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Non-linear model based control of a propylene polymerization reactor

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Abstract

A modified generic model controller is developed and tested through a simulation study. The application involves model-based control of a propylene polymerization reactor in which the monomer conversion and melt index of the produced polymer are controlled by manipulating the reactor cooling water flow and the inlet hydrogen concentration.

Non-linear control is designed using a simplified non-linear model, in order to demonstrate the robustness of the control approach for modeling errors. Two model parameters are updated online in order to ensure that the controlled process outputs and their predicted values track closely. The controller is the static inverse of the process model with setpoints of the measured process outputs converted to setpoints for some of the state variables.

The simulation study shows that the proposed controller has good setpoint tracking and disturbance rejection properties and is superior to the conventional generic model control and Smith predictor control approaches.
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1. Introduction

Control of polymerization reactors is probably one of the most challenging issues in control engineering. The difficulties in operating such processes are numerous. Firstly, the process dynamics are often highly non-linear because of the complicated reaction mechanisms associated with the large number of interactive reactions. Secondly, on-line monitoring of polymer quality is often hampered by a lack of on-line measurements for key quality variables such as composition (or monomer conversion), molecular weight and copolymer composition [1]. If measuring quality variables is at all possible, there may still be a number of problems associated with these measurements, such as (i) sampling problems, (ii) large dead times, (iii) off-line analysis, and (iv) sometimes large measurement errors and/or high noise levels. A more detailed discussion of measurement difficulties in the field of polymerization can be found, amongst others, in Kiparssides [2]. To cope with the lack of on-line measurements of polymer quality, researchers have employed different inferential and estimation techniques [1,3–5].

Many articles have been published in the area of polymer reactor control in the last few years. They can be divided into linear and non-linear control approaches. There are numerous examples in the literature of linear control approaches applied to polymerization reactor control, such as, PI cascade control [6], dynamic matrix control [7,8], generalized predictive control [9] and adaptive internal model control [10]. Examples of the application of non-linear control approaches are, amongst others, globally linearizing control [11–13] and non-linear model predictive control [14,15]. There are also some approaches in which linear control is used, combined with non-linear models for setpoint updating [16].

Another type of control that has received moderate attention is generic model control (GMC). This method uses a non-linear process model and assuming a desirable process output trajectory, a non-linear control law can be derived. A recent example of its application in combination with extended Kalman filtering is found in Arnpornwichanop et al. [17].

In the current paper an approach similar to generic model control is being proposed, although its implementation and tuning is simpler. It implements the non-linear model of the process directly and gives an on-line estimation for the delayed measurements (Fig. 1); thus, there is no need to design an estimator, such as a Kalman filter. This control strategy is applied to the polymer-

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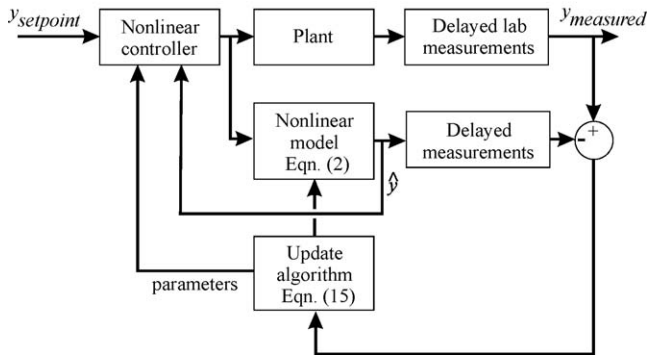 *E-mail address:* b.roffel@utwente.nl (B. Roffel).

Fig. 1. Reactor control based on simplified non-linear model, using model and controller update.

ization of propylene in a fully-filled hollow shaft reactor [18]. In case of a perfect non-linear model, a perfect non-linear controller can be designed. In case of a simplified non-linear model, the control system is improved by updating two model parameters of the simplified process and control models using an online model parameterization method. The efficiency of this control algorithm is compared to the performance of a conventional PI control system with Smith predictor dead time compensation.

The advantages of the proposed control approach over other approaches are: (i) there is no need for use of an extended Kalman filter to estimate unknown states or parameters, (ii) there is no need to solve the coupled set of non-linear ordinary differential equations, and (iii) the controller shows a good robustness the adaptation of the model parameters, as a result of which errors in dynamics and kinetics can easily be dealt with.

2. Non-linear control

Consider a process, which can be described by the following equations:

$$\frac{dx}{dt} = f(x, p) + g(x, u) + l(x, d)$$
$$y = h(x) \tag{1}$$

where x is the vector of state variables, y the vector of measured variables, u the vector of input variables, d the vector of disturbance variables, p the vector of process parameters, and h, f, g, l are the non-linear function vectors.

Let the model be a simplified description of the process with a different parameter set p and be given by:

$$\frac{d\hat{x}}{dt} = f(\hat{x}, \hat{p}) + g(\hat{x})u + l(\hat{x})d$$
$$\hat{y} = h(\hat{x}) \tag{2}$$

where the hat refers to the model values. In the development of the generic model control algorithm it is assumed that the derivative of y obeys the following equation [19]:

$$\frac{dy}{dt} = K_1(y_{sp} - y) + K_2 \int_0^{t_f} (y_{sp} - y)\, dt \tag{3}$$

were K_1 and K_2 are tuning parameters and y_{sp} is the setpoint value of the process output. Using Eq. (2), the derivative of the

state variable can be expressed as:

$$\dot{\hat{x}} = \dot{\hat{y}} \left[\frac{dh(\hat{x})}{d\hat{x}} \right]^{-1} \tag{4}$$

Substitution of the derivative of $\hat{x}$ in Eq. (2) results in:

$$\dot{\hat{y}} \left[\frac{dh(\hat{x})}{d\hat{x}} \right]^{-1} = f(\hat{x}, \hat{p}) + g(\hat{x})u + l(\hat{x})d \tag{5}$$

from which the equation for the control input vector can be derived:

$$u = \left\{ \begin{array}{c} K_1(y_{sp} - y) + K_2 \int_0^{t_f}(y_{sp} - y)\, dt \\ \dfrac{-(dh/d\hat{x})[f(\hat{x}, \hat{p}) + l(\hat{x})d]}{(dh/d\hat{x})g(\hat{x})} \end{array} \right\} \tag{6}$$

If the model is not linear in the control vector u, its values have to be computed through iteration. The parameters K_1 and K_2 are tuning parameters. If the model is not perfect, control performance will deteriorate, and the integral action in the controller will eliminate offset. However, it is preferred to use parameter estimation in order to update the model and thus account for parameter and structural errors. Farza et al. [20] suggested a simple non-linear observer, although other estimation schemes are possible, such as, e.g. a Kalman filter.

The tuning parameters K_1 and K_2 enable us to tune such that even some overshoot can be realized. This can primarily be realized through adjustment of K_1. A disadvantage of tuning for some overshoot in one variable is that it also affects the response of the other controlled variables. A smoother response without overshoot will show a smoother response of the other controlled variables.

