

Constrained nonlinear receding horizon control for maximizing production in polymerization processes [★]

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Abstract

Constrained nonlinear receding horizon control scheme is proposed to optimize the production in emulsion polymerization processes. The control has the ability to rigorously handle the process constraints (input saturation, maximum allowed heat production, maximal temperature values and rate of change) and can be implemented experimentally due to the low dimensional control parametrization being used. The controller is validated first by simulation to show its efficiency. Then, the controller performance is validated experimentally on a laboratory scale reactor during the polymerization of styrene.

Key words: Polymerization; Nonlinear predictive control; Constrained parameters; Productivity.

1 Introduction

Polymerization processes are not considered as rapid since the sampling time is in general in seconds (1-20 seconds for monomer conversion). However, rapid changes can be observed in these systems, such as particle nucleation, but there are no online sensors for observing these phenomena. Controlling these phenomena is of high importance since these systems are exothermic and irreversible. Moreover, they are sensitive to impurities. One of the main concern of the industrialization of these processes is maximizing the productivity and ensuring its security.

In emulsion polymerization, the reaction rate is related to the reaction temperature and to the concentration of monomer. The concentration of monomer in the polymer particles is proportional to the reaction rate. Controlling this parameter can be done by manipulating the feed rate of monomer. It was usually used to control the

production rate (Buruaga et al. (1997)[2], Zeaiter et al. (2002)[6] and Vicente et al. (2003)[5], Sheibat-Othman et al. (2004)[4]). In all these cases, the reaction temperature was maintained at a fixed value. However, the reaction temperature has a direct influence on the reaction rate. Gentric et al. (1999)[3] used the input/output linearization and optimization of the reactor temperature in order to minimize the process time and to control the polymer molecular weight. For a best control of the process productivity, the reaction temperature profile should be optimized as well as the concentration of monomer in the polymer particles. Araujo and Giudici (2003)[1] used an iterative dynamic programming technique to minimize the reaction time and control the polymer composition by manipulating both the reaction temperature and the monomer flow rate.

In this work, the nonlinear receding horizon control is used to maximize the process productivity by manipulating the monomer flow rate and the reaction temperature. The process constraints, due to physical limitations such as the maximum admissible heat or the maximum possible flow rate are explicitly taken into account in the control scheme. The calorimetry is used to estimate the heat produced by the reaction which allows us to estimate the reaction rate. The number of moles of free monomer in the reactor and the concentration of radicals in the polymer particles are then estimated. Based

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on this information, the controller is applied to control the heat produced by the reaction. The controller is validated experimentally on a 3 liters reactor during the polymerization of styrene.

The paper is organized as follows. First, the system model is described and the control problem is formulated in section 2. The principle of receding horizon control is briefly recalled in section 3 together with the particular formulation adopted in the present paper. In particular, the control parameterization and the definition of a modified cost function that takes into account the state constraints handling are described. Then, some validating simulations are proposed in order to assess the implementability, the efficiency and the robustness properties of the proposed feedback law. Finally, the strategy is validated experimentally during the emulsion polymerization of styrene .

2 Problem formulation

First, the system dynamic model is described, then the control objective and the associated operational constraints are detailed.

2.1 The system model

A simplified model of emulsion polymerization process is given by the following four dimensional ODE's:

$$\dot{N}^T = F \quad (1)$$

$$\dot{N} = F - R_p(N^T, N, T, V) \quad (2)$$

$$\dot{T} = \frac{1}{\rho_m C_p V} \left[(-\Delta H) R_p + UA(T_j - T) + Q_{feed} \right] \quad (3)$$

$$\dot{V} = \frac{MW_m}{\rho_m} F \quad (4)$$

where N^T (*mole*) is the total number of moles of monomer introduced to the reactor. N (*mole*) is the number of residual monomer. F (*mole/s*) is the monomer input flow rate. R_p (*mole/s*) is the rate of transformation of monomer into polymer. T ($^{\circ}$) is the reactor temperature. T_j ($^{\circ}$) is the jacket temperature. V (cm^3) is the total volume of the reaction. The reaction rate $R_p(N^T, N, T, V)$ is a state function that is given by

$$R_p = \mu \cdot V \cdot k_{p0} e^{-EA/(RT)} \cdot M(N^T, N) \quad (5)$$

where μ is the concentration of radicals in the monomer particles and M is the concentration of monomer in the polymer particles. This latter is given by :

