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Conceptual modelling and optimization of jacketed tubular reactors for the production of LDPE

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Abstract: This paper deals with the model based optimization of tubular reactors for the production of LDPE. Due to the high complexity, solving an optimization problem of an industrial application is not straightforward. Often researchers seek the shortest way to reach their final goal by going directly to the development of a high-complexity model and optimize this with respect to a certain objective. Although this approach seems the fastest way to success, it can be a bumpy road with a lot of dead ends. Therefore, a *divide and conquer* strategy is adopted, i.e., first develop a conceptual low-complexity model, set up the optimization problem and then use the obtained knowledge during the optimization of more complex models. The aim of this paper is to give the results of the three steps which have to be accomplished in order to achieve this subgoal. First, the multizone process of LDPE production is modelled as a sequence of conceptual modules which simulate the steady-state characteristics of one reaction and cooling zone. Then, this model is fitted to industrial data such that it quantitatively describes the real process. Finally, a multiple objective design optimization problem is formulated, i.e., where along the reactor and which amount of initiator has to be injected to maximize the profit at different economic situations.

Keywords: Chemical industry; multiobjective optimizations; process parameter estimation; plastics industry; multiple-criterion optimization; differential equations.

1. INTRODUCTION

Polyethylene, which includes HDPE, LDPE and LLDPE, is without a doubt the most widespread polymer worldwide. Its annual production is estimated at 80 million tonnes. Low density polyethylene (LDPE), made in high-pressure reactors, represents about 30 % of the total volume of produced polyethylene and is used for a large number of applications, e.g., packaging, adhesives, insulators, coatings and films. This widespread use is the result of the wide range of possible molecular and structural properties of the various grades of LDPE and its copolymers.

Low density polyethylene is commonly produced in high-pressure autoclaves and tubular reactors. From the 90's on, when the majority of LDPE (60 %) was produced in the autoclave reactors, the tubular reactors gradually got the upper hand. Nowadays the tubular reactor stands for 60 % of the total production of LDPE. A high-pressure LDPE tubular reactor consists of a spiral wrapped metallic pipe with a large length to diameter ratio and a total length ranging from 1500 to 3000 m. The (co)polymerization of ethylene is carried out under extreme conditions, e.g., at very high pressures in between 2000 and 3000 bar. Because of the high pressure, the thickness of the reactor wall is of the same order as the inner diameter of the tube. Also the

reactor works at a high temperature level of 400 to 600 K due to the exothermicity of the free-radical polymerization reaction. The heat of reaction is removed through the reactor wall by a cooling system in the jacket around the tube. The ethylene conversion in this process is known to be very low, in the order of 25-35 %, and the polymer produced in these tubular reactors has a typical density in the range of 915-930 kg/m^3. Downstream of the tubular reactor, the unreacted monomer and other species have to be removed from the produced polymer. A commercial reactor can have multiple reaction and cooling zones and includes a number of initiators, monomers and solvents feeding points.

In literature substantial research has been conducted on the modelling of the LDPE tubular reactor. Several studies have developed models of different complexity for *(i)* steady-state simulation [Kiparissides, 1996, Brandolin et al., 1996, Agrawal and Han, 1975, Bokis et al., 2002] and optimization [Yoon and Rhee, 1985, Brandolin et al., 1991, Yao et al., 2004] and *(ii)* transient simulation [Häfele et al., 2005, 2006] and optimization [Asteasuain et al., 2001]. The numerous examples prove that the use of the model based optimization approach for industrial applications leads to significant improvements and benefits for the manufacturers. A key component in this approach is to find the
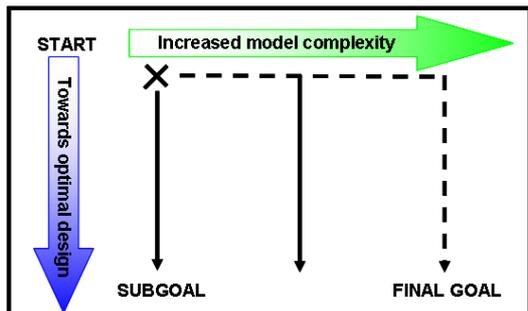
Fig. 1. *Divide and conquer* strategy for model based optimization of LDPE tubular reactors.

balance between *(i)* having an accurate predictive model of the process and *(ii)* minimizing the computational effort (calculation time) of the optimization problem. Both depend on model complexity. Where to draw this gray line depends on the application and the research objective. Every made assumption or simplification excludes some non-idealities from the reactor model, e.g., Agrawal and Han [1975], Yoon and Rhee [1985] assume a constant heat transfer coefficient due to no fouling in the reactor, Yao et al. [2004] neglect the pressure drop and Yoon and Rhee [1985], Zhou et al. [2001] simplify the complex LDPE polymerization reaction mechanism.

