

Real-Time Optimizing Nonlinear Control Applied to a Continuous Reactive Distillation Process

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Abstract: Chemical processes are usually operated under feedback control of critical operating conditions to maintain process safety, product quality and economic performance. Increasingly, linear model predictive is applied to these coupled multivariable control problems and in some cases the reference values are adapted infrequently by an optimization based as a rigorous nonlinear stationary plant model (RTO). However, in between these optimizations that plant may operate suboptimally due to the presence of disturbances. In recent years, it has been proposed to use nonlinear model-based optimization of the available degrees of freedom of a process over a finite moving horizon to integrate set-point optimization and feedback control (for a survey see Engell (2007)). In this paper we demonstrate the feasibility and the potential of the optimizing nonlinear control for a challenging example, a reactive distillation column. The performance of the controller is an economics-driven response since the profit function represents the main part of the objective criterion while the quality specifications as well as the physical limitations of the process were incorporated as optimization constraints. A performance comparison between the tracking controller and the optimizing nonlinear controller is also shown.

Keywords: Reactive distillation, Non-linear model predictive control, Non-minimum phase behavior, Tracking and optimizing control, Static gain sign change.

1. INTRODUCTION

Chemical reactions are usually restricted by the chemical equilibrium between reactants and products which limits conversion and selectivity. The combination of reaction and separation into one single unit reduces the investment and the operational costs and the consumption of energy. Reactive distillation (RD) represents one of the most successful applications of the concept of integrated reaction and separation. Using reactive distillation, the conversion can be increased beyond the equilibrium due to the continuous removal of reaction products from the reactive zone according to Le-Châtelier's principle. This also helps in overcoming azeotropes and improving energy integration also. Enhanced reaction selectivity can be one of the advantages of RD columns (Noeres et al. (2003)). In the show-case example of the methyl acetate production process of Eastman, a plant consisting of two reactors and eight distillation columns was replaced by a single reactive distillation column (Agreda, Partin, and Heise (1990)). Disadvantages of reactive distillation are the complex dynamics in the catalytic area that result from the interaction of reaction and separation and the reduced number of degrees of freedom due to the identical temperatures and pressures for reaction and separation.

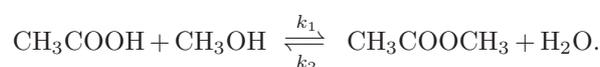
The paper is structured as follows: in section 2 the case study and the rigorous process model are described. A short overview on NMPC, the mathematical formulation

of the different controllers, and a brief illustration of the algorithm are given in section 3. Section 4 shows the performance of the resulting closed loop and section 5 summarizes and concludes this work.

2. CONTINUOUS REACTIVE DISTILLATION

2.1 Process description

The process considered here and shown in Fig. 1 consists of three main parts: the reboiler, the condenser and reflux splitter, and the column itself. The column comprises four sections, in two sections the chemical reaction takes place due to the presence of a catalyst. The system has four input streams which have to be adapted in order to achieve the desired product purity and conversion. These inputs are the reboiler heat duty, the condenser reflux ratio and the flow rates of the two reactants. The exothermic chemical reaction that takes place in the catalytic sections in the middle part of the column is the equilibrium limited heterogeneously catalyzed esterification of acetic acid (HAc) and methanol (MeOH) producing methyl acetate (MeAc) as the desired product and water (H₂O) as a by-product:



The product MeAc is the valuable and most volatile component, while MeOH (key reactant), H₂O and HAc boil at

higher temperatures. The reaction mixture considered here exhibits two minimum azeotropes namely MeAc-MeOH and MeAc-H₂O. At the corresponding temperatures, the vapour phase and the liquid phase have identical compositions so that the product purity that can be achieved in a single distillation column is limited. The presence of the additional substances, such as the acetic acid coming from the top, in the reactive distillation column enables distillation with higher purity in a single column. The vapour stream is fully condensed at the top and split up into two streams in the reflux section. One stream is removed as product (distillate), while the other stream (reflux) and the acetic acid feed realize a counter current flow pattern in which higher-boiling components of the vapour stream get condensed and are taken downwards. In turn, low-boiling components of the liquid stream evaporate and join the vapour stream upwards. The catalyst is a strong acidic ion-exchange resin, type Lewatit K2621 (Noeres et al. (2004)). At the top of each packing, there exists a liquid distributor for dispersing the liquid stream to reduce the wall effects that would decrease the mass transfer between the vapour phase and the liquid phase. The side-product water and un-reacted acetic acid are withdrawn as bottom products.