$$M(N, N^T) = \begin{cases} \frac{(1-\phi_p^p)\rho_m}{MW_m} & \text{if } \Gamma(N^T, N) \geq 0 \\ \frac{N}{MW_m \left(\frac{N^T - N}{\rho_p} + \frac{N}{\rho_m} \right)} & \text{otherwise} \end{cases} \quad (6)$$

where

$$\Gamma(N^T, N) := \frac{MW_m}{\rho_m} N - \frac{1 - \phi_p^p}{\phi_p^p} \left[\frac{MW_m}{\rho_m} (N^T - N) \right] \geq 0$$

where the following notations have been used

MW_m molecular weight of monomer
 ρ_m, ρ_p monomer and polymer densities
 EA the activation energy of the reaction
 R Universal gas constant
 k_{p0} pre-exponential factor
 ΔH Reaction enthalpy
 U Heat transfer coefficient
 A heat transfer area between the jacket and the reactor
 C_p heat capacity of the reaction medium
 Q_{feed} heat exchanged with the entering components
 ϕ_p^p volume fraction of polymer in the polymer particles

Note that the first two equations (1)-(2) express the mass balance of N^T and N . Equation (3) is the heat transfer balance between the reactor and the jacket while the last equation (4) represents the evolution of the reaction volume due to the flow input.

Note that equations (1)-(4) give the nonlinear state space model of the polymerization reactor with the state being defined as follows

$$x := \left(N^T \ N \ T \ V \right)^T \in \mathbb{R}^4$$

2.2 State measurement

In the remainder of this paper, the state represented above is assumed to be completely measurable by calorimetry and using a balance. The heat produced at some instant t , namely $Q_R(t) = (-\Delta H) \cdot R_p(t)$ is computed using the heat balance and the measurement of the reaction temperature T . This enables $R_p(\cdot)$ to be known and hence used to measure N by integrating equation (2). This scheme leads to measurements that can be acquired once each 10 s .

μ is also a state of the system that is not taken into account in x since it is not modelled here since it is sensitive to impurities and its model is very complicated and infers several other nonmeasurable variables. μ is not measured but can be estimated online from the other measurements. This is to keep in mind in the prediction scheme.

2.3 The operational constraints

Assume that the reaction duration is t_f . In order to properly express the control objective, the operational constraints have to be clearly stated. These are the following :

(1) The maximal admissible flow rate :

$$F(\tau) \in [0, F_{max}] \quad ; \quad \forall \tau \in [0, t_f]$$

(2) The admissible range for the jacket temperature :

$$T_j(\tau) \in [T_j^{min}, T_j^{max}] \quad ; \quad \forall \tau \in [0, t_f]$$

(3) The maximal rate of variation of the control input T_j (due to thermal inertia)

$$|\dot{T}_j| \leq \dot{T}_j^{max}$$

(4) The maximal allowed heat production :

$$Q_R(\tau) := (-\Delta H) \cdot R_p(\tau) \leq Q_R^{max} \quad ; \quad \forall \tau \in [0, t_f]$$

2.4 The control objective

The control objective is to control maximize the quantity of polymer produced during the interval $[0, t_f]$ while respecting the operational constraints (1)-(5) defined in section 2.3. Maximizing the reaction rate implies minimizing the process time for a constant quantity of monomer to consume. This can be formally written as follows

$$\max_{F(\cdot), T_j(\cdot)} [N^T(t_f) - N(t_f)] \quad \text{under (1)-(4) of section 2.3}$$

This objective has to be satisfied by a feedback design in order to be robust against model uncertainties.

3 The proposed control scheme

Since a receding horizon control scheme is adopted in the present paper, let us first of all recall the basic features underlying this principle.

3.1 Control parametrization

The control parametrization used in the present paper for the control inputs T_j and F are depicted on Fig. 1 and Fig. 2 respectively. More precisely, the admissible profiles for T_j over some time interval $[0, t_f]$ are given by the following three dimensional parametrization with the parameter vector given by (w_1, w_2, t_1) :

$$T_j(t) = Sat_{T_j^{min}}^{T_j^{max}} \left(T_j(0) + \int_0^t w(\tau) d\tau \right) \quad (7)$$

$$w(t) := \begin{cases} w_1 & \text{if } t < t_1 \\ w_2 & \text{if } t \in [t_1, t_f] \end{cases} \quad t_1 \in [0, t_f] \quad (8)$$

where w_1 and w_2 are restricted to $[-\dot{T}_j^{max}, \dot{T}_j^{max}]$ in order to meet the constraint (3) of section 2.3 while the

constraint (2) is structurally imposed by the use of the saturation function in equation (7).