If parameter update ensures that the model output tracks the true process output, the integral term in Eq. (6) is not required, since there will be no sustained offset in the controlled variables. Hence if $K_2 = 0$ and tuning of K_1 is done very conservatively to suppress variable interaction, one may wonder why one would not use a controller with both tuning values K_1 and K_2 set equal to zero, i.e. use a controller that is based on a static process model with parameter update. This may give a conservative response for setpoint changes, which approaches the open loop response of the system, however, disturbance rejection properties are expected to be good. The controller can then be calculated by the following set of equations:

$$u = \frac{-f(\hat{x}_{sp}, \hat{p}) - l(\hat{x}_{sp}, d)}{g(\hat{x}_{sp})}, \quad \hat{x}_{sp} = h^*(y_{sp}, \hat{x}) \tag{7}$$

where the estimated setpoint values of the output vector could be filtered values of the true setpoint values and the parameter $\hat{p}$ needs to be updated. In Eq. (7) the dimension of the y vector is usually smaller than the dimension of the x vector, therefore not all state variables setpoint values can be calculated, consequently, some setpoint values are set equal to the current values of the state variables from the model. This is also one of the main differences with generic model control where all the state variables follow from the process model and none of them have setpoint targets.

The parameter update should be realized such that the predicted process output values do track the true process measurements. Assume that it is required that the predicted process output values follow the true process outputs according to a first order response with time constant τ:

$$\tau \frac{\mathrm{d}\hat{y}}{\mathrm{d}t} = y - \hat{y} \tag{8}$$

In a steady state situation, when u and d are constant, an offset between y and $\hat{y}$ can only be minimized through adjustment of the model parameter $\hat{p}$. Eq. (8) can then be rewritten as:

$$\frac{\mathrm{d}\hat{p}}{\mathrm{d}t} = \frac{1}{\tau} \frac{y - \hat{y}}{\partial \hat{y}/\partial \hat{p}} \tag{9}$$

In some cases it may be easier to rewrite Eq. (9) in a somewhat different manner. In that case, Eq. (8) is differentiated with respect to $\hat{p}$ and substituting back into Eq. (9), which gives:

$$\frac{\mathrm{d}\hat{p}}{\mathrm{d}t} = -\frac{1}{\tau^2} \frac{y - \hat{y}}{\partial[\mathrm{d}\hat{y}/\mathrm{d}t]/\partial \hat{p}} \tag{10}$$

3. Process description

The hollow shaft reactor is an experimental extruder-like continuous reactor with internal recycling of the reaction medium. It has been designed for polymerizations at high viscosities up to a few hundred Pa s, and to work under high pressure and temperature, 250 bar and 250 °C. The reactor possesses the following properties: minimum dead volume, maximum recycle ratio, fast and predictable macro mixing; the recycle ratio and macro-mixing do not depend on the viscosity of the reaction mass in a wide range of viscosities [18].

The reactor is used for liquid-pool propylene polymerization with a multi-site heterogeneous Ziegler–Natta catalyst. The inlet flow to the reactor consists of pure monomer, catalyst and hydrogen, the latter is used as a transfer agent to provide a better control of the molecular weight of the produced polymer. A coolant removes the heat released due to polymerization.

One of the first considerations in establishing a control strategy is to arrange the system inputs and outputs into manipulated, controlled and disturbance variables. The polymerization process studied in this work has five inputs (manipulated and disturbance variables) and four controlled variables. Assuming fast cooling water dynamics, input variables include cooling water flow (F_w), outlet liquid flow rate (F) and feed rate of monomer ($F_{in}y_{m,in}$), hydrogen ($F_{in}y_{H_2,in}$), and catalyst ($F_{in}y_{cat,in}$). Reactor pressure (P), polymer melt index (MI), reaction conversion (C) and temperature (T) could be used as controlled outputs.

The reactor system is equipped with an automatic valve at the outlet that controls the reactor pressure P by manipulating the outlet flow F. In a pilot setup, it is aimed to keep the catalyst and monomer feed rates constant. Consequently, they will not be used in designing the control system. The control problem can therefore be simplified to a system with two manipulated inputs $F_{in}y_{H_2,in}$ and F_w, and two controlled variables MI and C.

4. Dynamic process model

The process model consists of dynamic material balances, a dynamic energy balance and algebraic equations for kinetic rate expressions and physical properties, as described in Appendix A. The mechanism of propylene polymerization is explained elsewhere [21]. In order not to complicate the model description too much, a number of assumptions were made, also listed in Appendix A.

The measurements of the process outputs, i.e. the monomer conversion and polymer melt index are subject to measurement delays, the delay for the conversion is 1 h and for the melt index it is 2 h.

The detailed model as described in Appendix A is used as the process description. If the model used for prediction of the controlled output is the same as this set of equations, a perfect prediction is obtained and the non-linear controller is a perfect non-linear controller.

5. Model simplification

In order to demonstrate the robustness of the control approach to modeling errors, the following deliberate simplifications were introduced. The rate of reaction, Eq. (A.9), is approximated by:

$$R_p = k_1 K_3 m y_c \bar{\rho}_m X \tag{11}$$

where K_3 is a tunable parameter with an initial value of 0.91, $\bar{\rho}_m$ the constant value for the monomer density and a 5% error in the calculation of k_1 is introduced.

Since the density is assumed constant, the equation for the outlet flow, Eq. (A.13), can be simplified to:

$$F = \left(\frac{F_{in}}{\rho_m} + R_p \left(\frac{1}{\rho_p} - \frac{1}{\rho_m} \right) \right) \rho \tag{12}$$

Eq. (A.19) was approximated by a linear first order differential equation:

$$0.9 \frac{\mathrm{dMI_c}}{\mathrm{d}t} = \mathrm{MI_i} - \mathrm{MI_c} \tag{13}$$

and the exponent in Eq. (A.16) was assumed to have a value equal to one. Another tunable parameter K_4 was therefore introduced in Eq. (A.16) to compensate for structural and parametric model mismatch:

$$\mathrm{MI_i} = K_4 \kappa X \tag{14}$$

with the initial value of K_4 equal to 0.88.

If the inaccuracies in the model are not known, a sensitivity analysis should be performed to find out which equations have the largest impact on the controlled variables in order to be a candidate for introduction of the parameter update.

Summarizing, the simplified model consists of Eqs. (A.1)–(A.20), with Eq. (A.9) replaced by Eq. (11), Eq. (A.13) replaced by Eq. (12), Eq. (A.16) replaced by Eq. (14) and Eq. (A.19) replaced by Eq. (13).

6. Parameterization of the simplified model

The implementation of a simplified model in non-linear controller design will usually result in unacceptable performance, the main problem being offset in the controlled variables. Thus, the parameters in the simplified model should be updated for prediction and control to be effective. Different updating approaches can be used. McAuley and MacGregor [1] implemented the recursive prediction error method for updating a set of parameters in the instantaneous melt index and density correlations. Because of its flexibility, other researchers [6,22,1] preferred to use extended Kalman filtering or other types of observers such as the Luenberger estimator [23]. Rhinehart and Riggs [24] used Newton's method and an intuitive relaxation method to calculate the model parameter update, our proposed method shows some resemblance to this method. In our case we do not use relaxation as a tuning parameter, instead, we propose to use a first-order time constant, which will be more acceptable from an engineering point of view.