2.2 Process modelling

Generally, there are two types of dynamic models that have been employed for reactive distillation: the equilibrium model and the rate-based (non-equilibrium) model. For a detailed comparison study between these different models for a packed column we refer to (Peng et al. (2003)).

In this work, the reactive distillation column is described by an equilibrium stage model where it is assumed that the vapour and the liquid stream leaving the stage are in thermodynamic equilibrium with each other. The core of the model are the **MESH** equations, consisting of the material balance equations, the phase equilibrium relations, and the summation equations or the constitutive relations. **H** stands for the heat or the enthalpies of the liquid and/or the vapour phases (Taylor and Krishna (2000)). The equilibrium stage model is extended by some additional equations for the pressure drop of the packing, dynamics of the tray hydraulics etc. The hold-up on each stage is considered to be the hold-up only of the liquid-phase. For additional modelling assumptions and detailed description of the reaction kinetics see (Kreul et al. (1998)).

In this simulation study, each packing section of the reactive distillation process comprises three theoretical trays yielding an overall model with 14 trays including reboiler and condenser trays with total reboiler and total condenser. Applying mass and heat balances around all parts of the plant, a set of differential equations is obtained. A large number of algebraic equations is needed to calculate the physical properties, the phase equilibria and the reaction kinetics as well as the tray hold-ups. Modelling results in a large differential-algebraic system (DAEs), nearly 600 states, in which the dynamics of the process is described by 90 differential states, the rest being algebraic variables.

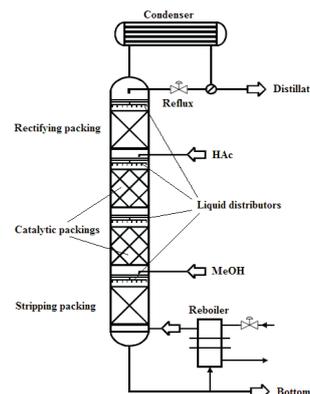


Fig. 1. Reactive distillation column.

2.3 Process characteristics

The model is strongly nonlinear and shows a change of the static gain sign in the channels of reboiler heat supply and condenser reflux (see Fig. 2). The process exhibits non-minimum phase behaviour as illustrated in Fig. 3. Therefore nonlinear model predictive control was proposed for controlling this system. In Fernholz et al. (1999), it was proposed to use a MIMO-control structure for this process with the heat duty supplied to the reboiler and the reflux ratio as the manipulated variables and the concentrations of MeAc and H₂O in the distillate as the controlled variables. In this work, the reactant feed flow rates are also exploited as additional degrees of freedom while the conversion of MeOH is used as a controlled variable instead of the concentration of H₂O in the distillate.

2.4 Model reduction

To simplify the process model used for the online optimization, in the model that is used inside the controller, perfect liquid distribution is considered throughout the column and thus the dynamics of all liquid distributors are ignored. With this assumption about 12% of the states (all of them being algebraic variables) can be removed and the computation time is reduced by about 30%.