The parametrization of the input flow rate F over a

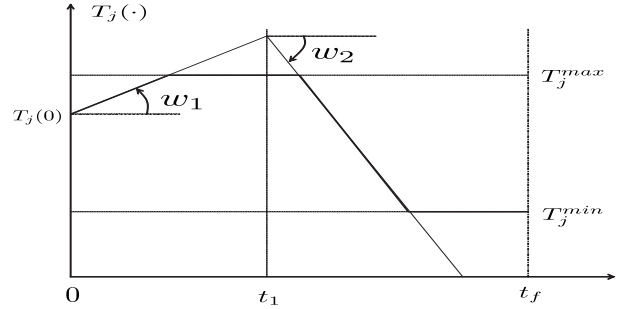


Fig. 1. Parametrization of the control input $T_j(\cdot)$ over a time interval $[0, t_f]$. This parametrization is used in the definition of the receding horizon control formulation. $T_j(\cdot)$ is defined by three parameters: the two slopes w_1 and w_2 that have to belong to the admissible range $[-\dot{T}_j^{max}, \dot{T}_j^{max}]$ and the switching time $t_1 \in [0, t_f]$.

time interval $[0, t_f]$ is given by (see Fig. 2) :

$$F(t) = F(0) + (F^f - F(0))[1 - e^{-\lambda t}] \quad (9)$$

this is a scalar parametrization with the only parameter $F^f \in [0, F_{max}]$ enabling the constraint (1) of section 2.3 to be structurally satisfied.

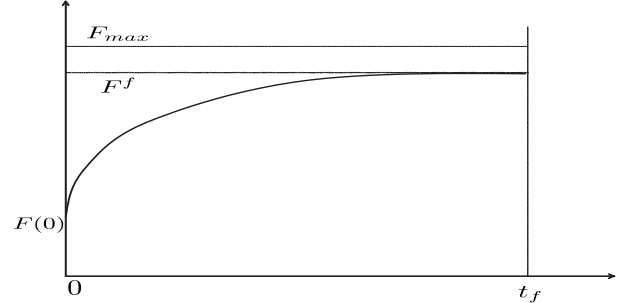


Fig. 2. Parametrization of the control input $F(\cdot)$ over a time interval $[0, t_f]$. This parametrization is used in the definition of the receding horizon control formulation. $F(\cdot)$ is defined by one parameter, namely its asymptotic value F^f .

To summarize, the control input parametrization is defined over a time interval $[0, t_f]$ by the following four dimensional parameter vector

$$p := \begin{pmatrix} w_1 / \dot{T}_j^{max} \\ w_2 / \dot{T}_j^{max} \\ t_1 / t_f \\ F^f / F_{max} \end{pmatrix} \in \mathcal{P} := [-1, 1]^2 \times [0, 1]^2 \quad (10)$$

This control parametrization structurally leads to the satisfaction of the operational constraints (1)-(3) of section 2.3. The satisfaction of the remaining constraint (4) of section 2.3 is guaranteed by a suitable modification of the cost function. This is explained in the following section.

In the remainder of this paper, the control profile $T_j(\cdot)$ and $F(\cdot)$ associated to a particular choice of t_f and $p \in \mathcal{P}$ are denoted as follows

$$T_j(\cdot, p, t_f) \quad ; \quad F(\cdot, p, t_f)$$

3.2 Modified cost function

Assume that the reaction takes place during T_{batch} units of time. Assume some instant $t \in [0, T_{batch}]$. The prediction horizon at that instant is then given by :

$$T_p(t) = T_{batch} - t \quad (11)$$

Each choice of the control parametrization $p \in \mathcal{P}$ results in the following two open-loop control profiles over $[t, T_{batch}]$,

$$T_j(\cdot, p, T_p(t)) \quad ; \quad F(\cdot, p, T_p(t)) \quad (12)$$

The modified cost function at instant t is given by :

$$J(p, t) = \left[1 - \frac{1}{\varepsilon} \max \left(0, \sup_{\tau \in [t, T_{batch}]} \frac{Q_R(\tau, p) - Q_R^{max}}{Q_R^{max}} \right) \right] \times \left[N(T_{batch}, p) - N^T(T_{batch}, p) \right] \quad (13)$$

where $Q_R(\cdot, p)$, $N(\cdot, p)$ and $N^T(\cdot, p)$ are the predicted values given the initial state $x(t)$ and the control profiles (12) that would be used on $[t, T_{batch}]$. $\varepsilon > 0$ is a small parameter.

