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Optimizing Control of a Continuous Polymerization Reactor ⋆

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Abstract: In this contribution we study the application of non-linear model predictive control to a continuous polymerization of acrylic acid in tubular reactors with multiple side injections of monomer. The background of this work is to transfer the polymerization from semi-batch to continuous operation. Model Predictive Control (MPC) is the obvious candidate to control such a multi-input system. The reactor configuration and polymerization reaction make the application of MPC very challenging. The controller employs a discretized dynamic pde model of the process and optimizes the productivity of the plant online while keeping the product quality parameters within the predefined constraints. The spatial domain of the model is discretized by applying the Weighted Essentially Non Oscillatory (WENO) scheme. Besides testing the controller for a nominal case in which the control model is identical to the existing plant, the controller has been simulated for two model-plant mismatch cases, caused by fouling and feed impurities. For the both cases, a moving window estimation scheme is applied to estimate the unknown parameters and to update the model used by NMPC. The results show that the controller can increase the product throughput considerably and has a robust performance in the presence of the model-plant mismatch. Moreover, the effect of formulating the quality constraints as soft constraints is studied.

Keywords: continuous polymerization, tubular reactors, optimizing control, pde models, parameter estimation.

1. INTRODUCTION

In the framework of the European Project F3-factory (Buchholz 2009) new flexible continuous modular production concepts are being developed which employ intensified equipment. One of the case studies is the production of water soluble acrylic acid-based polymers in tubular reactors instead of semi-batch reactors. The reactor considered in this work consists of eight tubular reactor modules which are connected in series and equipped with static mixers to obtain efficient mixing of the reactants at laminar flow conditions. The free radical homo-polymerization of acrylic acid is investigated as a first step towards more complex reaction systems. In the plant, shown in figure 1, there are four possible locations for feeds of monomer (u_1, u_2, u_3 and u_4). The jacket temperature can be set via a thermostat, offering an additional control input.

We assume that the monomer concentrations at four positions distributed along the reactor can be measured by infrared spectroscopy. This measurement system has been established in parallel work. The molecular weight of the polymer is assumed to be measured at the outlet of the reactor by means of an online viscosity measurement. The system has several inputs (the feed flow rates of monomer and the zone temperatures) and reacts with large time delays due to the plug flow between the inputs and outputs. Model Predictive Control (MPC) is the obvious candidate for the control of such a multi-input control problem. While the standard implementations of MPC employ cost functions which penalize the deviations of the states or outputs from the targets and the control moves, in this work the controller has been formulated to maximize the product throughput while meeting the constraints on product quality (Engell 2007). The controller copes with model inaccuracies, in particular a fouling case which decreases the heat transmission between the reactor and jacket, as well as a retardation of the start of the reaction due to impurities in the feed.

2. PROBLEM DESCRIPTION

2.1 Process

This case study deals with the continuous production of poly acrylic acid (PAA) in a modular, intensified tubular reactor, which is currently tested in the lab in Dortmund. The results in this paper are based on a realistic model of this process. The polymerization is performed in aqueous solution at 20% solid content by a free radical mechanism. Sodium persulfate acts as thermal initiator and the nominal reaction temperature is $80°C$. Throughout the whole process, all components including the polymer are soluble in water, thus precipitation of solids does not occur. As the product is used directly as an aqueous polymer solution, no further separation of solvent is required.

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Figure 1 shows the P&ID diagram of the polymerization plant. It consists of eight tubular reactor modules equipped with static mixing elements and jackets. The internal volume of modules one to four is 45 ml where modules five to eight are larger and each has a volume of 130 ml. This configuration leads to a total residence time of 42 minutes at the nominal flow rate of 1 kg/hr.

Through the side injections of the monomer, which are distributed along the reactor, it is possible to influence the product quality. The reactor is divided into four sections consisting of two reactor modules each. A side injection at the beginning of every section is installed and the temperature of each section is controlled independently via the jacket.