Despite some assumptions researchers often go straight to the use of high-complexity models to tackle the optimization problems of the LDPE reactor. It is well know that high complexity non-linear optimization problems are difficult to solve and for example a well chosen initialization has a big influence on the convergence of the optimization. So why not start with conceptual (low-complexity) modelling and gradually increase the complexity of the model (Fig. 1)? Using conceptual models for simulation and optimization gives the researcher generic insight on the industrial application and the obtained profiles are useful as initialization for the more complex models.

In this paper the first part of the *divide and conquer* strategy, i.e., the conceptual modelling and optimization of LDPE tubular reactors is discussed in detail (subgoal in Fig. 1). The paper is organized as follows. Section 2 gives a global overview of the chosen conceptual approach, which consists of three major steps. In the following sections each step, i.e., *(i)* conceptual modelling of a tubular LDPE reactor (Section 3), *(ii)* estimation of the model parameters on industrial data (Section 4) and *(iii)* formulating and solving a multi-objective design optimization (Section 5) is elaborated in detail and the obtained results are discussed. Finally, Section 6 summarises the main conclusions.

2. APPROACH

Setting up a well-defined model based optimization problem of an industrial application is not always as straightforward as it seems. In literature, researchers choose the shortest way to success by directly developing a high-complexity model and use the model to optimize with respect to a certain objective function (dashed arrow in Fig. 1). Due to the high complexity of the model conver-

gence is not ensured, good initialization is required and calculation time increases rapidly. Hence, for this research project a sustainable strategy of *divide and conquer* is chosen, i.e., first develop a conceptual (low-complexity) model, set up the (multiple objective) optimization problem and then use the obtained knowledge during the optimization of more complex models. This paper focusses on the first part, i.e., the conceptual optimization of jacketed tubular reactors for the production of LDPE (arrow in Fig. 1). To obtain this subgoal three steps has to been followed.

Step 1: Process Modelling. LDPE is produced in a high-pressure tubular reactor that exists of multiple reaction zones (Fig. 2) with its characteristic multi-peak temperature and conversion profiles along the tube. Because the reaction mechanism stays the same in each zone, every reaction module exhibits similar shaped profiles. The length and distribution of the reaction zones or peaks along the tube are mainly defined by the initiator, i.e., the location of initiator injector, the amount and composition of the initiator cocktail. So the multizone tubular reactor is modelled as a series of 1-peak reactor modules, where each module represents the model of one reaction zone.

Step 2: Parameter estimation. After developing the model, which is capable of qualitatively describing the characteristic features of the LDPE production process, the unknown model parameters have to be estimated in order to simulate the actual measured industrial data. First, the parameters of a 1-peak reactor module are estimated on the data of each reaction zone separately, after which the 4-peak reactor model can be fit on the data of the entire reactor. By following this parameter estimation strategy, *(i)* it is known in advance whether the 1-peak reactor module can quantitatively describe each reaction zone, *(ii)* you get an idea which model parameters can be kept constant along the multizone reactor and *(iii)* good initial guesses for the 4-peak reactor can be made.

Step 3: Multiple objective optimization problem formulation (MOO). In this conceptual study about tubular reactors in steady-state, and also in future work, the focus is put on formulating and solving optimization problems with multiple and conflicting objective functions (costs). In contrast to single objective optimization, no unique solution exists in MOO and it is necessary to determine a set of points that all fit a predetermined optimality definition, i.e., Pareto optimality. A solution is Pareto optimal if there exists no other solution that improves at least one objective function without worsening another.