The relevance of the cost function (13) is shown by the following straightforward result :

Proposition 1 Provided *that there are admissible control profiles over $[t, T_{batch}]$ that lead to the satisfaction of constraint $Q_R(\tau) \leq Q_R^{max}$ over $[t, T_{batch}]$, a parameter vector $\hat{p}(x(t), T_p(t))$ that minimizes $J(p, t)$ leads to state trajectories satisfying*

$$Q_R(\tau) \leq (1 + \varepsilon)Q_R^{max} \quad (14)$$

for all $\tau \in [t, T_{batch}]$

In other words, proposition 1 states that whenever there are solutions that respect the constraint (4) of section 2.3, the modified cost function (13) leads to trajectories

that do not violate this constraint by more than $\varepsilon\%$.

PROOF

This comes from the fact that if p_1 is an admissible parametrization that meets the constraint, then one has

$$J(p_1, t) = N(T_{batch}, p_1) - N^T(T_{batch}, p_1) < 0 \quad (15)$$

(such p_1 exists by assumption). This is because for such p_1 , one clearly has

$$\sup_{\tau \in [t, T_{batch}]} \frac{Q_R(\tau, p_1) - Q_R^{max}}{Q_R^{max}} \leq 0$$

and therefore

$$\max \left(0, \sup_{\tau \in [t, T_{batch}]} \frac{Q_R(\tau, p_1) - Q_R^{max}}{Q_R^{max}} \right) = 0$$

Now let p^* be a solution that violates the constraint by more than $\varepsilon\%$, this clearly means that

$$\sup_{\tau \in [t, T_{batch}]} \frac{Q_R(\tau, p^*) - Q_R^{max}}{Q_R^{max}} > \varepsilon$$

and hence

$$1 - \frac{1}{\varepsilon} \max \left(0, \sup_{\tau \in [t, T_{batch}]} \frac{Q_R(\tau, p^*) - Q_R^{max}}{Q_R^{max}} \right) \leq 0 \quad (16)$$

and since $N(T_{batch}, p^*) - N^T(T_{batch}, p^*) < 0$, equation (16) clearly implies that

$$J(p^*, t) \geq 0 > J(p_1, t)$$

which clearly indicates that p^* is not an optimal solution as soon as it violates the constraint by more than $\varepsilon\%$. This clearly ends the proof. \diamond

3.3 Receding horizon feedback

Let $\tau_s > 0$ be some sampling period for control. The instants $(k\tau_s)_{k \geq 0}$ becomes the decision instants where the optimization problems $P(x(k\tau_s), T_p(k\tau_s))$ are solved in order to update the control inputs to apply to the system on $[k\tau_s, (k+1)\tau_s]$. More precisely, let $\hat{p}(k\tau_s)$ be given by :

$$\hat{p}(k\tau_s) := \text{Arg} \min_{p \in \mathcal{P}} J(p, k\tau_s) \quad (17)$$

the receding horizon control scheme amounts to apply the following control over the sampling period $[k\tau_s, (k+1)\tau_s]$:

$$T_j(k\tau_s + \tau) := T_j(\tau, \hat{p}(k\tau_s), T_p(k\tau_s)) \quad \forall \tau \in [0, \tau_s] \quad (18)$$

$$F(k\tau_s + \tau) := F(\tau, \hat{p}(k\tau_s), T_p(k\tau_s)) \quad \forall \tau \in [0, \tau_s] \quad (19)$$

where $T_p(t)$ is given by (11).

4 Simulation results

Simulation of the closed loop system performance under the proposed constrained receding horizon control are proposed to assess the efficiency and the robustness of the proposed feedback in presence of model uncertainties. The parameters of the polymerization of styrene were used in the simulations.

Very often, model uncertainties are due to the presence of complex phenomena that are difficult to model by explicit mathematical framework. This is the case for instance when the evolution of the concentration of radicals μ is considered. Remember that μ appears in the very definition (5) of the reaction rate R_p .