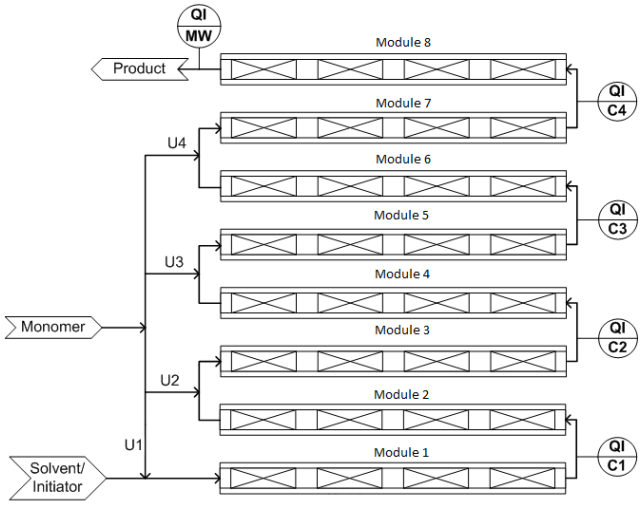

Fig. 1. Flow sheet of the modular continuous polymerization plant with side injections, heating system and analytics ($C1-4$: monomer concentration; MW : molecular weight).

The measurements from the process which are assumed to be available to the controller are the monomer concentration measured at C_4 and the average molecular weight at the reactor outlet. These information are available at the real plant, as described below.
The monomer concentration can be measured inline by mid-IR spectroscopy in between the reactor modules (C1-4). For this purpose, attenuated total reflection (ATR) fiber optic probes are installed at the reactor flanges. The monomer concentration can be extracted from the acquired spectra with a relative error of 1%. This measurement is available at roughly every 25 seconds.

At the outlet of the reactor an online sensor for the liquid viscosity is installed. The average molecular weight (M_w), which is an important measure for the product quality can be derived from this measurement. For this purpose, a correlation is applied, based on the empirical Mark-Houwink equation, which describes the correlation between the intrinsic viscosity and the viscosity average molecular weight. At constant temperature, known polymer concentration c and with negligible changes of the shape of the molecular weight distribution (MWD), an extended correlation for the absolute viscosity η holds:

$$\eta = K M_w^{\alpha} c. \tag{1}$$

The polymer concentration is known from the monomer feed assuming full conversion. The shape of the MWD does not change significantly in continuous free-radical solution polymerization over a wide range of operating conditions. That is why M_w at the reactor outlet is available at the same sampling rate as the viscosity, which is up to 1 Hz for the installed vibrating rod viscosimeter. (Chevrel et al. 2012).

3. PROCESS MODEL

Based on the balance equations for energy and all relevant components in the reaction mixture, a rigorous dynamic model of the process has been formulated. It is assumed that radial mixing is perfect and axial dispersion is negligible. The free radical polymerization of acrylic acid is modeled by the terminal model approach. A thermally activated initiator decomposes in two radical fragments, which start the polymer chains by reacting with a monomer molecule. The resulting macroradicals propagate by adding more monomer units. As the speed of all reaction steps is assumed to be independent of the chain length, the concentration of the macroradicals is lumped into one species λ_0. Chain termination happens when two macroradicals meet, resulting in dead polymer chains that do not participate in the reaction anymore. The temperature dependent rate coefficients $k_i(T)$ are modeled by an Arrhenius approach. The resulting nonlinear partial differential equations model (PDE) is shown in the equations 2 to 10.

$$\frac{\partial c_I}{\partial t} = -u\frac{\partial c_I}{\partial z} + \frac{\partial}{\partial z}\left(D_{ax}\frac{\partial c_I}{\partial z}\right) - k_d(T)c_I \tag{2}$$

$$\frac{\partial c_M}{\partial t} = -u\frac{\partial c_M}{\partial z} + \frac{\partial}{\partial z}\left(D_{ax}\frac{\partial c_M}{\partial z}\right) - k_p(T)\lambda_0 c_M \tag{3}$$