In general a multiple objective optimal control problem can be formulated as follows:

$$\min_{u(\xi),\ \xi \in [0,\xi_f],\ \xi_f} \mathbf{J} = (J_1, \ldots, J_m)^{\mathrm{T}} \qquad (1)$$

where each individual objective function J_i can be of the following type:

$$J_i = \underbrace{h_i[\mathbf{x}(\xi_f), \mathbf{p}]}_{\text{Terminal cost}} + \underbrace{\int_0^{\xi_f} g_i[\mathbf{x}(\xi), u(\xi), \mathbf{p}]d\xi}_{\text{Integral cost}} \qquad (2)$$

with $\mathbf{x}$ and $\mathbf{u}$ the state and control variables.
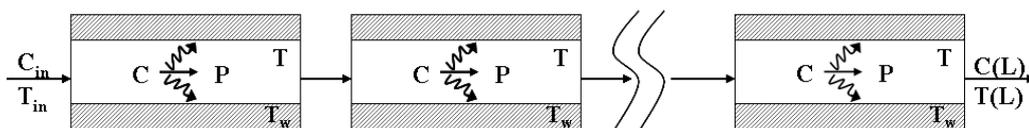
Fig. 2. Schematic view of a high pressure multizone jacketed tubular reactor for the production of LDPE.

subject to:

$$\frac{d\mathbf{x}}{d\xi} = \mathbf{f}[\mathbf{x}(\xi), u(\xi), \xi] \quad (3)$$

$$\mathbf{0} = \mathbf{I}[\mathbf{x}(0)] \quad (4)$$

$$\mathbf{0} = \mathbf{S}[\mathbf{x}(\xi_f)] \quad (5)$$

$$\mathbf{0} \geq \mathbf{C}(\mathbf{x}(\xi), u(\xi), \xi) \quad (6)$$

The vector $\mathbf{f}$ represents the dynamic system equations on the interval $[0, \xi_f]$, in this case the model equations developed in step 1, with consistent initial and terminal boundary conditions given by the vectors $\mathbf{I}$ and $\mathbf{S}$. The vector $\mathbf{C}$ indicates path and/or terminal inequality constraints on the states and the controls.

A deterministic multiple shooting method is employed to tackle the optimal control problems and to generate the resulting Pareto front.

3. MODELLING

The LDPE production process is carried out in a multizone jacketed tubular reactor with the main monomer feed at the inlet and where the initiators, most often mixtures of peroxides, are fed to the reactor through lateral injections. A peak, following the initiator injections, consists out of a short reaction zones with a high heat generation and a cooling zone where the reactor is mainly used as a heat exchanger, in order to reach appropriate temperatures for the next initiator addition. In other words, the model for the complete tubular reactor (Fig. 2) is built up as a series of modules, which describe the working principle of one peak. A 1-peak module is composed of the mass balance of the monomer and initiator and the energy balance along the length of the reactor.

3.1 One-peak module

Every 1-peak module represents a classic tubular reactor with one initiator injection point at the inlet and with a surrounding cooling jacket present to remove the heat of reaction. In the tubular reactor the supercritical single-phase ethylene-polyethylene mixture allows the reaction to take place as a free-radical initiated polymerization. Instead of incorporating the full complex polymerization mechanism existing of several initiation, propagation and termination reactions, the model describes the polymerization as a single exothermic, irreversible, first-order reaction of the monomer M and an initiator radical I^*. Assuming that κ is the average chain length of the produced polymer P, the simplified reaction looks as follows:

$$\kappa M + I^* \xrightarrow{k_0} P.$$

When assuming (i) there is no axial dispersion, (ii) steady-state conditions, and (iii) an Arrhenius law dependence of the reaction rate on the temperature, the reactor is described by the following first-order differential equations with respect to the spatial coordinate z [m]:

$$\frac{dx_1}{dz} = \frac{k_0 C_{i,in}}{v}(1-x_1)(1-x_3)e^{\frac{-\gamma}{1+x_2}} \quad (7)$$

$$\frac{dx_2}{dz} = \frac{k_0 C_{i,in}\delta}{v}(1-x_1)(1-x_3)e^{\frac{-\gamma}{1+x_2}} + \frac{\beta}{v}(u-x_2) \quad (8)$$

$$\frac{dx_3}{dz} = \frac{k_0\epsilon}{v}(1-x_1)(1-x_3)e^{\frac{-\gamma}{1+x_2}} \quad (9)$$

subject to the initial conditions:

$$x_1(0) = 0 \quad (10)$$

$$x_2(0) = 0 \quad (11)$$

$$x_3(0) = 0 \quad (12)$$

with $x_1 = (C_{in} - C)/C_{in}$, the dimensionless monomer concentration C [mole $\cdot$ L^{-1}], $x_2 = (T - T_{in})/T_{in}$, the dimensionless reactor temperature T [K], $x_3 = (C_{i,in} - C_i)/C_{i,in}$, the dimensionless initiator concentration C_i [mole $\cdot$ L^{-1}], and $u = (T_w - T_{in})/T_{in}$, the dimensionless jacket temperature T_w [K], respectively. v [m/s] represents the fluid superficial velocity, while k_0 [s^{-1}] is the kinetic constant. The constants δ, γ, β and ϵ are defined as follows:

$$\delta = -\frac{\Delta H}{\rho c_p}\frac{C_{m,in}}{T_{in}} \qquad \gamma = \frac{E}{RT_{in}}$$

$$\beta = \frac{4h}{\rho c_p d} \qquad \epsilon = \frac{C_{m,in}}{\kappa}$$

with ΔH [J $\cdot$ kmole^{-1}] the heat of reaction ($\Delta H < 0$ for an exothermic reaction), and ρ [kg $\cdot$ m^{-3}], C_p [J $\cdot$ kg^{-1} $\cdot$ K^{-1}], E [J $\cdot$ mole^{-1}], R [J $\cdot$ mole^{-1} $\cdot$ K^{-1}], h [W $\cdot$ m^{-2} $\cdot$ K^{-1}], and d [m], the fluid density, the specific heat, the kinetic constant, the activation energy, the ideal gas constant, the heat transfer coefficient, and the reactor diameter, respectively. κ [-] represents the average chain length of the produced polymer.

3.2 Multi-peak model

The multi-peak model is nothing more than a sequence of the 1-peak modules, with different model parameters fitted on industrial data (obtained in Section 4). Every time a 1-peak module is added to the model an extra state variable is defined, i.e., an extra dimensionless initiator concentration because at the beginning of each peak a different type of initiator can be injected. It also makes the model more robust in case a mixture is added containing previous used initiators.

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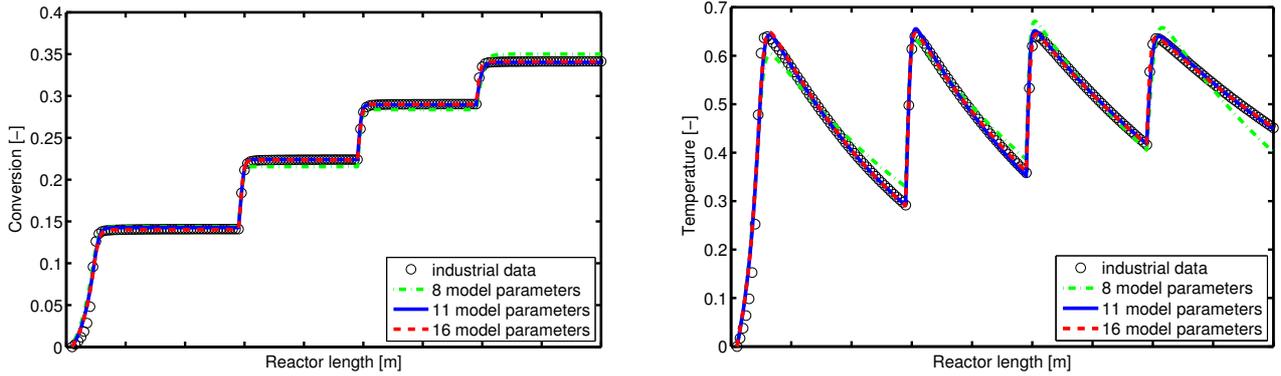
Fig. 3. Parameter estimation of three case studies on the industrial data of 4-peak reactor: conversion (left) and temperature (right).

4. PARAMETER ESTIMATION

Simulations proved that the characteristic temperature and conversion profiles can qualitatively be described by the model obtained in Section 3, but is it also capable of reproducing the industrial data? In Section 4.1 the mathematical formulation is given, followed by the parameter estimation on each peak separately in Section 4.2. Finally, the 4-peak model is fitted to a complete industrial data set in Section 4.3.