As shown in Appendix B, update of the model parameters K_3 and K_4 proceeds according to the following equation:

$$K_{j,k+1} = K_{j,k} + \alpha_{pv,k} e_{pv,k} \tag{15}$$

in which k is the time step, $j = 3$ when the process variable pv is the conversion and $j = 4$ when the process variable is the melt index; e is the error between the measured process output and the estimated process output using the simplified model. The coefficient α depends on the process conditions.

7. Non-linear controller design

Starting point for the controller design is the static simplified model. The setpoint for the melt index $MI_{c,sp}$ can be written as a setpoint for ratio of hydrogen to monomer concentration by using Eq. (14):

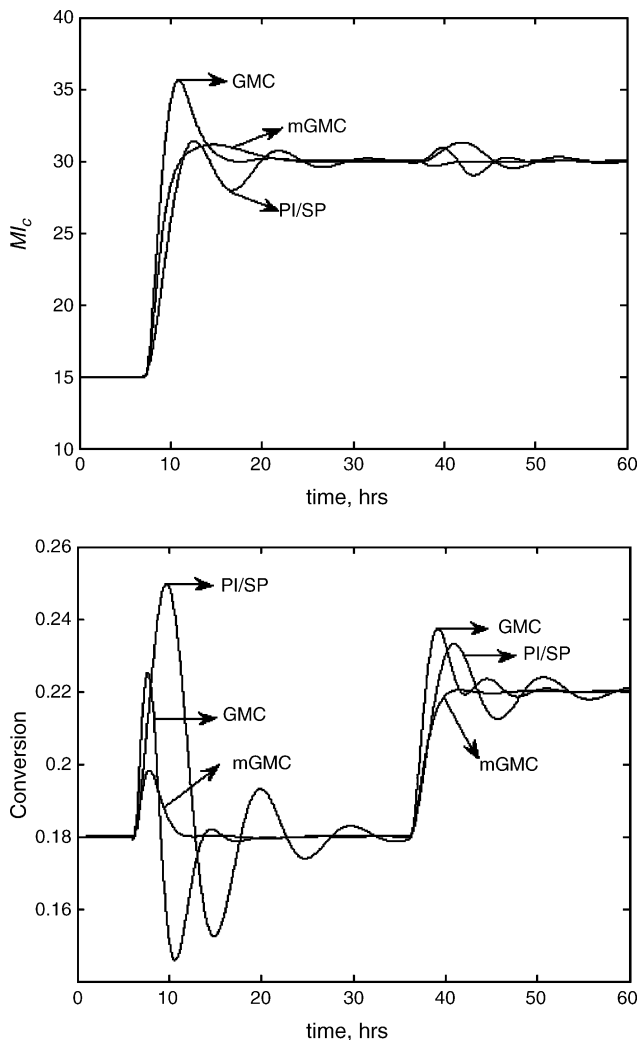$$X_{sp} = \frac{MI_{c,sp}}{\kappa K_4} \tag{16}$$

Fig. 2. Response to step changes in melt-index and conversion setpoints.
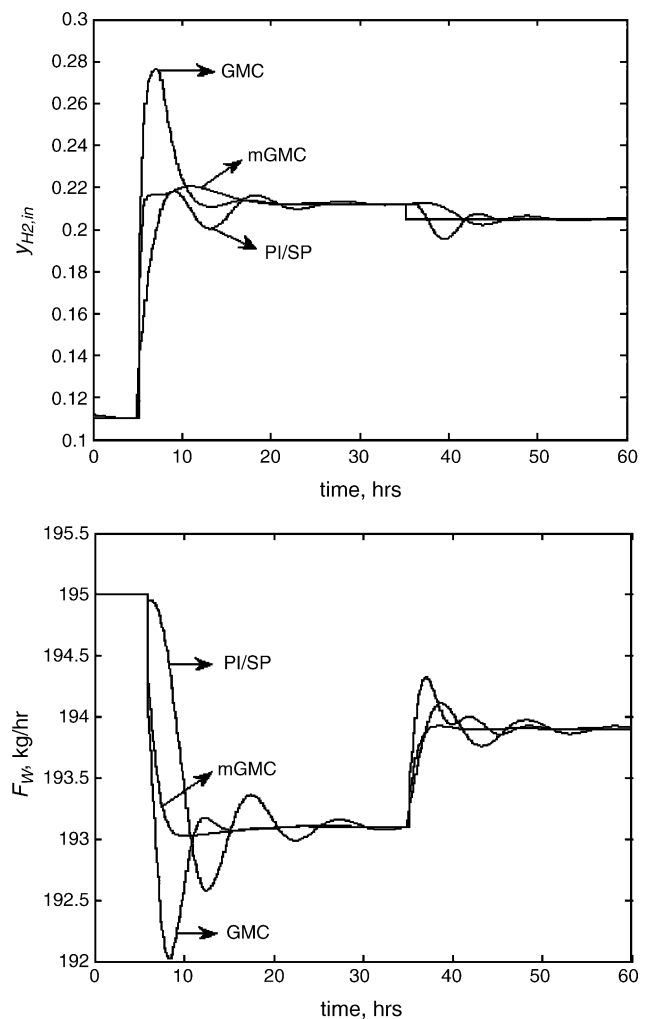

Fig. 3. Response of manipulated variable to step changes in melt-index and conversion setpoints.

The setpoint for the conversion can be written as a setpoint for the monomer concentration by using Eq. (A.20):

$$y_{m,sp} = y_{m,in}(1 - C_{sp}) \qquad (17)$$

By combining Eqs. (A.2)–(A.6), the inlet hydrogen concentration can be written as:

$$y_{H_2,in} = y_{H_2,sp} + \frac{R_{H_2}}{F_{in}} \qquad (18)$$

in which R_{H_2} follows from the simplified model equations and F_{in} is a measured variable. Eq. (A.12) can be used to calculate the specific heats of the reactor inlet flow and the fluid inside the reactor, using the reactor temperature from the simplified model. The static version of the reactor energy balance, Eq. (A.11) can subsequently be used to calculate the reactor jacket temperature:

$$T_{j,sp} = T_m + \frac{1}{UA}[F_{in}(C_{p,in}T_{in} - C_p T_m) - R_p \Delta H_{R,p}] \qquad (19)$$

after which the linear relationship between the jacket temperature and cooling water flow can be used to compute the water flow through the reactor jacket. T_m represents the reactor temperature from the simplified model.

The control law of Eqs. (18) and (19) is rather similar to the one that can be derived for generic model control, however, there are two main differences: (i) this controller does not have proportional integral control action to ensure that the process output follows a prescribed trajectory. In this case model updating ensures that there will be no process-model mismatch and the process output approaches setpoint; (ii) the setpoints for the controlled process outputs are converted to setpoints for the same number of state variables. This can easily be achieved, since in reactor modeling component concentrations and temperatures are often measured and they are also the state variables of the model. The control approach as described in this section is therefore called mGMC, modified generic model control.