$$\frac{\partial \lambda_0}{\partial t} = -u\frac{\partial \lambda_0}{\partial z} + \frac{\partial}{\partial z}\left(D_{ax}\frac{\partial \lambda_0}{\partial z}\right) + 2fk_d(T)c_I - 2k_{tc}(T){\lambda_0}^2 \tag{4}$$

$$\frac{\partial \lambda_1}{\partial t} = -u\frac{\partial \lambda_1}{\partial z} + 2fk_d(T)c_I + k_p(T)\lambda_0 c_M - k_{tc}(T)\lambda_0\lambda_1 \tag{5}$$

$$\frac{\partial \lambda_2}{\partial t} = -u\frac{\partial \lambda_1}{\partial z} + 2fk_d(T)c_I + k_p(T)c_M\left(\lambda_0 + 2\lambda_1\right) - k_{tc}(T)\lambda_0\lambda_2 \tag{6}$$

$$\frac{\partial \mu_0}{\partial t} = -u\frac{\partial \mu_0}{\partial z} + k_{tc}(T){\lambda_0}^2 \tag{7}$$

$$\frac{\partial \mu_1}{\partial t} = -u\frac{\partial \mu_1}{\partial z} + k_{tc}(T)\lambda_0\lambda_1 \tag{8}$$

$$\frac{\partial \mu_2}{\partial t} = -u\frac{\partial \mu_1}{\partial z} + k_{tc}(T)\lambda_0\lambda_2 + k_{tc}(T){\lambda_1}^2 \tag{9}$$

$$\frac{\partial T}{\partial t} = -u\frac{\partial T}{\partial z} + \frac{\partial}{\partial z}\left(\lambda\frac{\partial T}{\partial z}\right)\frac{2k}{R}\left(T_{jac} - T\right) + k_p(T)\lambda_0 c_M\left(-\Delta h_p\right) \tag{10}$$

The temperature in the reactor jacket T_{jac} is assumed to be constant along the reactor axis, which can be achieved with a high coolant flow rate. The dynamics of temperature changes in the thermostat are assumed to be negligible, which is approximately true for lab scale equipment. The concentrations of initiator and monomer are denoted with c_I and c_M respectively. The method of moments is applied to describe the polymer chain length distribution, with λ_0 as the concentration of macroradicals and μ_0 as the concentration of deactivated polymer chains. Higher moments are λ_1, λ_2, μ_1 and μ_2, which are required to calculate the molecular weight averages. The weight average molecular weight is chosen as main quality measure of the product and is defined as:

$$M_w = \frac{\mu_2 + \lambda_2}{\mu_1 + \lambda_1} \tag{11}$$

As mentioned in the section 2.1, the controller uses only M_w and C_4. None of these variables depends on μ_0, thus the balance describing the dynamics of this state can be left out from the model used in the controller.
The spatial domain is discretized using the weighted essentially non-oscillatory (WENO) scheme, a method which is especially designed to cope with steep fronts of the states' profile along the axial domain without the need of a very fine discretization (Bouaswaig et al. 2009) (Liu et al. 2011). The 8 remaining pdes are discretized in 200 points which result in a system of 1600 odes that is solved by the Matlab solver $ode15s$.

3.1 Model Predictive Control

The reactor in focus at this work is controlled by a model-based controller where the input moves are optimized over a finite prediction horizon using the rigorous process model. Standard implementations of MPC employ a tracking criterion as cost function which penalizes deviations of the outputs or states and possibly of the inputs from the target values (Findeisen et al. 2004). In this work, we follow the idea of online optimizing control (Engell 2007). The target of the controller is to maximize the reactor throughput under the constraints of product quality. For the continuous reactor, maximizing the throughput implies to maximize the sum of all injections of monomer into the reactor (u_1, u_2, u_3 and u_4 in figure 1). In order to keep the product quality within the specifications, constraints are set for the last measurement point of the monomer concentration (C_4) and for the molecular weight of the product measured at the outlet of reactor (M_w). The optimization problem is formulated as:

$$\begin{aligned} \max_{u_{1k},u_{2k},u_{3k},u_{4k}} \; & \Phi\ (x(t_k), u(t_k), N_c, N_p) \\ := \; & \sum_{j=k}^{k+N_p} (u_{1j} + u_{2j} + u_{3j} + u_{4j}) \\ s.t. \; & \begin{cases} M_{w,min} \leq & M_w \leq M_{w,max} \\ & C_4 \leq C_{max} \end{cases} \end{aligned} \tag{12}$$