4.1 Mathematical formulation

In general, a parameter estimation can be formulated as a dynamic optimization problem with the model parameters $\mathbf{p}$ as decision variables and the mean squared error (MSE) as objective function J:

$$\min_{\mathbf{p}} J = \frac{\sum_{j=1}^{J_e} \sum_{m=1}^{2} \left[(x_m(z_j, \hat{u}(z), \mathbf{p}) - \hat{x}_m(z_j))^2 \right]}{n_D - n_p} \quad (13)$$

with x_m the modelled m^{th} state variable of equations (7) and (8), i.e., the dimensionless monomer concentration and the dimensionless temperature. $\hat{x}_m$ and $\hat{u}$ are the industrial measured m^{th} state variable, respectively control variable or dimensionless jacket temperature, in the number of measure points J_e. n_D and n_p are the number of industrial data points and the number of unknown model parameters.

4.2 One-peak model

The one-peak model is composed of the mass balance of the monomer and initiator and the energy balance along the length of the reactor (equations (7)-(9)). The model has four known or measurable parameters, i.e., fluid superficial velocity v, kinetic constant k_0, initiator inlet concentration $C_{i,in}$ and reactor length L. The model also contains four dimensionless model parameters to be estimated, i.e., δ, γ, β and ϵ.

The parameter estimation of each of the peaks separately produces useful information for the later multiple peak estimation. Most important, it proved that the conceptual, low-complexity model is capable of quantitatively describing each reaction zone of the real-life tubular reactor. Only in the reaction zone of the first peak some inaccuracies between the s-shape of the data and of the simulated

model are noticeable due to nature of the initiator at the inlet. Instead of assuming only one type of initiator, industry uses a mixture of initiators, which are active at different temperatures. The use of a mixture gives rise to the more gradual increase of the reactor temperature than is obtained from the model. Secondly, the results give some insight on which model parameters of the multi-peak model can be kept constant along the reactor. The parameter β, which determines the heat transfer through the jacket, differs from peak to peak because towards the outlet the increase in polymers to monomer ratio leads to a decrease in heat exchange. It also causes an increase of fouling at the reactor wall, which gives rise to an additional heat transfer resistance. Due to the use of different types of initiator at each injection point, the ratio of monomer vs initiator consumption, i.e., parameter ϵ, varies in each reaction zone. The parameter γ, which has an influence on the rate of reaction, of the first peak differs from the second peak, but remains constant for the following peaks, while δ, a measure for the heat of reaction, is constant along the complete reactor. Finally, the obtained results provide useful initial parameter values and boundaries for the multi-peak estimation. The parameter values however, cannot be disclosed due to confidentiality reasons.

4.3 multi-peak model

To emphasize the importance of the previous section, the 4-peak parameter estimation is performed for three different sets of model parameters (Table 1), i.e., a set (i) based on the profile of the data points, (ii) based on the a priori knowledge from Section 4.2 and (iii) with all available model parameters.

Table 1. Overview 4-peak parameter estimation

Case	n_p	Free model parameters	MSE
i	8	β_{1234}, δ_{1234} γ_1, γ_{234}, ϵ_1, ϵ_2, ϵ_3, ϵ_4	1.2×10^{-4}
ii	11	β_1, β_2, β_3, β_4, δ_{1234} γ_1, γ_{234}, ϵ_1, ϵ_2, ϵ_3, ϵ_4	3.3×10^{-5}
iii	16	β_1, β_2, β_3, β_4, δ_1, δ_2, δ_3, δ_4 γ_1, γ_2, γ_3, γ_4, ϵ_1, ϵ_2, ϵ_3, ϵ_4	2.8×10^{-5}

As depicted in Fig. 4 the first case, with 8 parameters and the heat transfer assumed constant, is not able to describe the industrial data. The model cannot fit the temperature profiles of cooling zones and simulates incorrect conversion

values at the end of each peak. Thus by not using the knowledge gathered in the previous section, not only incorrect cooling profiles were obtained, but this also resulted into incorrect ϵ-values. Both the second and third case, with 11 and 16 parameters respectively, describe accurately the industrial data (except for the temperature in the first reaction zone, as predicted in Section 4.2). Based on the MSE values the third case still has a minor advantage on the second case, but note that 50 % more unknown parameters have to be estimated. Therefore, in line with the conceptual approach of this research the model with 11 parameters is chosen.

5. MULTIPLE OBJECTIVE OPTIMIZATION

First, in Section 5.1 the MOO, based on equation (1) to (6), is formulated. Then, in Section 5.2 an overview of the optimization results for the 4-peak LDPE reactor is given, followed in Section 5.3 by the results for a range of multizone reactors going from 1-peak till 6-peak reactors.