8. Conventional proportional-integral control with dead time compensation

In many polymer producing companies, classical control techniques such as proportional integral (PI) control is still being used, the designed non-linear controller will therefore be compared to a conventional PI controller with Smith predictor dead time compensation. Using the relative gain array method (RGA)
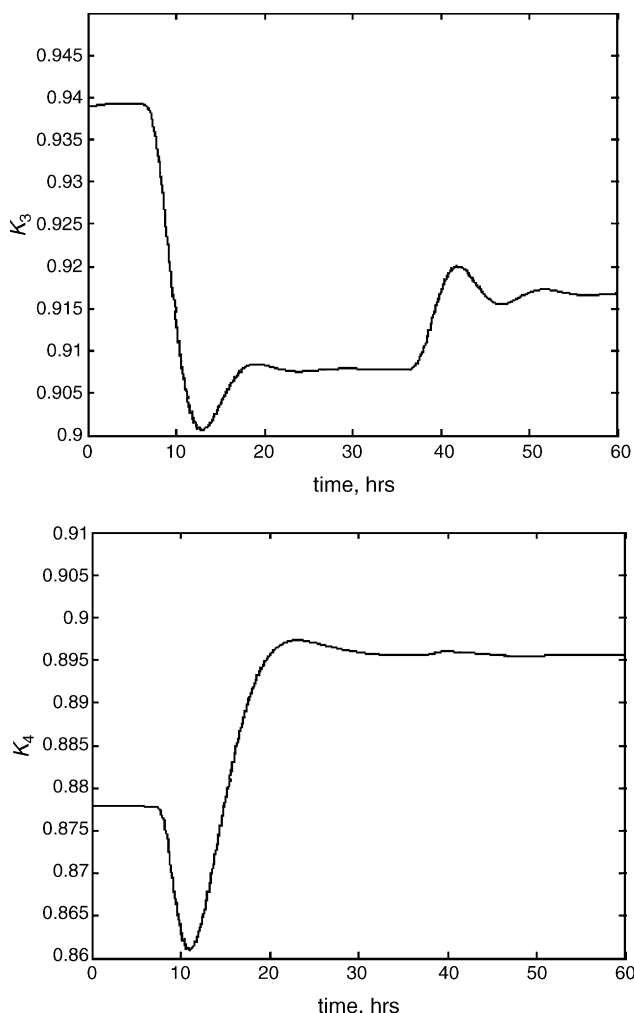

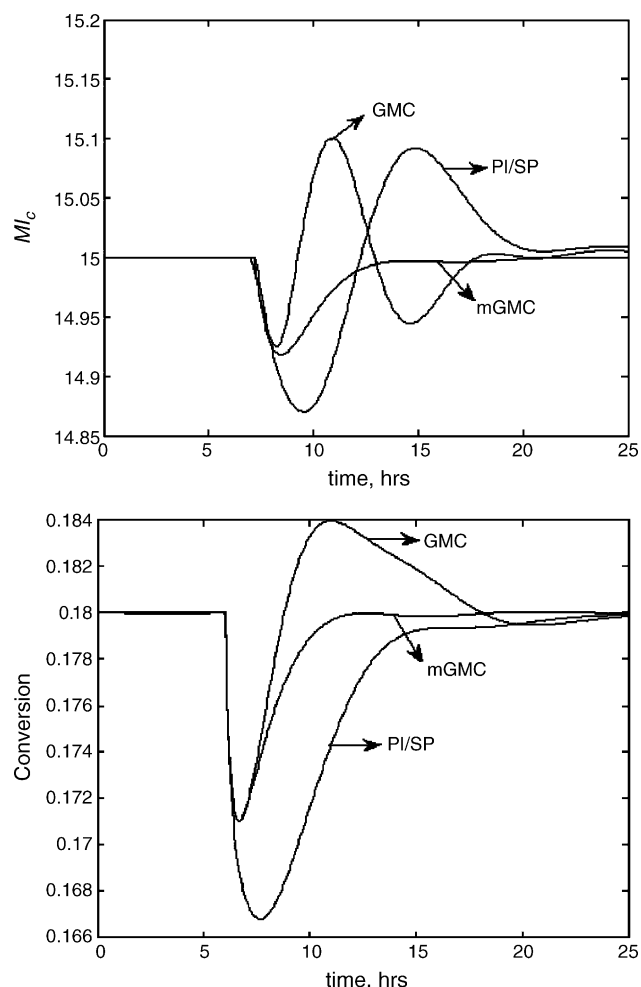Fig. 4. Update of model parameters during setpoint changes.

Fig. 5. Controlled variable responses to a +20% disturbance in feed rate.

[25], it was found that the melt index can be best controlled by the inlet hydrogen concentration, $y_{H_2,in}$, and the conversion by the jacket temperature, T_j (i.e. cooling water flow). Due to the presence of measurement dead times of 1 and 2 h for conversion and melt index, respectively.

9. Results and discussion

Since the process model is represented by a simplified model, the responses will show process/model mismatch. As a result, the parameter update scheme will come into effect to ensure that the model output tracks the process output. It should be mentioned that the update scheme uses fixed values of $\alpha_{C,k}$ in Eq. (A.4) and $\alpha_{MI,k}$ in Eq. (A.7), equal to 0.6 and 0.001, respectively, since changes in these values were found to be limited to a maximum change of 20%.

9.1. Performance of the non-linear and PI control algorithms

In this section, the performance of the following control approaches will be discussed: (i) the generic model controller, (ii) the modified generic model controller and (iii) the PI–Smith

predictor controller. Performance is examined for four different cases:

- polymer grade change,
- conversion setpoint change,
- disturbance rejection,
- error in dead time of the melt index measurement of 1 h and conversion measurement of 0.5 h.

In the closed-loop simulations, it is assumed that the values of MI_c and C are available every 2 and 1 h, respectively; within these time intervals, estimated values are obtained using the property models and the parameter-updating scheme. The controller algorithms are executed every 6 min.

In addition to monitoring the controlled variables, also the manipulated variable moves are monitored.