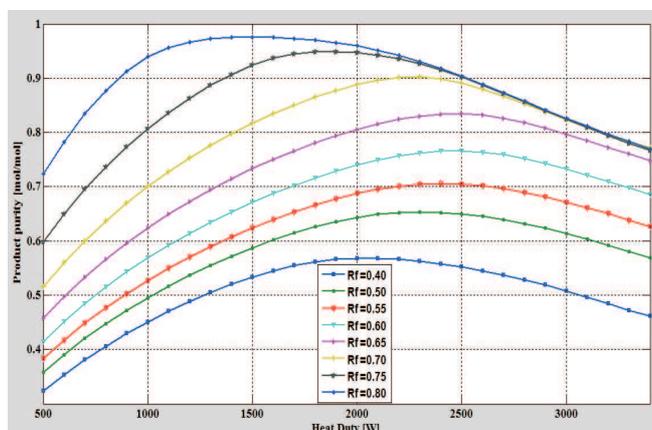


Fig. 2. Input multiplicity in the channel of reboiler heat duty, Rf stands for reflux ratio.

Several simulation studies were performed to compare the performance and the differences between the original model and the model with neglected distributors dynamics. In most cases the differences are small. In some cases differences in the behaviours occur, particularly in the condenser dynamics as can be seen in the example illustrated in Fig. 3. This simulation shows that the composition trajectories of the MeAc purity in the distillate are similar but the simplified model changes the concentrations faster than the original one. Nonetheless the simplified model captures most of the dynamics of the equilibrium-based stage model correctly.

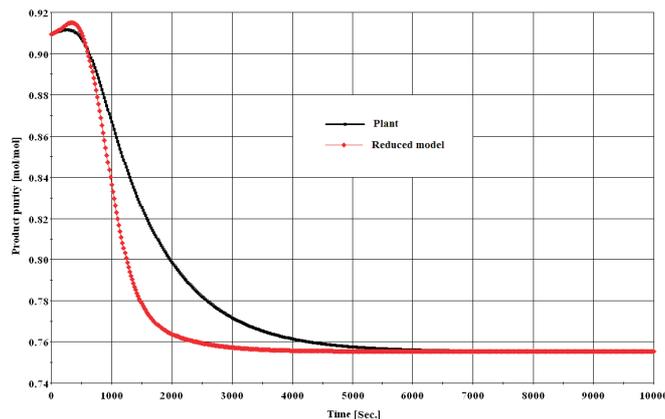


Fig. 3. Comparison of the trajectories of the simplified and the original models for the computation of methyl acetate purity by applying step change in the reflux ratio, also showing the non-minimum phase characteristic of the process.

3. NONLINEAR MODEL PREDICTIVE CONTROL

The principle of NMPC is to choose a set of future control actions or decision variables over the control horizon length of M in order to minimize an objective function and to satisfy all the imposed constraints over a pre-defined prediction horizon P using a nonlinear model. Figure 5 demonstrates the idea behind NMPC. The objective index, which can be a tracking, a maximizing or an optimizing criterion is usually subject to some linear and/or nonlinear constraints besides the dynamic model itself. Although the optimization is performed for a control horizon M , only the first control action is actually implemented. Next, plant output measurements are obtained, and then a compensation of plant-model mismatch is carried out using an open-loop observer as the difference between the plant measured output y^{meas} and the model output y^{model} at the current time interval to adjust the output prediction. Then the optimization problem is solved again.

3.1 Mathematical formulation

Tracking Control The tracking index is the squared sum of the differences between the predicted outputs and the setpoint over the prediction horizon. The slew rate term is added to the objective index to penalize excessive control

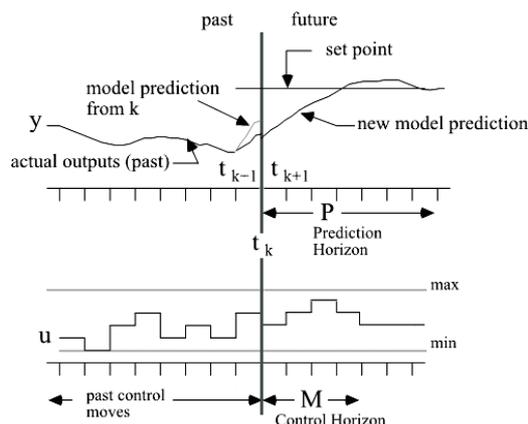


Fig. 4. Concept of NMPC.