The temperature (T_{jac}) does not enter the cost function directly but is used to fulfill the constraints. The dynamic model of the system is implicit in the constraints of this formulation since we are using a sequential approach for the implementation of the NMPC. We assume that the initial condition of the model is known.
In section 4.1(a), we show the simulation results in which the constraints of this optimization problem are treated as hard constraints and are applied for the whole prediction horizon. In section 4.1(b), the performance of the controller is compared with the case that the quality constraints are formulated as soft constraints. For the implementation at the real plant, model-plant mismatch have to be taken into account. This is discussed in section 4.2 and we show there that a moving horizon scheme can be used to estimate the unknown parameters and to update the model used by NMPC which gives a robust performance of the controller. For the results presented in this work, the algorithm SNOPT from the TOMLAB package is used to solve the optimization problem that appears in NMPC.

4. RESULTS

4.1 Regulation scenario

a) Quality constraints as hard constraints

As the first case, it is assumed that the NMPC controller uses a model which can perfectly reflect the behavior of the plant and maximizes the product throughput as stated in equation 12. Tuning the controller parameters (N_p and N_c) has a great influence on the performance of the controller for this problem. The residence time of the reactor for the nominal flow rate, as mentioned before, is about 2600 seconds which implies that the prediction horizon should be at least this long. However, the controller continuously manipulates the total flow rate of the reactor which causes a drift in the states and can expedite or postpone the effect of a particular control move depending on the next control movement. In order to take this behavior into account and to ensure the stability of system, the prediction horizon is chosen longer (6000 seconds). The control horizon has the length of one and the manipulated variables stay constant for the whole prediction horizon. Using a longer control horizon, results in too many decision variables for the optimizer which increases the computation time considerably and is not favorable for the real-time application. The sampling time is 100 seconds.

For the nominal case (no regulation), the produced polymer has a molecular weight of $77.1 Kg/mol$ (M_w) and a monomer content of $321 mol/m^3$ at (C_4). By admitting some deviation of the molecular weight from the nominal value, and small increase of the monomer content, the regulation problem can be formulated as in equation 13.

$$\begin{aligned} \max_{u_1,u_2,u_3,u_4} \; & \sum_{j=k}^{k+60} (u_1 + u_2 + u_3 + u_4) \\ s.t. \; & \begin{cases} 75000 \leq & M_w \leq 80000 \\ & C_4 \leq 330 \end{cases} \end{aligned} \tag{13}$$

The cost function itself is trivial to solve and the problem is defined by its constraints. Considering the quality constraints as hard constraints and the prediction horizon of 6000 seconds ($N_p = 60$) implies that 120 constraints for this optimization problem must be satisfied. Providing a feasible initial point for the optimizer helps to solve the problem efficiently. This can be done by considering that at the beginning of the simulation, the reactor is in steady state and the produced polymer fulfills all the necessary constraints. This combination of the inputs is chosen as the initial point for the solver at the beginning of the simulation ($t = 0$). For the next sampling times, the output of the optimizer at the previous sampling time is considered as the initial point.

Figure 2 depicts the reactor outputs and manipulated variables for this simulation. The controller tends to inject the largest portion of the monomer at the first feed point (u_1), which makes sense because the injected monomer from this position has the longest residence time inside the reactor. For the fluctuation of the outputs (figure 2(a) and 2(b)) before reaching the steady state, the reasons are:

(1) The system is highly nonlinear and does not have a monotically increasing/decreasing step response.

