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# Modeling and Optimization of a Grade Change for Multistage Polyethylene Reactors

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**Abstract:** Grade changes in polyethylene reactors, i.e., changes of operating conditions, are performed on a regular basis to adapt to market demands. In this paper, a dynamic optimization procedure is presented built upon the Modelica language extended with Optimica constructs for formulation of optimization problems. A Modelica library for the Borstar<sup>®</sup> multistage polyethylene reactors at Borealis AB, consisting of two slurry and one gas phase reactor, has been constructed. Using JModelica.org, a framework to translate dynamic optimization problems to NLP problems, optimal grade transitions between grades currently used at Borealis AB, can be calculated. Optimal inflows and grade key variables are shown.

**Keywords:** optimization, reactor control, polymerization, chemical industry

## 1. INTRODUCTION

Polyethylene reactors are able to produce different grades by manipulating inflows of raw material. It is imperative for polyethylene manufacturers to change product grades to increase profitability as market demands change, but also due to market competition and raw material pricing. The result is product campaigns, varying in length between a few days up to weeks. During grade transitions it is therefore of importance that production of off-specification material, i.e., material that does not fulfill specification of any grade, is minimized. On the other hand, there is also a cost in raw material and time that has to be taken into account when performing a grade change, see e.g., van Brempt et al. [2004].

The grade transition problem has been the subject of several papers. For gas phase reactors, McAuley and MacGregor [1992] uses the control variable parametrization method (CVP) with control profiles approximated by series of ramps, while in Gisnas et al. [2003] optimization results in bang-bang type solutions. A series of two slurry reactors has been considered by Takeda and Harmon Ray [1999], also using the method of CVP, and in Prata et al. [2008] a grade change for a series of a slurry and a gas phase reactor was performed with a discretization scheme based on direct single and multiple shooting.

This paper presents an optimization procedure for a grade change of a Borstar<sup>®</sup> polyethylene plant including three polyethylene reactors in series, two slurry and one gas phase reactor, and is an extension of Larsson et al. [2010] where one reactor was considered. The developed plant model is encoded in the Modelica language and a simultaneous optimization method based on collocation is applied using the optimization extension Optimica and the framework of JModelica.org.

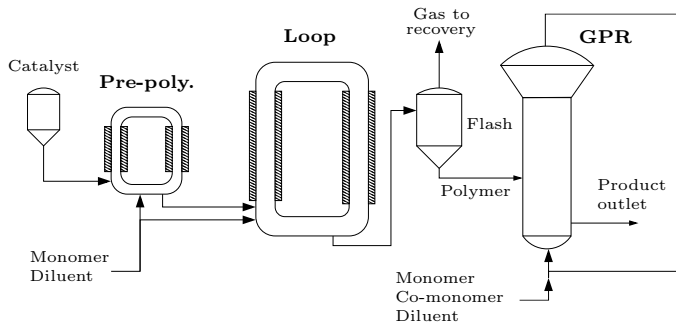


Fig. 1. Reactor chain of a Borstar<sup>®</sup> process: pre-polymerization, loop, and gas phase reactor.

## 2. BORSTAR<sup>®</sup> POLYETHYLENE PLANT

The Borstar<sup>®</sup> polyethylene plant incorporates in total three polyethylene reactors, two slurry and one gas phase reactor (GPR), see Figure 1. It is a bimodal plant and can thus produce polymer with a two-peaked molecular weight distribution. The pre-polymerization slurry reactors main function is to induce the polymerization in a pre-specified composition and has a negligible production compared to the two other reactors. It has catalyst, monomer ethylene and the chain transfer agent hydrogen along with the diluent propane as inflows. Using the second reactor, i.e., the loop slurry reactor, polymer with low molecular weight is shaped. The loop reactor has the same material inflow as the pre-polymerization reactor except that no catalyst is added. Temperature and pressure is high resulting in a super-critical state of the polyethylene slurry. The slurry stream from loop reactor is fed to a flash tank where gases are separated from polymer, which are transported to recovery area and the GPR, respectively. To the subsequent fluidized bed GPR, apart from the same type of raw material added to the loop reactor, also the co-monomer butene is added together with nitrogen. This gives the high molecular weight polymer resulting in the bimodal polymer product.