actions and to smoothen the response. The mathematical formulation of the tracking controller is as follows:

$$\begin{aligned} & \min_{\Delta u_1(k+1), \dots, \Delta u_1(k+M)} \Phi_T(x, u) \quad (1) \\ & \quad \vdots \\ & \quad \Delta u_k(k+1), \dots, \Delta u_k(k+M) \end{aligned}$$

$$\Phi_T = \sum_{n=1}^N \left(\sum_{i=1}^P \gamma_{n,i} (y_{n,ref}(k+i) - y_n(k+i))^2 \right) + \sum_{b=1}^R \left(\sum_{j=1}^M \alpha_{b,j} \Delta u_b^2(k+j) \right)$$

s.t.

$$\begin{aligned} x(i+1) &= f(x_i, \vartheta_i, u_i, i), \quad i = k, \dots, k+P \\ u_{min} &\leq u(i) \leq u_{max}, \quad i = k, \dots, k+M \\ -\Delta u_{min} &\leq \Delta u(i) \leq \Delta u_{max}, \quad i = k, \dots, k+M \\ u(i) &= u(k+M), \quad \forall i > k+M. \end{aligned} \quad (2)$$

Here x refers to the differential state variables while ϑ denotes algebraic variables. f represents the system dynamics. P is the prediction horizon, M is control horizon. α and γ represent the weights on the control outputs and inputs respectively. y_{ref} refers to the setpoint or the desired output, y_n is the corrected model prediction. N is number of the controlled outputs, R is the number of the control inputs. Compensation for plant-model mismatch is usually done using these equations:

$$\begin{aligned} d_n(k) &= y_n^{meas}(t|k) - y_n^{model}(t|k) \\ y_n(t+k) &= y_n^{model}(t+k) + d_n(k) \end{aligned} \quad (3)$$

where y^{meas} is the output measurement of the plant, y^{model} is the predicted output, d_n denotes the current difference between the model and the plant (Fernholz and Engell (2000)).

Moving Horizon Optimizing Control The replacement of the conventional quadratic cost criterion that penalizes the deviations of the controlled variables from the reference values and the input variations by an economic criterion couples feedback control and the optimal operation of the plant. Product quality specifications as well as process limitations are included in the optimization problem as constraints. This approach has many advantages (Engell (2007)). For a historical view of the changing role of process control in operation and profit/loss measures besides the perspective of how process control has influenced business decision-making see (Edgar (2004)). A general motivation on how model predictive control techniques and dynamic process optimization can be integrated to improve economic performance of chemical processes is given in (Backx et al. (2000)).

A moving horizon optimizing controller that employs the reduced order nonlinear process model of the RD described above is considered here. The production revenues are maximized online over a finite optimizing horizon as in (Toumi and Engell (2004)) for a chromatographic separation process. The product purity and the conversion of the key component methanol are considered as quality constraints. No tracking of previously fixed set-points or reference trajectories is imposed.

The MeAc purity and the conversion of MeOH are averaged over the prediction horizon. The formulation of the economical objective is as follows:

$$\Psi = \left\{ \text{Product revenue} - \text{energy cost} - \sum_{i=1}^{N_f} \text{cost of feed}_i \right\},$$

which can be rewritten mathematically as:

$$\Psi = \int_{t_0}^t \left(\dot{P}(t)C_P - \dot{H}(t)C_E - \sum_{i=1}^{N_f} \dot{R}_i(t)C_{RM,i} \right),$$

where Ψ is the profit, N_f is the number of feed streams, \dot{P} is the product flow rate, C_P is the product unit price, \dot{H} is the boil-up rate, C_E is the price per energy unit, \dot{R}_i are the feed flow rates, $C_{RM,i}$ are the unit price of the feeds. The mathematical setup of the moving horizon optimizing NMPC controller is shown below:

$$\min_{\substack{\Delta u_1(k+1), \dots, \Delta u_1(k+M) \\ \vdots \\ \Delta u_k(k+1), \dots, \Delta u_k(k+M)}} \Phi_{OP}(x, u) \quad (4)$$

$$\Phi_{OP} = \left(\sum_{i=1}^P \gamma_i \Psi_{k+i} \right) + \sum_{b=1}^R \left(\sum_{j=1}^M \alpha_{b,j} \Delta u_b^2(k+j) \right)$$

s.t.