(2) The inputs are applied at different positions acting with different delays in the measured variables. For example, suppose the controller manipulates u_1 and u_4 simultaneously. Since u_4 acts in a position closer to the sensors, its effect is sensed earlier. After a while when the effect of change in u_4 is almost over, the change in u_1 affects the measurements and depending on u_1 (and the other manipulated variables), a fluctuation in the outputs can be observed.

(3) As mentioned before, manipulating the flow rate inside the reactor shifts the states in the axial direction which causes an extra fluctuation of the states.

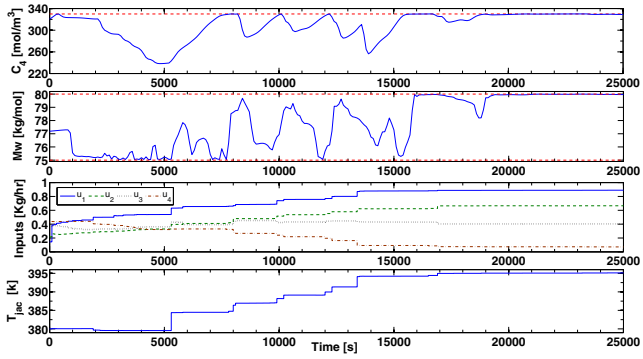

Fig. 2. Manipulated and controlled variables for the regulation scenario ($N_P = 60, N_C = 1, T_s = 100sec$).

The controller drives the operating point of the reactor to a higher temperature and increases the total feed of monomer to the reactor as expected. Figure 3 shows that the controller increases the product throughput by 76.1% from the initial condition.

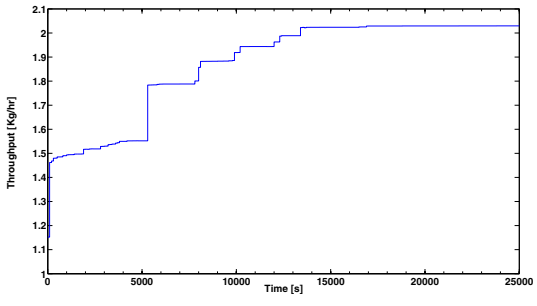

Fig. 3. Throughput increase by the controller.

b) Quality constraints as soft constraints

Considering the quality constraints of the product as hard constraints has the benefit that the required specification for the products are strictly met for the whole simulation time and no off-specs are produced. However, a large settling time results. Moreover an optimization problem with hard constraints is more difficult to solve. It is possible to handle the quality constraints as soft constraints which means their violations from the predefined bounds are tolerated but penalized in the cost function. The cost function (Φ) is formulated as:

$$\Phi = -\Phi_1 + \gamma\Phi_2 = -\Phi_1 + \gamma\left(\Phi_{21} + \Phi_{22} + \Phi_{23}\right) \tag{14a}$$

$$\Phi_1 = \sum_{j=k}^{j=k+N_p} (u_1 + u_2 + u_3 + u_4) \tag{14b}$$

$$\Phi_{21} = \sum_{j=k}^{j=k+N_p} (max((C_{4j} - C_{4u}), 0))^2 \tag{14c}$$

$$\Phi_{22} = \sum_{j=k}^{j=k+N_p} (max((M_{wj} - M_{wu}), 0))^2 \tag{14d}$$

$$\Phi_{23} = \sum_{j=k}^{j=k+N_p} (min((M_{wj} - M_{wl}), 0))^2 \tag{14e}$$

where the parameters with a subscript of "l" and "u" are the lower and upper bounds of these parameters. The first part of the cost function, (J_1), maximizes the product throughput and the second part, (J_2), minimizes the violation of controlled variables from the predefined bounds. γ is a tuning parameter (but not a decision variable) and determines the relative importance of J_1 and J_2. The control horizon is one and the prediction horizon is 60. The simulation results are shown in figures 4 and 5.