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### 3. PLANT MODEL

Several assumptions are made when modeling, keeping model complexity reasonable and usable for grade change optimization. The two loop reactors are assumed to have perfect temperature and pressure control, while control systems for reactor content volumes are in regulatory mode and incorporated in the model. Both loop reactors have high volumetric circulation rates compared to outflow rates. This results in recycle ratios well above 30, yielding no considerable gradients of molecular species nor temperatures exists along the reactors, see Zacca and Ray [1993]. The latter is also supported by measurements along the reactor. Thus, the loop reactors can be considered well-mixed. The outtakes of polymer from the loop reactors are settling legs, making the outflow have higher concentration of polymer than the reactor contents, see Reginato et al. [2003]. Hence, the approach of non-ideal CSTR for the loop reactors is suitable, see Reginato et al. [2003] and Touloupides et al. [2010], where the settling legs are modelled using discharge factors.

The temperature in the GPR is assumed to be controlled to a constant value and the regulatory system for bed level is incorporated in the model, using the outflow as control variable. The content in the GPR is assumed well mixed and conversion per pass through is low, making the gas composition approximately uniform in the bed, see e.g., Xie et al. [1994] and McAuley and MacGregor [1992]. Thus, the reactor may be modelled as a CSTR with outflow and reactor contents concentrations equal.

The model, derived by Borealis AB and used today in a non-linear model predictive controller of the plant, includes both first principles, semi-empirical, and empirical relations. The main inflows are, as described in Section 2, the raw materials, diluents and catalyst which gives a total of 12 control inputs available for optimization at a grade change. If the reactors are numbered from left to right with index  $j$ , i.e.,  $j \in \{1, 2, 3\}$ , then for every component  $i$  in Table 1 that is a fluid, gas or catalyst, the mass balance read

$$\dot{m}_{ij} = q_{i,j-1} + u_{ij} - q_{ij} - r_{ij}, \quad (1)$$

where  $q_{ij}$  and  $r_{ij}$  is the outflow and reaction rate of component  $i$ , respectively, in reactor  $j$  and  $u_{ij}$  is the controlled inflow of component  $i$ . Note that  $q_{ij}$ ,  $r_{ij}$  or  $u_{ij}$  might be zero depending on which reactor or component that is considered.

For the polymer, i.e., polyethylene and incorporated butene, it is important to indicate in what reactor it has been formed due to the process bi-modality, and therefore an extra index,  $k \in \{1, 2, 3\}$ , is supplied. For a solid component in a reactor it is formed in, i.e.,  $j = k$ , the mass balance is

$$\dot{m}_{ijk} = h_{ij} - q_{ijk}, \quad (2)$$

where  $h_{ij}$  is the production rate of the polymer component  $i$  in reactor  $j$  and is a function of the reaction rates of the raw materials in Eq. (1). For a polymer component in a reactor it has not been formed in, i.e.,  $j \neq k$ , it reads

$$\dot{m}_{ijk} = q_{i,j-1,k} - q_{ijk}, \quad (3)$$

which is a pure transportation of the component through the reactor. For every fluid or gas component, the instantaneous molar concentration  $X_{ij}$  may be calculated. Molar

Table 1. Components summary and abbrev.

Component	$i$	Component	$i$
Catalyst	$c$	Propane	$p$
Ethylene	$e$	Nitrogen	$n$
Hydrogen	$h$	Polyethylene	$pe$
Butene	$b$	Incorp. butene	$pb$

concentrations and all component masses for reactor  $j$  are collected in vectors  $\mathbf{X}_j$  and  $\mathbf{m}_j$ .

The reaction rates used above are calculated using extended Arrhenius expressions of the form

$$r_{ij} = R_{ij}(\mathbf{c}_j, P_j, \mathbf{m}_j, \mathbf{X}_j) \exp\left(\frac{k_{ij1}}{T_j} + k_{ij2}\right), \quad (4)$$

where  $R_{ij}$  is a non-linear function of the catalyst properties  $\mathbf{c}_j$ , reactor pressure  $P_j$  and the component masses and concentrations. The reaction rate is inversely proportional to the temperature  $T_j$  in the exponent with two empirical constant  $k_{ij1}$  and  $k_{ij2}$  for each component  $i$  and reactor  $j$ . Analogously, reaction rates are collected in the vector  $\mathbf{r}_j$ .