$$x(i+1) = f(x_i, v_i, u_i, i), \quad i = k, \dots, k+P$$

$$u_{min} \leq u(i) \leq u_{max}, \quad i = k, \dots, k+M \quad (5)$$

$$-\Delta u_{min} \leq \Delta u(i) \leq \Delta u_{max}, \quad i = k, \dots, k+M$$

$$u(i) = u(k+M), \quad \forall i > k+M$$

$$\frac{\sum_{i=1}^P \text{Purity}_{MeAc,k+i}}{P} \leq L_{Purity}$$

$$\frac{\sum_{i=1}^P \text{Conv}_{MeOH,k+i}}{P} \leq L_{Conv}$$

where,

$$L_{Purity} = (\text{Purity}_{MeAc,min} - \Delta \text{Purity}_{MeAc,k})$$

$$L_{Conv} = (\text{Conv}_{MeOH,min} - \Delta \text{Conv}_{MeOH,k})$$

$$\Delta \text{Purity} = \text{Purity}_{MeAc,k}^{Plant} - \text{Purity}_{MeAc,k}^{Model}$$

$$\Delta \text{Conv} = \text{Conv}_{MeAc,k}^{Plant} - \text{Conv}_{MeAc,k}^{Model}.$$

Here Ψ_{k+i} represents the profit at the time step $k+i$, $\text{Purity}_{MeAc,k+i}$ denotes the purity of MeAc in the top product stream at the time interval $k+i$. $\text{Conv}_{MeOH,k+i}$ is the MeOH conversion at the time step $k+i$, $\text{Purity}_{MeAc,min}$ is the minimal allowed purity, $\text{Conv}_{MeOH,min}$ is the minimum conversion. P is the prediction horizon length, $u_{k,\dots,k+M}$ are the optimization decision variables. $\Delta \text{Purity}_{MeAc}$ and $\Delta \text{Conv}_{MeOH}$ compensate for model-plant mismatch and potential uncertainties. The degrees of freedom of the optimization problems are the feed flow rates of methanol and acetic acid, the reflux ratio and the reboiler heat duty.

The product purity is defined as the mole fraction of the desired product in the distillate which is the ratio of the number of moles of the product MeAc in the distillate to the total number of moles of all the components in the distillate including the moles of the product itself while the conversion is defined as the ratio of the MeAc product flow in the distillate and the bottom to the methanol feed flow rate. These quantities can be formulated mathematically as:

$$\text{Purity}_{MeAc,k+i} = \frac{m_{MeAc,k+i}}{m_{T,k+i}}$$

$$\text{Conv}_{MeAc,k+i} = \frac{D_{MeOH,k+i} + B_{MeOH,k+i}}{F_{MeOH,k+i}} \quad (6)$$

Where $m_{MeAc,k+i}$ is the number of moles of the MeAc in the distillate at time $k+i$, $m_{T,k+i}$ is the total number of moles in the distillate at time $k+i$. $D_{MeOH,k+i}$ is the number of moles of MeOH in the distillate at the time $k+i$, $B_{MeOH,k+i}$ is the number of moles of MeOH in the bottom at the time $k+i$. $F_{MeOH,k+i}$ is the number of the