Figs. 2 and 3 show the closed-loop responses of the controlled and manipulated variables for a change in melt index setpoint from 15 to 30 at time $t = 5$ h and a change in conversion setpoint from 0.18 to 0.22 at time $t = 35$ h. Fig. 2 shows the response of the controlled outputs, Fig. 3 shows the responses of the manipulated variables for completeness. Controller tuning settings are given in Table 1. In Fig. 2 it can be seen that the generic model
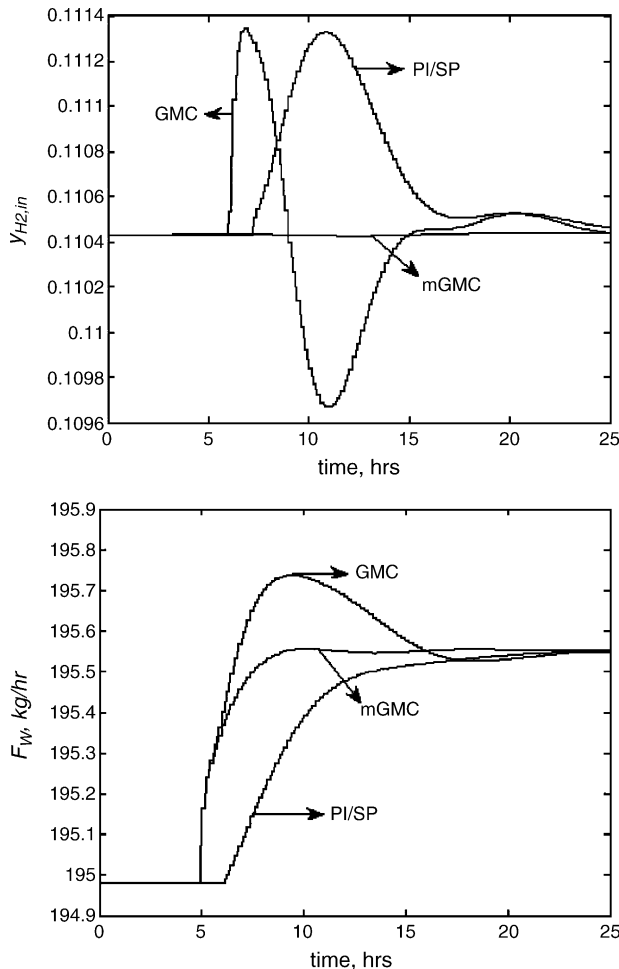

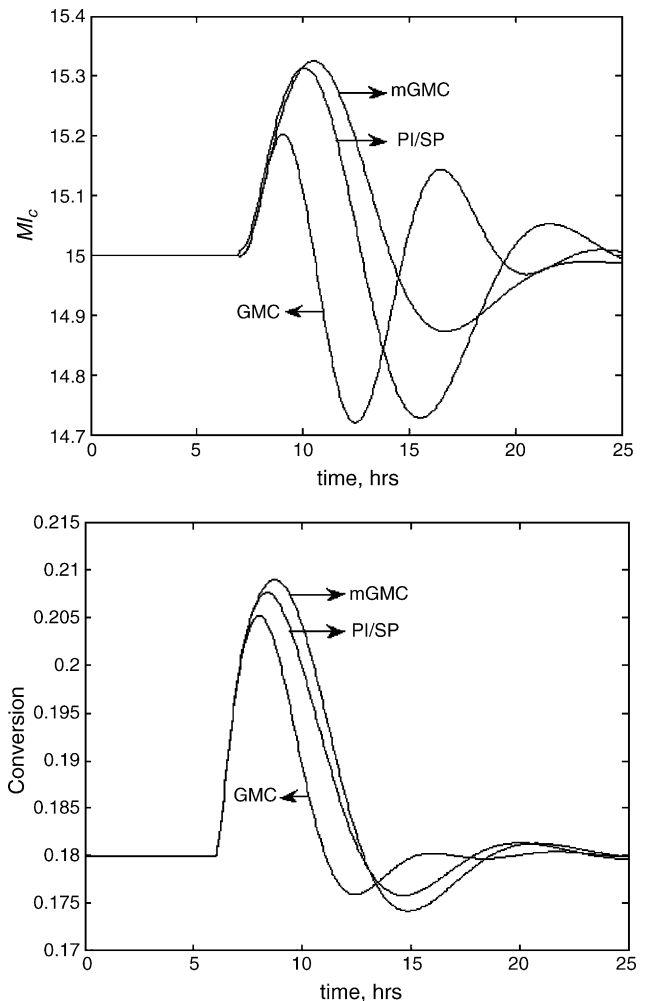Fig. 6. Manipulated variable responses to a 20% disturbance in feed rate.

Fig. 7. Controlled variable responses to a 20% disturbance in catalyst activity.

Table 1
Controller tuning for setpoint changes

PI/SP conversion controller	$K_c = 2$, $T_i = 2.0$
PI/SP melt index controller	$K_c = 0.00125$, $T_i = 4.0$
Melt index setpoint filter	$\tau = 0.2\,h$
Conversion setpoint filter	$\tau = 0.1\,h$

controller gives a rather large overshoot for a setpoint change in the cumulative melt index, at the same time the interaction is visible in the conversion response (around $t = 10\,h$). Detuning the GMC controller improves the response, since the interaction between the variables is reduced but also reduces the speed of response.

On the one hand the GMC controller decouples the process variables through the static inverse of the process model, on the other hand a PI controller is added which introduces process variable interaction. The Smith predictor controller also suffers from the interaction between the process variables, detuning slows down the response. As can be seen, the mGMC controller outperforms the other two controllers. Fig. 4 shows the parameter update during these transients. As can be seen, the response is smooth.

Another issue that should be considered is load or disturbance rejection. The first type of disturbance that will be considered is a measurable change in the propylene inlet flow rate; the rejection tests were conducted with a 20% increase in inlet flow rate at $t = 5\,h$, retaining the controller settings for setpoint changes. As can be seen from Fig. 5, also in this case the mGMC controller outperforms the other two controllers. The melt index is not affected much by the disturbance, the conversion suffers from a momentary decrease (at $t = 6\,h$) which is the largest for the Smith predictor controller (Fig. 6).

Another type of unmeasurable disturbance that is considered is a 20% change in the catalyst activity. As can be seen from Fig. 7, the GMC controller outperforms the other two controllers. This is due to the aggressive tuning of the integral action in case of GMC control, this also causes the response to be slightly more oscillatory than the other two responses. All controllers reach a new steady state around the same time, the maximum deviation for the mGMC and SP/PI controller are somewhat larger than for the GMC controller in case of the melt index. The response of the conversion to this change is very much the same for all controllers (Fig. 8).
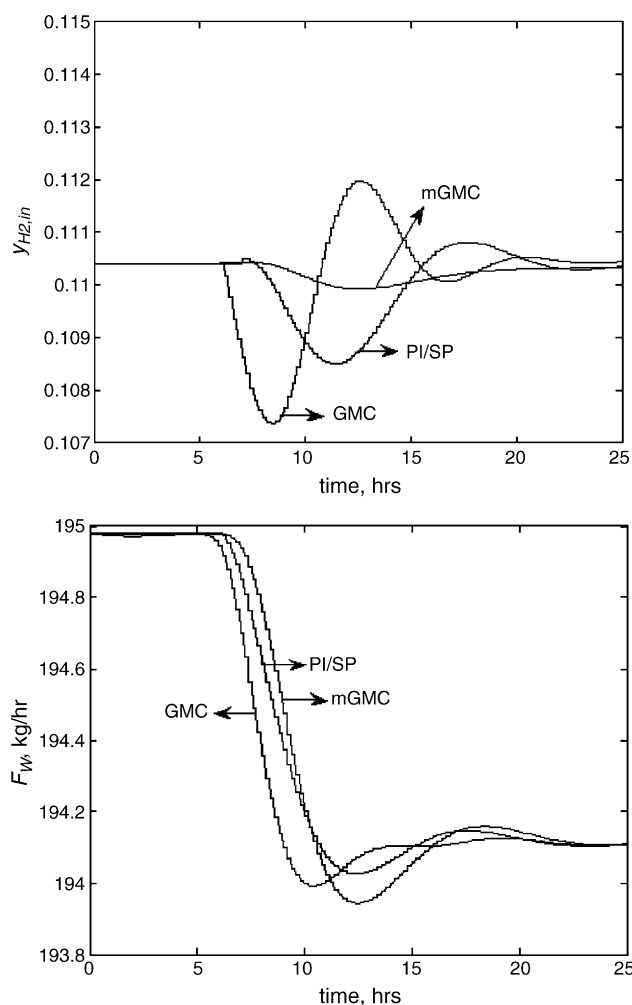