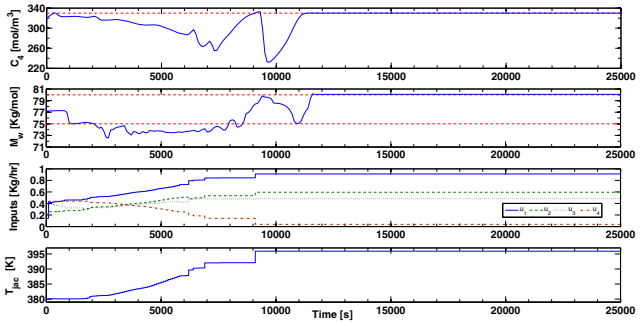

Fig. 4. Manipulated and controlled variables obtained by formulating the quality constraints as soft constraints (optimization problem stated in equation 16.)

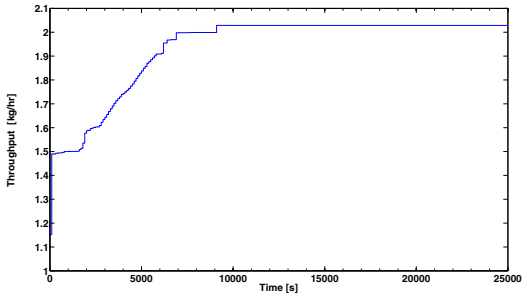

Fig. 5. Throughput of the reactor for the regulation case with the soft quality constraints. The optimizing controller increases the throughput by 76.0%.

Comparing the profiles of the controlled variables in figures 2 and 4 shows that the formulation with soft constraints results in a shorter settling time but off-specs are produced for a while in the transition phase. For this case, the throughput increases continuously with small steps at almost every sampling time which is not the case for the formulation with hard constraints. The main difficulty with this formulation is the tuning of the parameter γ because it affects the productivity of the reactor and the

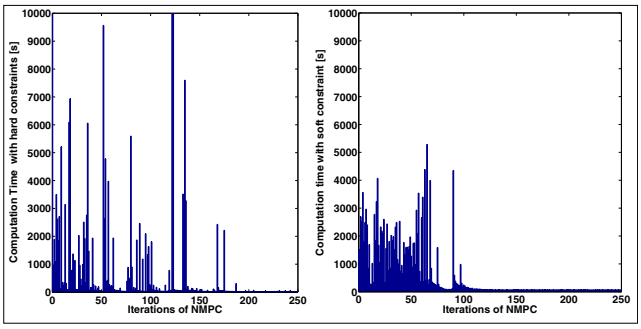

Fig. 6. Computation times in the cases of using hard and soft constraints.

violations of the controlled variables from the bounds. For the presented simulation, γ has a value of 15. With this value, the controller increases the product throughput by 76.0%. Furthermore, as shown in figure 6, the formulation with soft constraints results in a much shorter computation time which is favorable for the real-time application [1].

4.2 Regulation in the presence of model-plant mismatch

a) Fouling

It is almost impossible to build a model for a given plant which can perfectly reflect its behavior. Model-plant mismatch can result from neglecting some dynamics of the system or some parameters of the system can change over time.

Fouling is a phenomenon which is likely to happen in almost every chemical reactor. In this study, the controller attempts to maximize the product throughput while fouling decreases the heat transmission between the reactor and the jacket. A common approach to compensate model inaccuracies is to compare the available measurements with the simulation results at every sampling time and to correct the model outputs by this error in the next sampling time (bias correction). By applying this method, it is assumed that the error caused by the model inaccuracies stays almost constant for the whole prediction horizon which is not the case here. We therefore use a moving window approach to estimate the unknown parameters and to update the model used by the controller. Moving window estimation is an optimization-based approach which uses the current and past measurements to estimate parameters which can be formulated as follows:

$$\min_{\underline{\alpha}} \sum_{j=1}^{p} \left\| y_{k-j} - \hat{y}_{k-j} \right\|_2^2 \qquad (15)$$

where $\underline{y}$ is the vector of measurements, $\underline{\hat{y}}$ is the vector of estimated outputs, $\underline{\alpha}$ are the decision variables (unknown parameters) and p is the length of the window. At every sampling time (subscript k) a horizon including the last 35 samples (subscript j) is used to solve the above mentioned optimization problem. The sampling time of the parameter estimator is the same as of the controller and equals 100 seconds. We assume that fouling affects the reactor differently at each segment of the reactor (every two modules of the reactor constitute a segment). This assumption is reasonable because the material has a higher viscosity in the last segments which causes more fouling. By solving the optimization problem given by equation 15 the fouling factors are estimated. A fouling factor is defined as:

$$\alpha_i = \left(1 - \frac{heat\ transfer\ coef.\ with\ fouling}{nominal\ heat\ transfer\ coef.}\right) * 100, \quad i = 1, 2, 3, 4$$