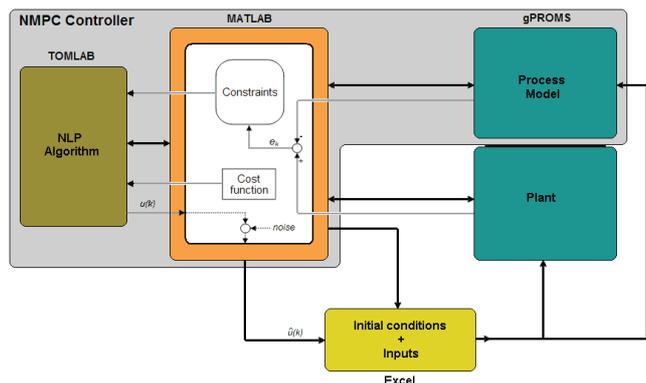


Fig. 5. The designed control algorithm.

MeOH moles in the feed stream at the time $k + i$. The reactant feeds are considered to be pure components.

3.2 Control algorithm

The control algorithm illustrated in Fig. 5 was implemented in gPROMS (see gPROMS User's Guide (2008)), using MATLAB and TOMLAB (see TOMLAB User's Guide (2010)). The Excel spreadsheet server is used to update the initial conditions and the control inputs at each iteration. The differential-algebraic equations of the optimization model and the plant model are integrated using gPROMS' DAE solver, while the optimization is conducted in the TOMLAB optimization environment using the "glcSolve" global NLP solver. The routine glcSolve employs an extended version of the DIRECT algorithm which is a modification of the standard Lipschitzian approach that eliminates the need to specify a Lipschitz constant (Jones, Perttunen and Stuckman (1993)). glcSolve is invoked for a predefined number of function evaluations considering the input values that give the best function value as the optimal solution. Small noise is added to the obtained optimal solution before its implementation at the plant. After the simulation of the plant and the reduced models with the newly obtained optimal inputs, a compensation for the model plant mismatch is done by adding the differences to the purity and the conversion constraints. The initial conditions of the optimization model are updated by the current state of the plant model and then the optimization is performed again.

The optimization solver first searches for a feasible operating point and then minimizes the objective function. The number of function evaluations of the NLP solver was limited to 80 in order to meet the real-time requirements. The objective function and the constraints are implemented in MATLAB which also performs data management and some coordinating tasks such as calling the gPROMS simulation, extracting and exchanging the required data among different modelling platforms as well as updating the initial conditions in the Excel spreadsheet.

3.3 Tuning parameters

The parameters of both controllers are shown in Table 1 below.

Table 1. Shows the parameters of both controllers.

Controller	Tracking and Optimizing
Prediction horizon, P	20
Control horizon, M	1
Regularization, α	[0.002 5.00 · 10 ⁻⁷ 0.45 0.45]
Sampling time, h	4 min.
Plant order	581
Model order	511
Output weights, γ	1

4. CLOSED-LOOP PERFORMANCE AND DISCUSSION

A 2 % stochastic uncertainty is introduced in all of the input channels and there is plant-model mismatch. The performance of the optimizing and the tracking controllers is depicted in Fig. 6. Both controllers were able to react in an effective way.

The optimizing controller manages to track the purity reference and to keep the conversion above its lower limit, while maintaining the economical operation of the plant by maximizing the revenue. Comparing the performance of both controllers, the optimizing controller takes care of both the optimal economics and prevents off-specifications product in spite of the structural and input uncertainties which may lead to a slower regulation. Fig. 7 compares the accumulated profit of the optimizing and the tracking controllers. An increase of about 29% in the profit with respect to the tracking controller after the change of the setpoint is obtained. The prices of the reactant, the energy, and the product considered in this paper are depicted in Table 2.

The simulation of the plant model for one sampling time (4 minutes) takes around 3 seconds, while the simulation of the model took around 2 seconds for a simulated time of 1.67 hours. One call of the NMPC controllers takes about 3.8 minutes.

Table 2. Shows the prices of reactant, product and energy.