The states of the Ziegler-Natta catalyst in reactor  $j$ , such as mean activity and deactivated sites, have the non-linear dynamics of a function  $C_j$ ,

$$\dot{\mathbf{c}}_j = C_j(\mathbf{c}_j, \mathbf{r}_j, \mathbf{m}_j, q_{c,j-1}, u_{c1}), \quad (5)$$

where the two last variables are the inflow of catalyst from previous reactor and controlled input of catalyst to pre-polymerization reactor, respectively.

By using reactor geometry, knowledge of the reactor content properties and empirical relations for a super-critical state, the densities for fluids and solids in each reactor may be calculated. In general, densities have dependencies as

$$\boldsymbol{\rho}_j = \varrho_j(\mathbf{X}_j, P_j, T_j), \quad (6)$$

with the non-linear function  $\varrho_j$ , where  $\boldsymbol{\rho}_j$  is a vector containing densities for fluids and solids in reactor  $j$ .

Pressure in the GPR is given by reactor content properties and temperature through the non-linear function  $P_3$

$$P_3 = P_3(\mathbf{X}_3, \mathbf{m}_3, \boldsymbol{\rho}_3, T_3). \quad (7)$$

The resulting polymer properties depend on the ratios between monomer, co-monomer and hydrogen, see e.g. McAuley and MacGregor [1991], and the model therefore also includes the following ratios,

$$\begin{aligned} X_{he1} &= X_{h1}/X_{e1}, & X_{he2} &= X_{h2}/X_{e2}, \\ X_{he3} &= X_{h3}/X_{e3}, & X_{be3} &= X_{b3}/X_{e3}. \end{aligned} \quad (8)$$

Note that there is no butene-ethylene ratio in the pre-polymerization and loop reactors since co-monomer is only added in the GPR.

Due to the residence time in each reactor, also the bed average of the concentrations and concentration ratios defined above, emphasized with a bar, e.g.,  $\bar{X}_{ij}$ , are considered. These indicate during what conditions, in average, the polymer has been formed and are calculated by filtering the instantaneous values using the ratio between mass of solids and outflow of solids for considered reactor as time constant.

The bi-modality of the polyethylene molecular weight distribution is formed by producing polymers with different molecular weight in loop reactor and GPR. A measure of

the bi-modality is the split factor, calculated using masses of polyethylene produced in the different reactors as

$$S = \frac{m_{pb33} + m_{pe33}}{m_{pb33} + m_{pe33} + m_{pe32} + m_{pe31}}, \quad (9)$$

i.e., as the ratio of polymer mass in the GPR formed in the GPR to the total mass of polymer in the GPR.

For economical reasons, the production rate of solids in each reactor is considered when optimizing a transition, which is defined as the sum of production rate of polyethylene and incorporated butene in each reactor, i.e.,

$$Q_j = h_{pe,j} + h_{pb,j}. \quad (10)$$

The model contains, apart from Eqs. (1)–(10) also additional algebraic equations. If the inputs and outputs of the model are denoted  $\mathbf{u}$  and  $\mathbf{y}$ , respectively, and the states and algebraic variables are denoted  $\mathbf{x}$  and  $\mathbf{w}$ , the model can be written in the general non-linear index 1 differential algebraic equation (DAE) form

$$\begin{aligned} \mathbf{0} &= \mathbf{F}(\dot{\mathbf{x}}, \mathbf{x}, \mathbf{w}, \mathbf{u}) \\ \mathbf{y} &= \mathbf{g}(\mathbf{x}, \mathbf{w}, \mathbf{u}). \end{aligned} \quad (11)$$

The model has  $n_y$  outputs, used for defining a grade with correct production rate, and  $n_u = 12$  inputs and  $n_x = 55$  states,  $n_w = 180$  algebraic variables and 225 equations, disregarding  $\mathbf{g}(\cdot)$ , and has been subjected for calibration using plant measurement data in Andersson et al. [2011].

#### 4. MODELING AND OPTIMIZATION ENVIRONMENT

Modelica, a high level language for encoding of complex physical systems aimed at simulation and supporting object oriented concept is used for plant modeling. Main features of the language are that text-book style declarative differential and algebraic equations may be used and mixed, but it lacks language constructs for formulating dynamic optimization problems. Notations such as cost functions, constraints and mechanism to select input and parameters to optimize has been proposed in the Optimica extension, see Åkesson [2008], enabling the user to construct such problems based on Modelica models.

