references (independent of the model) can be calculated before the reaction. This optimizes the computation time. In the case where no constraints on the inputs and on the states are considered, the matrices of control can be calculated only once, at the beginning of the reaction, and an analytical solution can be found for Eq. (11). However, in the case of a polymerization reactor, we have constraints on the input and on the states. In this case, Eq. (11) is solved by iteration and the matrices of control have to be recalculated at each step.

### 3.4. Process constraints

As mentioned above, the process under consideration has constraints on the input and on the states. The monomer flow rate has minimum and maximum limits. The jacket temperature is constrained by minimum and maximum limits and by the rate of change. The constraints on the state consist of a limitation on $N$ due to a physical limit of the saturation of the monomer particles and to upper and lower limitations of the reactor temperature (temperature range in which the polymerization takes place normally). The desired output is then limited by a maximum allowable heat of production.

The process constraints are described on the entire horizon. At a sampling time $k$, $u_k$ is given by

$$\alpha_k \leq u_k \leq \beta_k$$

where $\alpha_k$ and $\beta_k$ are calculated as functions of two kinds of constraints: on the amplitude and on the variation of the inputs:

$$\begin{align*}
\alpha_{k+i-1} &= \sup [u_{\min}, (u_{k-1} - i \times \Delta u_{\min})] \\
\beta_{k+i-1} &= \inf [u_{\max}, (u_{k-1} + i \times \Delta u_{\max})] \quad i = 1, \ldots, N
\end{align*}$$

where $\Delta u_{\min}$ and $\Delta u_{\max}$ are the minimum and maximum possible variations of the input.

These constraints are calculated for the new control variables ($w_1$ and $w_2$) from the original control variables ($F$ and $T_j$). They are therefore calculated online as follows, first for $w_1$:

$$\begin{bmatrix} w_{1, \min} \\ w_{1, \max} \end{bmatrix} = \begin{bmatrix} -R_P \\ F_{\max} - R_P \end{bmatrix}$$

and then for $w_2$:

$$\begin{bmatrix} w_{2, \min} \\ w_{2, \max} \end{bmatrix} = \begin{bmatrix} \frac{U A}{MC_p} (T_{j, \min} - T) + \frac{1}{MC_p} (Q_R + Q_{\text{feed}} - Q_{\text{loss}}) \\ \frac{U A}{MC_p} (T_{j, \max} - T) + \frac{1}{MC_p} (Q_R + Q_{\text{feed}} - Q_{\text{loss}}) \end{bmatrix}.$$
The maximum rate of change of the flow rate was assumed to be the same as the maximum flow rate since the pump can attain quickly its maximum flow rate. The maximum change of the jacket temperature was supposed to be 0.2 °C/10 s.

4. Controller development—implementation

The controller developed above was applied to control the emulsion polymerization of styrene. Constraints on the inputs, the flow rate of monomer and the jacket temperature, and constraints on the states, the reactor temperature and the saturation of the polymer particles, were considered. The main objective of the controller was to maximize the reaction rate.

A horizon length of 10 times the sampling time is used in the simulations. The maximum allowable heat was chosen to be 50 W. The temperature range in which the reaction is assumed to take place normally was put at 50–85 °C. The jacket temperature was therefore constrained at 50–85 °C with a maximum possible change of 0.2 °C/10 s. This ensures respecting the constraint on the reaction temperature. The maximum possible flow rate was assumed to be 1e−3 mol/s and the maximum variation between two successive inputs was also assumed to be 1e−3 mol/s. This means that no limitations on the rate of change of the flow rate were considered. The values of the flow rate and temperature change rates employed here are coherent with the experimental reactors of 1–3 L used in the laboratory.

Fig. 2 shows the evolution of the heat produced by the reaction. The rate of convergence is a function of the capacity of the thermostated bath to heat the jacket. It can be seen that during the time of convergence, the polymer particles were saturated in monomer (Fig. 3) due to the initial conditions that are usually applied in emulsion polymerization. This means that, if only the flow rate of monomer is used in the control strategy, then the heat of the reaction could not be maximized due to a saturation on the state $N$. In a multivariable control strategy, the maximum heat could be obtained more rapidly than the monovariable controller.

Fig. 4 shows the evolution of the reaction temperature. As it is controlled by the jacket temperature, its convergence is limited by the capacity of the bath. The reaction temperature was not constrained since the constraints were considered on the jacket temperature.

The controlled flow rate of monomer is shown in Fig. 5. At the beginning of the reaction, the polymer particles were saturated in monomer and therefore the monomer flow rate was set to zero. When the concentration of monomer in the polymer particles (Fig. 3) decreases, the monomer flow rate is increased to maximize $Q_R$. It is worth saying that if only the monomer flow rate was used in the control strategy, then during all the interval where the concentration of monomer in the polymer particles was saturated in monomer no control would take place (flow rate equals zero). This is therefore a big advantage of the multivariable controller.

Fig. 6 shows the controlled jacket temperature. This input is constrained in the amplitude (50–85 °C) and in the rate of variation (0.2 °C/10 s). In fact the rate of change is a function of the capacity of the thermostated bath used to heat the jacket and of the reactor volume. A good bath can heat a laboratory scale reactor at more than 10 °C/min. However, we considered a small rate of variation (1.2 °C/min) in order to be close to industrial conditions. Actually, a pilot scale reactor of 200 L can be heated at about 1–2 °C/min. The strategy is therefore valid for large scale reactors.

5. Conclusions

The receding horizon control was found to be an interesting tool for controlling emulsion polymerization processes. It takes into account constraints on the input and on the states of the model and predicts the evolution of the process which gives a stable trajectory under steady-state conditions. Therefore, the process rate could be optimized under safe conditions since the heat produced by the reaction did not exceed the maximum allowable heat.

In order to apply the linear receding horizon controller, the process had to be linearized using input–output linearization technique. In this case, the receding horizon controller is therefore based on the linear part of the model and predicts therefore variations in this part only. There is no prediction on the nonlinear part. If high variations of the nonlinear part are supposed to take place, or if this part of the model is supposed to have modeling errors, the nonlinear theory of the receding horizon control should be applied. This requires however a good knowledge of the nonlinear model which is not necessary in the technique proposed in this paper, since the parameter $\mu$ is not modeled but is only estimated online. If the model of the nonlinear part is not available, the linear receding horizon control combined with the input–output linearization would give equivalent results to the nonlinear predictive controller.
Fig. 2. Control of the heat produced by the reaction by the constrained receding horizon control combined with the input–output linearization by manipulating the monomer flow rate and the reaction temperature.

Fig. 3. The constraint on the state $N$ by limitation of the concentration of monomer in the polymer particles.

Fig. 4. Control of the reaction temperature (the state $T$).
Fig. 5. A constrained input, the flow rate of monomer.

Fig. 6. Controlled jacket temperature with the constraints on the amplitude (50–85 °C) and on the variation (0.2 °C/10 s).

References