5.1 Mathematical formulation

Model. The model developed in Section 3.2 with the obtained model parameters from Section 4.3 is used. The control variable, the jacket temperature, is assumed to be constant along the reactor.

Constraints. The inlet and outlet temperatures are fixed due to the required conditions of the up- and down stream processes. To ensure a safe process operation the reactor temperature is bounded along the reactor. At the end of the reactor all initiator has to be converted in order to avoid further polymerization in downstream processes. Due to practical reasons the total reactor length is limited to 15000 meters, which is still longer than for real LDPE reactors.

Objectives. The goal is on the one hand to maximize the return or in other words to maximize the conversion x_1 at the end of the reactor $J_{\text{conversion}}$. On the other hand the investment cost J_{invest} has to remain minimal. This can for example be quantified by minimizing the reactor length L. The multiple conflicting objectives are reformulated to a single objective J by a convex weighted sum:

$$J = -(1 - A) \times J_{\text{conversion}} + \frac{A}{K} \times J_{\text{invest}} \qquad (14)$$

with trade-off coefficient $A \in [0, 1]$ and a scaling factor K. For each trade-off value A, the solution is known to be Pareto optimal. The trade-off value can easily be linked to real a economic meaning. For instance, when $K = \frac{p_{\text{conversion,ref}}}{p_{\text{invest,ref}}}$ (conversion/meter tube) is the ratio of the reference conversion price $p_{\text{conversion,ref}}$ (money/conversion) to the reference investment price of 1 meter tube $p_{\text{invest,ref}}$ (money/meter tube), the reference situation is optimized for $A = 0.5$. However, as the economic situation changes, deviations can be accounted for by selecting $A = \frac{K}{K_a + K}$ with $K_a = \frac{p_{\text{conversion}}}{p_{\text{invest}}}$ (conversion/meter tube) the ratio of the actual conversion and investment prices. Varying A from 0 to 1 not only covers the entire economic spectrum, i.e., from freely available reactor tubes to worthless reaction products, but also yields an approximation of the Pareto front.

5.2 4-peak multiple objective optimization

The degrees of freedom are *(i)* the inlet concentrations of the initiator at each of the four injection points as *(ii)* the length of each peak, i.e., the placement of the initiator injectors along the reactor.

The resulting Pareto front of the 4-peak reactor and the corresponding optimal temperature and conversion profiles are depicted in Figure 4. For A-values lower than 0.3, i.e., where the produced LDPE is worth a lot more than the needed investment cost, identical optimal designs of a 4-peak reactor are found. If the design is not limited by the investment cost (reactor length), maximal conversion of a multipeak reactor is obtained by pushing every peak to the temperature bounds, i.e. a maximum concentration of initiator is injected such that the reactor temperature just remains beneath the upper bound and the length of a peak is maximized in order to cool down until the mixture reaches the jacket temperature. For increasing A-values, the investment cost becomes more important and the corresponding peak lengths decrease. By shortening the cooling zone, the reactor temperature is higher at the next injection point, hence, a smaller amount of initiator can be injected in order to avoid reactor temperature violation and the conversion decreases. Around $A = 0.6$ the four peaks are reduced to three peaks because the length between two injections becomes to small to sufficiently cool the mixture. As the A-value reaches 0.9, the reactor is reduced to one peak. Further increase of the A-value leads to useless designs, i.e., the reactor is too expensive to build.
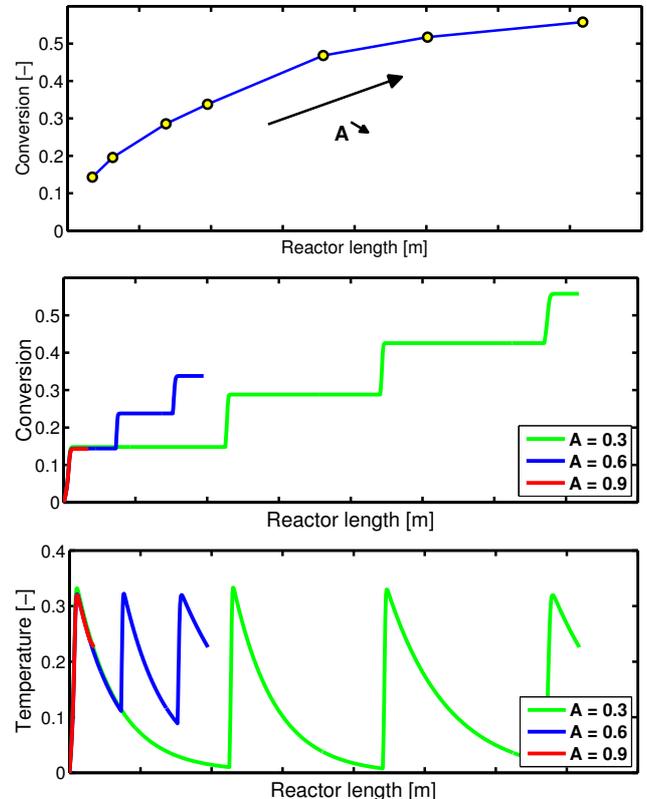
Fig. 4. 4-peak reactor: Pareto front (top) with corresponding optimal conversion (middle) and temperature profiles (bottom).