Fig. 8. Manipulated variable responses to a 20% disturbance in catalyst activity.
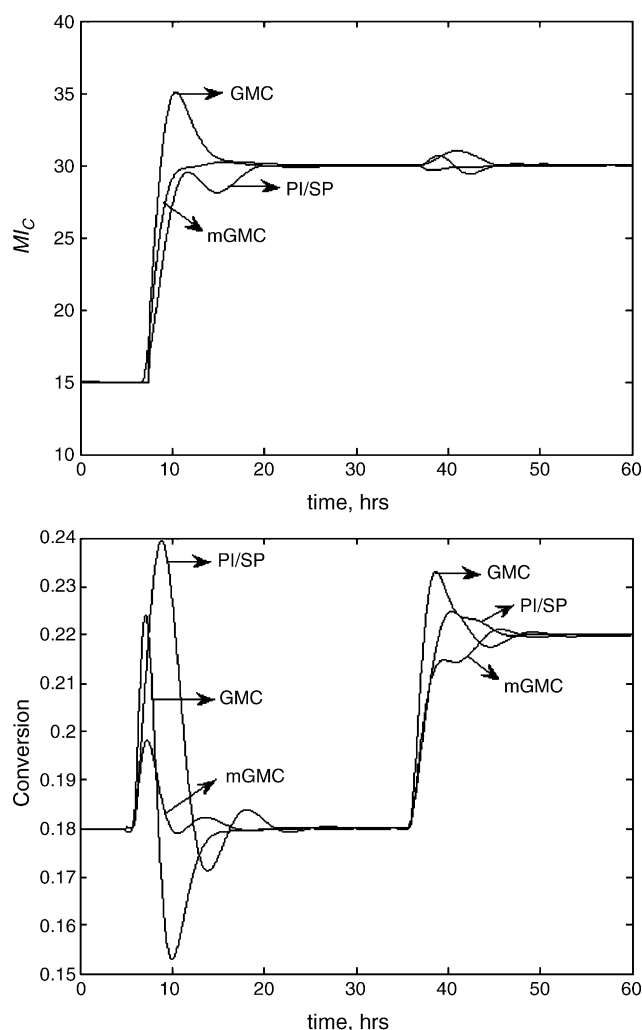
Fig. 9. Controlled variable responses to setpoint changes in case of dead time discrepancy.
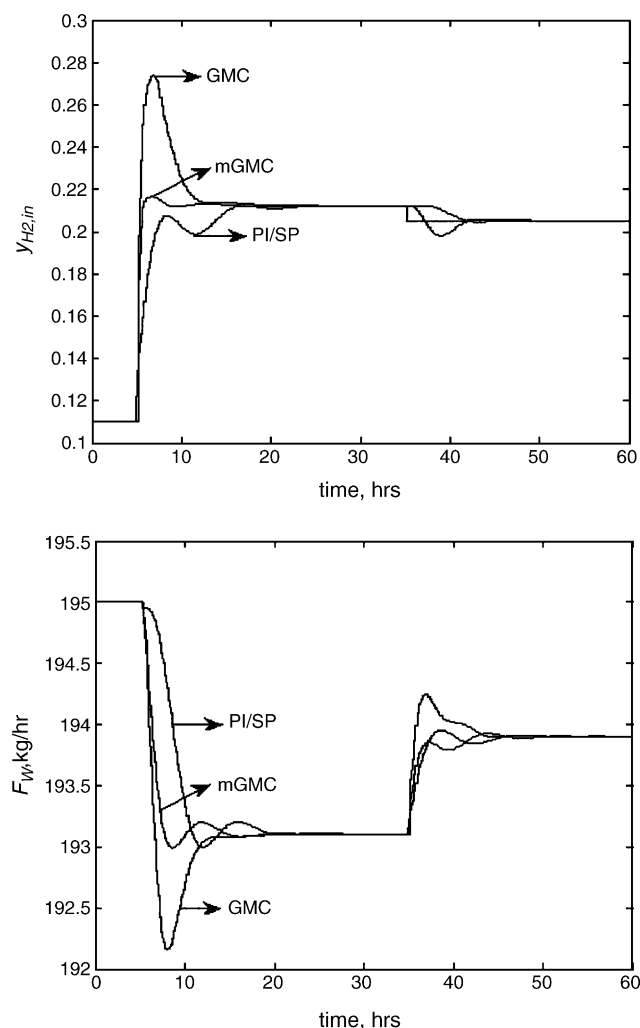
Fig. 10. Manipulated variable responses to setpoint changes in case of dead time discrepancy.

Another type of disturbance that could occur is a discrepancy in sampling times of the controlled outputs from the model and from the plant. The dead time of the melt index is assumed to be 2 h, the dead time of the conversion is assumed to be 1 h. Figs. 9 and 10 show the impact of a discrepancy in dead time between the process and the model, the dead time of the process melt index is decreased by 1.0 h and the dead time of the process conversion is decreased by 0.5 h. All controllers are affected, the mGMC controller performed better than the other two controllers.

10. Conclusions

Non-linear process model based control was studied for control of liquid propylene polymerization under varying conditions. The controller manipulated the hydrogen flow rate and cooling water flow to follow the setpoints for cumulative melt index and reaction conversion and to remove the effects of various process disturbances. The non-linear control strategy was called modified generic model control, it used the static inverse of the process model with setpoints

of the measured process outputs converted into setpoints for the state variables. In addition, model parameters were updated to ensure good setpoint tracking and disturbance rejection. Tuning of the proposed control strategy is simple, the time constant of the setpoint filters can be adjusted and the speed at which the parameter update is accomplished can be selected.

Performance of the control strategy was compared to a generic model controller and a proportional integral controller with Smith predictor dead time compensation.

Closed loop simulations revealed that for setpoint changes the modified generic model controller was superior to the other two controllers, also for measurable feed disturbances it outperformed the other control approaches.

For unmeasurable disturbances in the catalyst activity, the response of the melt index was somewhat faster for the generic model controller due to aggressive tuning of the integral action, this also lead to a more oscillatory response.

Appendix A. Dynamic model of the polymerization process

In order to develop a model of limited complexity, the following assumptions were made:

- The polymerization reactions are irreversible and first order with respect to each reactant.
- The reactor is ideally mixed. Thus, no temperature and concentration gradients are present. If the stirrer speed in the reactor is in the range of 100 rpm, the reactor is (macro) mixed within 40–80 s, meanwhile, the reactor average residence time may reach 1 h.
- The reactor is fully filled, no gas phase is present in the reactor.
- The energy produced due to mixer rotation is negligible.
- The catalyst decay through different chemical mechanisms at various types of active sites may be lumped together into a single deactivation. In addition, the active site concentration decreases in accordance with a first order decay mechanism constant [26].
- Monomer equilibrium concentration near the active sites is assumed the same as the monomer bulk concentration. Thus, it can be calculated using a monomer density correlation.
- The reactor contains two phases: (i) a liquid monomer phase and (ii) a polymer phase. The liquid phase consists of propylene monomer with dissolved hydrogen and the polymer phase consists of crystalline polymer and amorphous polymer, which is swollen with the monomer.