In order to test the robustness of the controller against high variation of μ with time, the following model is used for $\mu(t)$ during the reaction (note that this model is not known by the controller that only measures the value of μ at each sampling instant) :

$$\mu(t) = \left[1 + \frac{\psi_f \cdot t}{T_{batch}} \right] \cdot \mu_{nom} \quad ; \quad \mu_{nom} := \frac{\bar{n} \cdot N_p}{N_A} \quad (20)$$

Fig. 3 shows the behavior of the closed loop system for $\psi_f = 1$. Note that the controller avoids constraint violation (by more than 5%) despite the fact that μ is multiplied by 2 during the reaction duration.

5 Experimental results

Styrene was used for the experimental validation of the controller. The desired heat produced by the reaction was set to 60W. The maximum number of function evaluations has been taken equal to 150 in this experiment. The used recipe is given in table 1.

	Initial charge (g)	Feed
Styrene	145	1000
Dodecyl sulfate, sodium salt	4	8
Potassium persulfate	4	—
H2O	1000	500

Table 1
Experimental validation of the controller

Fig. 4 shows the evolution of Q_R during two experiments realized with the same recipe. No attempt was done to accelerate the convergence time to the set-point by increasing the reaction temperature during the first 15 minutes in order to anticipate the important heat

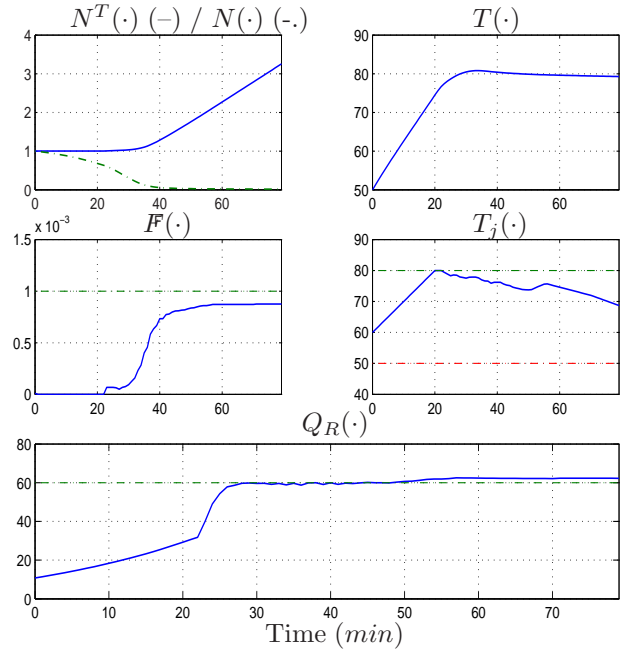


Fig. 3. Behavior of the closed loop system under the receding horizon feedback. $Q_R^{max} = 60$, $T_j^{max} = 80^\circ$, $T_j^{min} = 50^\circ$, $F_{max} = 0.001 \text{ mol/s}$. $\lambda = 1/10s^{-1}$. Sampling period $\tau_s = 1 \text{ min}$. $\varepsilon = 0.05$. Initial conditions $N(0) = N^T(0) = 1$, $T = 50^\circ$, $V(0) = 2$ with the model uncertainty on the concentration of radicals given by (20). Case where $\psi_f = 1$, namely, the value of μ in (5) increases linearly from μ_{nom} to $2 \cdot \mu_{nom}$ during the reaction time interval $[0, T_{batch}]$. The dynamic of μ is unknown to the controller but current values are accessible to measurement according to section 2.2

generated by the nucleation taking place at the beginning of the reaction. The jacket temperature was therefore fixed at $60^\circ C$ during this time (Fig. 5). By the same way, the monomer flow rate (Fig. 6) was fixed at 0 when $\Gamma(N^T, N) = 0$ which means that the polymer particles are saturated with monomer. This corresponds to the first 20 minutes of the reaction. Fig. 6 shows both the desired monomer flow rate calculated by the controller and the real one obtained experimentally while coupled with the local controller. The desired flow rate does not oscillate which means that the nonlinear controller is well tuned and oscillations are due to the local controller tuning. These oscillations affect the heat produced by the reaction as shown on Fig. 4. Q_R presents also some important oscillations that seem to be due to an important unpredicted change in the reaction rate that can be due to a change in mu . This experiment shows that even if the process model seems simple its dynamics can change rapidly in an unpredicted way.