5.3 Multi-peak multiple objective optimization
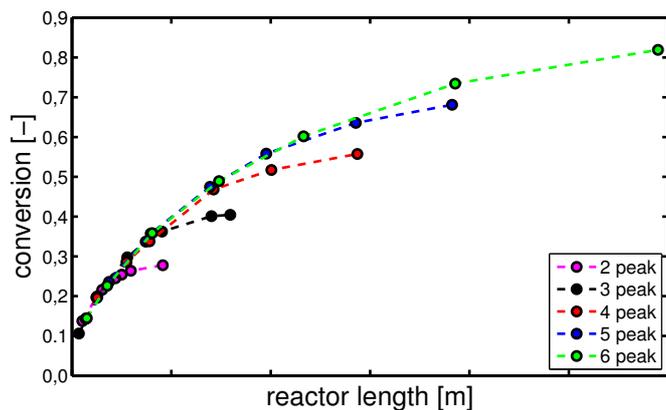
Fig. 5. Comparison of the Pareto fronts of the multi-peak reactors up to six injections points.

In Section 5.2 the Pareto front is obtained for a reactor with at most four initiator injections. To get a more global overview, the optimization is performed with all multi-peak reactor models up to six injections. The resulting Pareto fronts are depicted in Fig. 5. Despite the use of different multi-peak models, the results of the optimization for high A-values are practically the same, i.e., although multiple peaks are possible, the length of all peaks but one is set to zero due to the high investment cost of the tubes. As the A-value decreases, the produced LDPE becomes more valuable and the advantage of additional peaks towards higher conversion becomes clear. It is easy to deduct from Fig. 5 which multi-peak reactor is preferred at which market situation and what the additional profit is of adding an extra peak. Although no values are given due to confidentiality reasons, it should be noted that for the current market situation the resulting optimized reactor design based on the conceptual model does not differ much from the real-life reactor.

6. CONCLUSION

In this paper the first part of the *divide and conquer* strategy, i.e., the conceptual modelling and optimization of LDPE tubular reactors, is described in order to obtain some generic insight on the application and to use the profiles as good initialization for the optimization of the more complex models. First, the LDPE multizone high-pressure tubular reactor is modelled as a series of steady-state 1-peak modules. A 1-peak module is composed of the mass balance of the monomer and initiator and the energy balance along the length of the reactor. Simulations of this conceptual model proved that it was capable of qualitatively describing the typical profiles of the peaks. In a second step the model parameters are estimated on real industrial data. Despite the conceptual approach, the simulation results of the multi-peak model with obtained parameter values fit almost accurately the data. Only in the beginning of the first peak a deviation between simulation and data is detected, but this is due to the assumption of only injecting one initiator instead of a mixture. Finally, with the obtained model a multi-objective optimization problem is formulated in order to maximize

return, i.e., maximize the produced LDPE (conversion) while minimizing the investment cost. The two conflicting costs are reformulated to a single objective by a weighted sum, where trade-off coefficient A can be linked to the market situation. For high A-values the LDPE is worthless compared to the investment and small reactor with only one peak is designed. For decreasing A-values, the market price of LDPE increases and the optimizer automatically adds extra peaks in order to increase the conversion. The Pareto fronts obtained for the multipeak models up to six peaks already helps the designer in comparing the (dis)advantages of adding an extra injector versus varying the initiator amount and prolonging the peak lengths.

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