Figure 7 shows the result of the estimation of these coefficients. The sensors are located at the last segment of the reactor and the optimizer can sense the change of the unknown parameter at this segment immediately (decision variable α_4). For this reason, α_4 is estimated accurately at the very beginning of the simulation while the other unknown parameters are estimated accurately once their effect is visible at the measurements. The horizon for the estimator has to be chosen long enough to capture the effect of all unknown parameters.

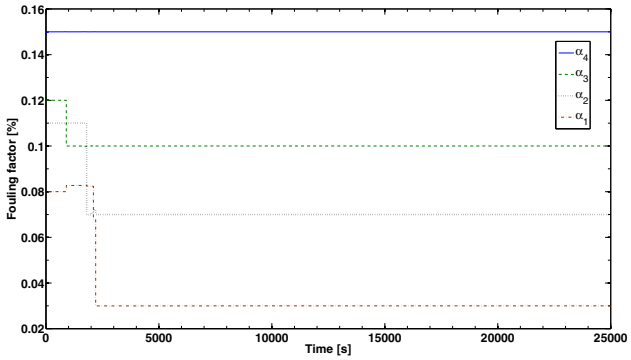

Fig. 7. Decrease of the heat transmission between the reactor and the jacket by 3%, 7%10% and 15%in the first, second, third and fourth segments of the reactor and estimation of the fouling factors using a moving window estimation scheme.

As shown in figure 8, the controller can cope with this type of model-plant mismatch properly and fulfill the constraints strictly and increases the product throughput by 75.7%. The slight difference to the nominal case (no fouling) results from the fact that a reactor affected by fouling is less efficient. Since the controller tends to inject the largest portion of the monomer at the first feed point, fouling in this segment is critical and affects the attainable throughput.

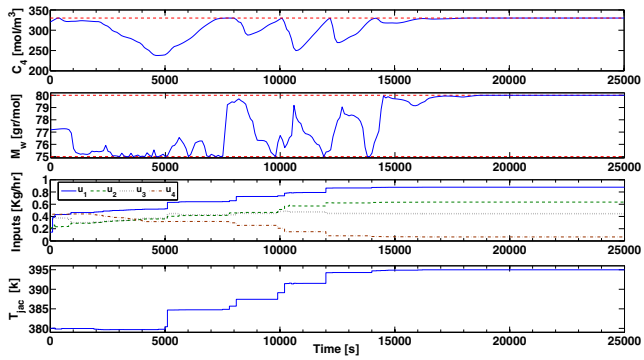

Fig. 8. Manipulated and controlled variables for the regulation scenario with model inaccuracies caused by fouling and estimation of the fouling factors.

b) Fouling and variation of the reaction rate

As the second case we investigate the robustness of the proposed controller against two possible inaccuracies of

[1] Intel Core i7-2600K CPU @ 3.40GHz - 8.0 GB RAM

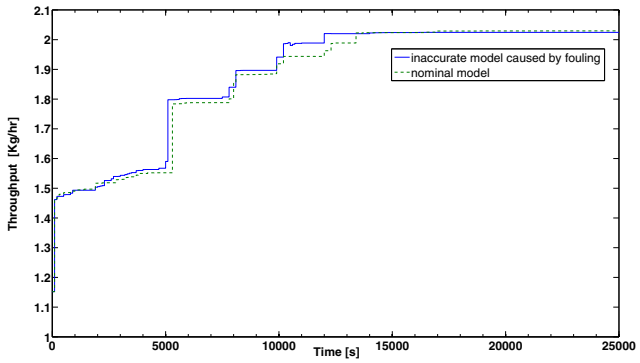

Fig. 9. Evolution of the throughput with and without considering model inaccuracies caused by fouling.

the model. We assume that besides fouling, impurities in the raw material have decreased the initiator reaction rate by 5%. Similar to the previous case, a moving window estimation scheme is used to estimate the fouling factors and the actual reaction kinetics.