6 Conclusions

Multi-variable constrained receding horizon control scheme has been proposed to maximize the production during polymerization reactions. Two control inputs are used: the inlet flow rate and the jacket tempera-

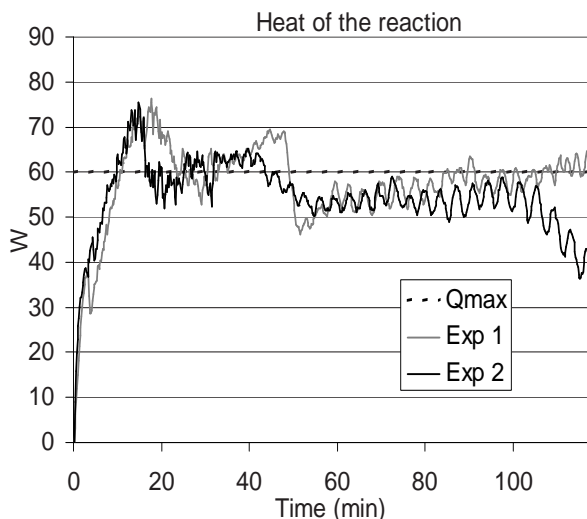


Fig. 4. Experimental validation of the controller. The heat produced by the reaction with a maximal constraint at 60 W

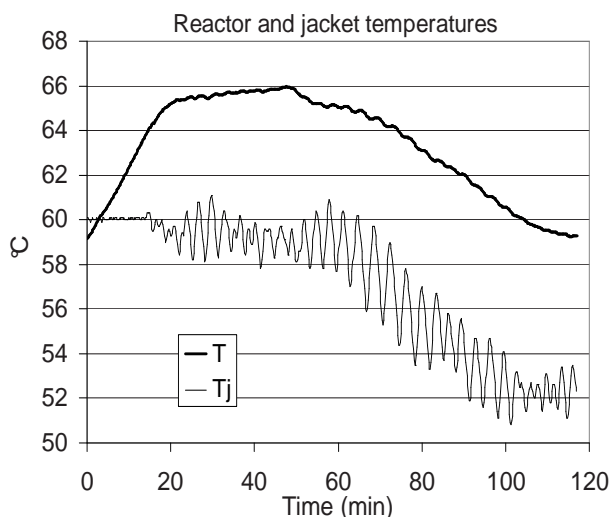


Fig. 5. Experimental validation of the controller. Reactor and jacket temperatures (*Exp2*)

ture. A key feature of the proposed scheme is the very simple control parametrization enabling a real time implementability of the proposed scheme despite the relatively low sampling time (60 s). Another interesting feature is the explicit handling of all operational constraints that would be difficult to be rigorously handled by more classical schemes.

Investigations also showed nice robustness properties against unmodeled dynamics like the one affecting the concentration of radicals in the polymer particles that might increase during the reaction due to the gel effect following very complex and unpredictable laws.

The computation times needed to solve the optimiza-

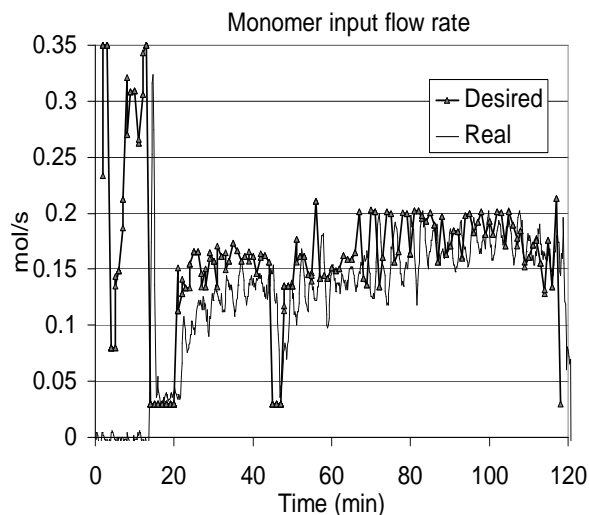


Fig. 6. Experimental validation of the controller. Controlled monomer flow rate and the reaction rate (*Exp2*)

tion problem (less than 0.5s) are even compatible with a higher measurement acquisition frequency (say 0.1 Hz) that is practically feasible. This leads to more reactive controller that would certainly make the results even better than it appears from the simulations proposed in this paper.

The controller was also found to be stable enough while implemented experimentally.

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