In this model, all variables should have a hat in order to show they are model values, however, it has been omitted for reasons of simplicity of notation.

The overall mass balance of the reactor can be described as:

$$\frac{dm}{dt} = F_{in} - F \tag{A.1}$$

where m is the total mass inside reactor and F the outlet mass flow rate in kg/h, $F_{in} = 1.0$ kg/h. The monomer mass balance is:

$$m\frac{dy_m}{dt} = F_{in}(y_{m,in} - y_m) - R_p \tag{A.2}$$

y_m is the mass fraction of monomer in the outlet flow stream, and R_p is the propagation reaction rate. The hydrogen mass balance can be described as:

$$m\frac{dy_{H_2}}{dt} = F_{in}(y_{H_2,in} - y_{H_2}) - R_{H_2} \tag{A.3}$$

in which y_{H_2} is the hydrogen mass fraction in g H_2/kg material inside the reactor and R_{H_2} is the apparent hydrogen consumption rate. An apparent consumption rate is used, since the reaction rate constants for the hydrogen reactions, transfer with hydrogen and dormant sites reactivation, are not known for the catalyst system used in this work.

The hydrogen consumption rate, R_{H_2}, can be calculated from:

$$R_{H_2} = \frac{2R_p}{42.1 P_n} \tag{A.4}$$

where 2 and 42.1 are the molecular weight for hydrogen and propylene, respectively, R_p is the polymerization rate. The number average degree of polymerization P_n can be calculated from:

$$P_n = \frac{2}{q\text{PD}} \tag{A.5}$$

where PD is the polydispersity of the produced polymer, for the catalyst used in this study it has an average value of 6.8. The polymerization termination probability q is experimentally determined from [21]:

$$q = d + eX, \quad X = \frac{0.02104 y_{H_2}}{y_m} \tag{A.6}$$

where X is the molar ratio of hydrogen to monomer in the reactor. The values of d and e are given in Table 2.

Based on the assumption that the catalyst is being activated before injecting it, the mass balance for the active catalyst, y_c, can be described as:

$$m\frac{dy_c}{dt} = F_{in}(y_{c,in} - y_c) - R_d \tag{A.7}$$

where R_d is the deactivation reaction rate. The concentration of the deactivated catalyst, y_d, can be calculated from the following balance:

$$m\frac{dy_d}{dt} = F_{in}(y_{d,in} - y_d) + R_d \tag{A.8}$$

The reaction rates are calculated using the following equations:

$$\begin{aligned} R_p &= k_1 m y_c \rho_m X \\ R_d &= k_d m y_c \end{aligned} \tag{A.9}$$

in which k_1 and k_d are rate constants and ρ_m is the monomer density. For the rate constants the following equations hold:

$$\begin{aligned} k_1 &= K_{01} + K_{02}T + K_{03}T^2 \\ k_d &= k_{d0}e^{-E_{a1}/RT} + k_{d1}e^{-E_{a2}/RT}(1 - e^{-E_{a3}/X}) \end{aligned} \tag{A.10}$$

Table 2

Thermodynamic and physical parameters for propylene polymerization

Parameter	Value
Physical parameters	
Reactor volume (V)	1.86×10^{-3} m^3
Reactor heat transfer area (A)	0.0961 m^2
Thermodynamic parameters	
Overall heat transfer coefficient (U)	1.62 MJ/h K m^2
Heat of propagation reaction ($\Delta H_{R,p}$)	2.03 MJ/kg
Specific heat of polypropylene ($C_{p,p}$)	2.25×10^{-3} MJ/kg K
Density of polypropylene (ρ_p)	900 kg/m^3
Specific heat of propylene ($C_{p,m}$)	
a	2.785×10^{-3} MJ/kg K
b	-9.18×10^{-6} MJ/kg K^2
c	2.93×10^{-8} MJ/kg K^3
Density of propylene monomer (ρ_m)	
$\rho_{m,a}$	-263.7 kg/m^3
$\rho_{m,b}$	6.827 kg/K m^3
$\rho_{m,c}$	-0.0143 kg/K^2 m^3
Parameters for Eq. (A.16)	
κ	6818.3
γ	1.03
Parameters for Eq. (A.17)	
β	-2.34
for $X < 0.00144$	
d	5.32×10^{-5}
e	0.115
else	
d	1.52×10^{-4}
e	0.0405
K_0 in Eq. (A.10)	[-204256.61, 1153.3314, -1.626207]
k_{d0}	3746 h^{-1}
k_{d1}	1.748×10^{-7} h^{-1}
E_{a1}/R	1620.8 K
E_{a2}/R	5570.7 K
E_{a3}	498.9

Since the mass of the reactor wall is not small compared to the mass of the reactor contents, the heat capacities of the reactor wall and reactor contents are lumped together in the reactor energy balance. This balance can be written as:

$$\begin{aligned} (mc_p + m_s c_{p,s})\frac{dT}{dt} &= F_{in}(c_{p,in}T_{in} - c_pT) - R_p \Delta H_{R,p} \\ &\quad - UA(T - T_{jacket}) \end{aligned} \tag{A.11}$$

where the subscript 's' refers to steel. The dependence of the jacket temperature on the cooling water flow can be calculated from a static energy balance and is approximated by a simple linear relationship. The specific heat of the reactor contents can be given by:

$$c_p = y_m(a + bT + cT^2) + C_{p,p}y_p \tag{A.12}$$

The values of the coefficients are summarized in Table 2. $C_{p,p}$ is the heat capacity of the polymer, it is assumed to have a constant value.

Since the reactor is completely filled and there is a significant change in density because the low-density monomer is converted

to the high-density polymer, the reactor outlet flow rate, F, will vary. It can be shown that the following equation can be used to calculate this flow [27]:

$$F = \left(\frac{F_{\text{in}}}{\rho_{\text{m}}} + R_{\text{p}} \left(\frac{1}{\rho_{\text{p}}} - \frac{1}{\rho_{\text{m}}} \right) - m y_{\text{m}} \frac{\text{d}T}{\text{d}t} \frac{1}{\rho_{\text{m}}^2} \frac{\text{d}\rho_{\text{m}}}{\text{d}T} \right) \rho \quad (A.13)$$

where ρ_{p} is the polymer density and ρ_{m} is the monomer density, which can be calculated from:

$$\rho_{\text{m}} = -\rho_{\text{m},a} + \rho_{\text{m},b}T - \rho_{\text{m},c}T^2 \quad (A.14)$$

The constants of this equation are summarized in Table 2. ρ is the density of the reaction mixture inside the reactor, it can be written as:

$$\rho = \frac{\rho_{\text{m}}\rho_{\text{p}}}{y_{\text{m}}\rho_{\text{p}} + y_{\text{p}}\rho_{\text{m}}} \quad (A.15)$$