As it can be seen in figure 10, the controlled variables are kept within the defined bounds for the whole simulation time and the product throughput is increased by 75.4%. The product throughput for this case and for the nominal model are shown in figure 11.

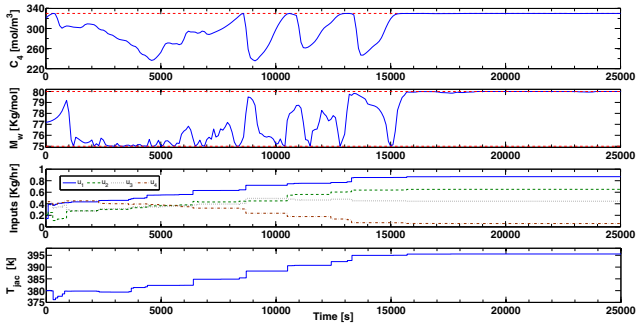

Fig. 10. Manipulated and controlled variables for the regulation case where fouling and feed monomer impurities have caused a model-plant mismatch. It is assumed that fouling has affected the model like the previous case and that feed monomer impurities have decreased the monomer reaction rate by 5%.

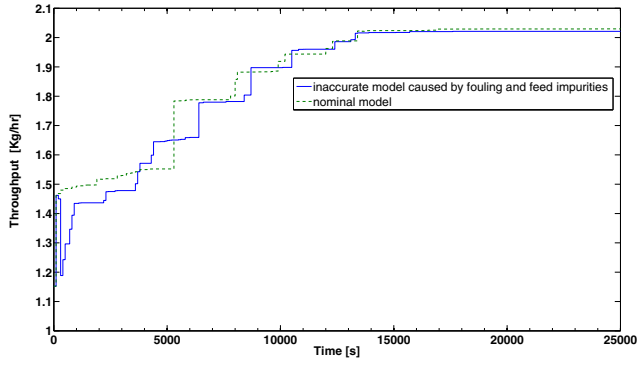

Fig. 11. Evolution of the throughput with and without considering model inaccuracies caused by fouling and different reaction rate.

5. CONCLUSION

An application of optimizing model-based control was studied in this work which aims at the maximization of the product throughput of a continuous polymerization reactor while the product quality is set as constraint. The simulation results show that the controller can increase the product throughput considerablly and cope with model inaccuracies properly. Two cases of such model inaccuracies caused by fouling and feed impurities were studied. For these cases a moving window estimation scheme is used to estimate the unknown (uncertain) parameters and to update the model. Using this updated model, the optimizing controller is able to meet the problem constraints.

Finding the solution of the optimization problem stated in eqn. 12 requires a high computational effort. However by being content to a suboptimal solution when the optimal solution can not be found on time, the controller can be used for the real time applications. The use of a moving window estimator adds another computational burden.

Treating the quality constraints as hard constraints has the benefit that the quality constraints are strictly met and no off-specs are produced but it results in a long settling time and a demanding optimization problem. On the other hand, soft constraints result in a less complicated optimization problem and a smoother and faster transient, but the quality constraints are violated slightly in the tranisient phase and off-specs are produced. However, since the reactor is a continuous reactor and the product is usually stored in large vessels, these violations of the constraints are averaged to the whole production time. The tuning of the parameter γ, which controls the relative importance of the throughput maximization and violations of the controlled variables from the bound, is the main difficulty of using soft constraints. It seems that the hard constraints can be replaced by the soft constraints if the proper values for the tuning parameter γ and possibly modified quality constraints are used. Consideration of the measurement noise and estimation the initial states of the model are two open issues.

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