Materials and energy	Unit price, €
MeAc, Mole	0.36
HAc, Mole	0.173
MeOH, Mole	0.005
Heat duty, kW	4.25e-6

5. CONCLUSIONS

By optimizing control, an improvement in the plant economics can be achieved. The optimizing controller has been applied successfully to a simulated complex model of a reactive distillation column with plant model mismatch. The controller manages to track the purity constraint and to keep the conversion above its lower limits, while it improves the economics by maximizing the revenue of the plant. More sophisticated numerical algorithms for simulation and optimization are still needed to enable real-time dynamic optimizing control for industrial-scale plants.

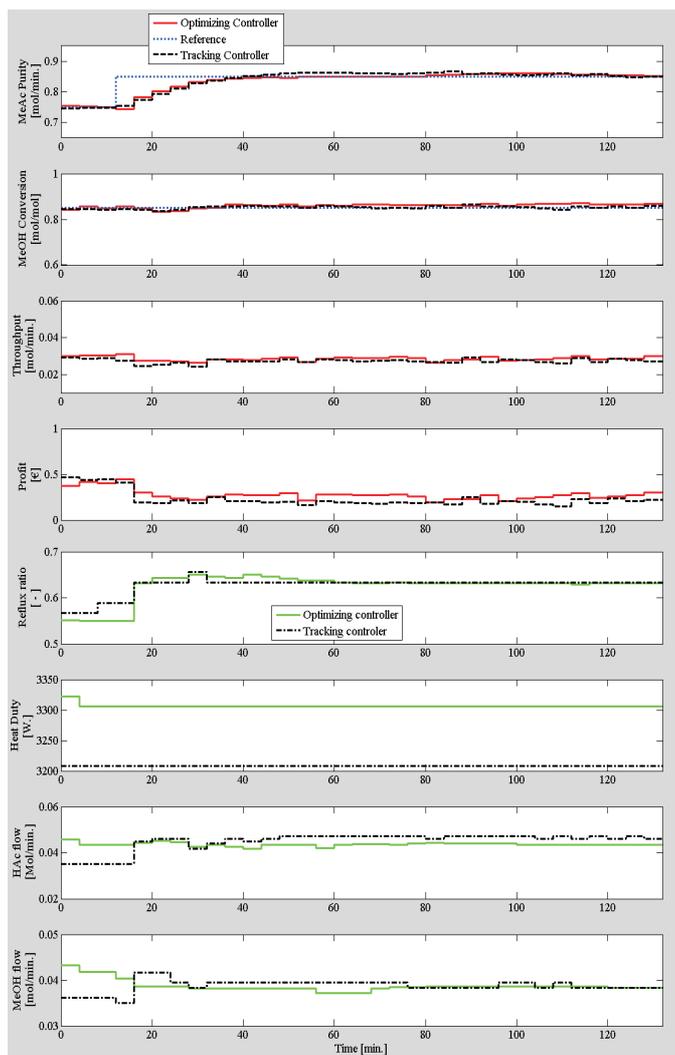


Fig. 6. Performance of the optimizing and tracking NMPC controllers under structural uncertainties and stochastic disturbances that act in all input channels.

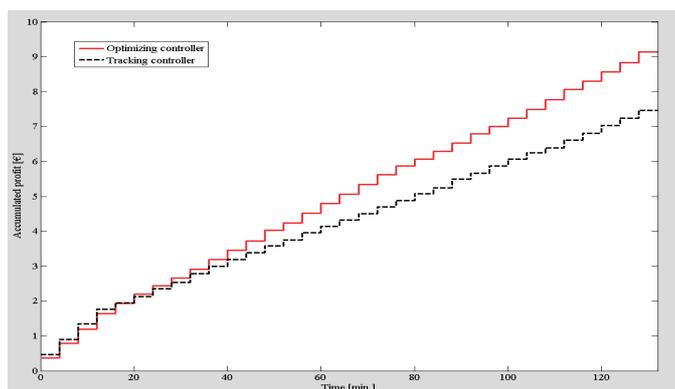


Fig. 7. Comparison of the accumulated profit of the optimizing and the tracking controllers.

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