The easily available measurements of the melt index are often utilized to control the polymer quality in a homo-polymerization reactor. In polyolefin production plants, it is well-known that the concentration ratio of hydrogen to monomer, X, has a strong effect on the instantaneous melt index MI_{i}. In the literature [1,28] different relationships have been proposed to relate MI_{i} to X. In this work, the following relationship is used:

$$\text{MI}_{\text{i}} = \kappa X^{\gamma} \quad (A.16)$$

The numerical values of κ and γ are obtained from experimental work and are listed in Table 2. Because the direct on-line measurement of the instantaneous polymer molecular properties is not practically realizable, the melt index is correlated to the polymer average molecular weight (M_{w}). In this study the following semi-empirical equation is employed [29]:

$$\text{MI}_{\text{c}} = \alpha \bar{M}_{\text{w}}^{\beta} \quad (A.17)$$

the values of α and β were calculated by fitting MI measurements to the off-line measurements of M_{w}, the values are presented in Table 2. To calculate the cumulative melt index, the differential balance for the cumulative weight average molecular weight, $\bar{M}_{\text{w}}$, is employed [30]:

$$\frac{\text{d}\bar{M}_{\text{w}}}{\text{d}t} = \frac{1}{m_{\text{p}}}(y_{\text{p,in}}F_{\text{in}}[\bar{M}_{\text{w,in}} - \bar{M}_{\text{w}}] + R_{\text{p}}[M_{\text{w}} - \bar{M}_{\text{w}}]) \quad (A.18)$$

where m_{p} is the mass of polymer inside the reactor. Substituting Eq. (A.17) into Eq. (A.18), in addition to the assumption of a free-polypropylene inlet stream, i.e. $y_{\text{p,in}} = 0$, results in the following differential equation for the cumulative melt index:

$$\frac{m(1 - y_{\text{m}})}{\beta R_{\text{p}}} \frac{\text{d}}{\text{d}t}\text{MI}_{\text{c}} = [\text{MI}_{\text{i}}^{1/\beta}\text{MI}_{\text{c}}^{1-(1/\beta)} - \text{MI}_{\text{c}}] \quad (A.19)$$

which is a first-order relationship with variable gain and time constant.

To complete the model description, the monomer conversion is calculated from:

$$C = 1 - \frac{y_{\text{m}}}{y_{\text{m},t=0}} \quad (A.20)$$

Appendix B. Parameterization of the simplified model

To parameterize the conversion model, a relationship between the conversion and the model parameter K_3 has to be derived. This relationship can be obtained by differentiating the conversion equation, Eq. (A.20) with respect to time:

$$\frac{\text{d}C}{\text{d}t} = -\frac{1}{y_{\text{m},t=0}} \frac{\text{d}y_{\text{m}}}{\text{d}t} \quad (B.1)$$

Using Eqs. (A.2) and (11), this equation can be rewritten as:

$$\frac{\text{d}C}{\text{d}t} = \frac{-1}{y_{\text{m,in}}m}[F_{\text{in}}(y_{\text{m,in}} - y_{\text{m}}) - k_1 K_3 m y_{\text{c}} \bar{\rho}_{\text{m}} X] \quad (B.2)$$

Since the relationship between C and K_3 is represented by a differential equation, Eq. (10) is used for parameter update. The difference between the conversion using the plant measurement and the estimated conversion using the simplified model is used to make incremental adjustments to K_3 at each execution interval of the discrete controller:

$$e_{C,k} = C_{\text{plant},k} - C_{\text{model},k} \quad (B.3)$$

After a simple Euler discretization of Eq. (10), the updating of the parameter K_3 can be evaluated according to:

$$K_{3,k+1} = K_{3,k} + \frac{\Delta t e_{C,k}}{\tau^2(\partial(\text{d}C/\text{d}t)/\partial K_3)} = K_{3,k} + \alpha_{C,k}e_{C,k} \quad (B.4)$$

The partial derivative of the conversion-time derivative with respect to the adjustable parameter K_3 is calculated using Eq. (B.2):

$$\frac{\partial(\text{d}C/\text{d}t)}{\partial K_3} = \frac{k_1 y_{\text{c}} \bar{\rho}_{\text{m}} X}{y_{\text{m,in}}} \quad (B.5)$$

In the melt index model, Eq. (14) the parameter K_4 is a tunable parameter. The difference between the cumulative melt index from the plant measurement and the simplified model is used to make the corrections to the model parameter every controller execution interval:

$$e_{\text{MI},k} = \text{MI}_{\text{c,plant},k} - \text{MI}_{\text{c,model},k} \quad (B.6)$$

The value of K_4 is updated using the discretized version of Eq. (10):

$$K_{4,k+1} = K_{4,k} + \frac{\Delta t e_{\text{MI},k}}{\tau^2(\partial(\text{dMI}_{\text{c}}/\text{d}t)/\partial K_4)}$$
$$= K_{4,k} + \alpha_{\text{MI},k}e_{\text{MI},k} \quad (B.7)$$

The partial derivative of the cumulative melt index-time derivative with respect to the adjustable parameter K_4 is calculated by substituting Eq. (14) into Eq. (A.19) and differentiation with respect to K_4:

$$\frac{\partial(\text{dMI}_{\text{c}}/\text{d}t)}{\partial K_4} = \frac{R_{\text{p}}}{mK_4(1 - y_{\text{m}})}\text{MI}_{\text{i}}^{1/\beta}\text{MI}_{\text{c}}^{1-(1/\beta)} \quad (B.8)$$

Appendix C. Nomenclature

A	area of heat transfer (m^2)
C	reaction conversion
C_p	heat capacity of monomer (kJ/kg K)
F	outlet flow rate from the reactor (kg/h)
F_{in}	inlet flow rate to the reactor (kg/h)
$\Delta H_{R,p}$	heat of polymerization (MJ/kg)
k_d	deactivation constant (1/h)
k_p	propagation constant (m^3/g_{cat} h)
m	total mass inside the reactor (kg)
MI_i	instantaneous melt index (g/10 min)
MI_c	cumulative melt index (g/10 min)
M_w	weight average molecular weight (kg/kmol)
P_n	number average degree of polymerization
R_d	catalyst deactivation reaction rate (kg/m^3 h)
R_{H_2}	average hydrogen reaction rate (m^3/g_{cat} h)
R_p	propagation reaction rate (kg/h)
t	process time (h)
T	reactor temperature (K)
T_j	jacket temperature (K)
U	heat transfer constant (MJ/h m^2 K)
V	reactor volume (m^3)
X	hydrogen molar ratio (mol H_2/mol)
y_c	active catalyst mass fraction (g/kg)
y_d	deactivated catalyst concentration (g/kg)
y_{H_2}	hydrogen mass fraction (g/kg)
y_m	monomer mass fraction in the reactor (kg/kg)
y_p	polymer mass fraction in the reactor (kg/kg)

Greek symbols

ρ	density of reaction mixture (kg/m^3)
ρ_m	monomer density (kg/m^3)

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