Numerical solver interfaces are typically written in C or FORTRAN and the translation of Modelica models and optimization problems are performed in the framework JModelica.org, an open source project targeted at dynamic optimization, see Åkesson et al. [2010]. It features compilers supporting code generation of Modelica/Optimica models to C, a C API for evaluating model equations and their derivatives, optimization algorithms and supports the Optimica extension.

The JModelica.org platform contains an implementation of a simultaneous optimization method based on collocation on finite elements, Biegler et al. [2002]. In this method, states, inputs and algebraic variables, are parametrized by Lagrange polynomials based on Radau points of order three, two and two, respectively. This corresponds to a fully implicit Runge-Kutta method, and possesses well known and strong stability properties. The dynamic optimization problem is thus translated into a non-linear program (NLP), which may be very large. To efficiently solve the NLP, derivative information together with sparsity patterns of the constraint Jacobians need to be provided to the numerical solver. In JModelica.org, the simultaneous optimization algorithm is interfaced with the large-scale

NLP solver IPOPT, see Wächter and Biegler [2006], particularly developed to solve NLP problems arising in simultaneous dynamic optimization. Simultaneous methods handle non-linear systems well, and also, constraints on state, input and algebraic variables are easily incorporated, and is thus well suited for the optimization of a grade transition treated in this paper.

Using the Modelica language, a library of necessary entities was constructed, see Larsson et al. [2010]. The library contains models both for simulation, experiments as well as optimization models with Optimica constructs.

### 5. OPTIMAL GRADE TRANSITION

#### 5.1 Grade Definition

Many measures may to be used when defining a grade, such as densities, molecular weight, molecular distribution, melt flow indices and raw material concentrations, which are controlled by polymerization conditions. Commercial practice is to give grade specifications by melt index and density due to convenience in industrial settings, see Xie et al. [1994]. Melt flow indices are measures of the molecular weight distribution and may be used to calculate the flow ratio, yielding a polydispersity measure. However, as indicated in Xie et al. [1994], these measures only give relative properties and not detailed information about polymer structure, which if desired, requires detailed models not suitable for over all plant optimization as considered in this paper.

In McAuley and MacGregor [1991], relations between melt index, density and reactor concentration ratios, both monomer and co-monomer, were presented based on reaction kinetics. Only a limited number of products is produced, and thus only a few steady state operation points exist where melt index may give measurement data. Thus, using this operation data, which is taken with a two hour interval, will give models with large uncertainties. Therefore, the concentration ratios, verified with process data, will be used when defining a grade. The concentration ratio data are calculated from chromatography measurements, i.e., component concentration measurements, with a sampling interval of 1 minute.

Except from the concentrations above also the split factor is used when defining a grade. Additionally, also the GPR pressure and the production rates are considered, although not effecting polymer properties directly. Table 2 lists variable values from a grade transition performed at Borealis AB, normalized by grade A. The reactant concentrations are specified from polymer properties while inert gases are given specifications from e.g., desired partial pressures, heat removal from exothermic reaction and other operating conditions. Note that e.g., specifying  $X_{e1}$  and  $X_{he1}$  implicitly also specifies  $X_{h1}$ , and thus also the remaining gas, i.e.,  $X_{p1}$ , since reactor content volume is constant at stationarity. Similar argument holds for  $X_{p2}$  and  $X_{n3}$ . The split factor is set to give correct bi-modality of the polymer. This, together with specifications of  $Q_1$  and  $Q_2$  implicitly defines production rate in GPR, and thus also total production rate. GPR pressure is set to nominal reactor value.

Table 2. Normalized grade definitions.

Grade	A	B	Grade	A	B
$X_{e1}$	1	1.000	$X_{p3}$	1	1.009
$X_{he1}$	1	0.37	$X_{e3}$	1	0.8828
$Q_1$	1	1.064	$X_{he3}$	1	1.846
$X_{e2}$	1	1.160	$X_{be3}$	1	1.279
$X_{he2}$	1	2.371	$S$	1	0.9167
$Q_2$	1	1.134	$P_3$	1	1

### 5.2 DAE Initialization

A grade transition changes production from one steady state to another, both representing on-specification production. The DAE initialization and stationarity problem for a specific grade requires the output  $\mathbf{y}$ , containing all variables defining a grade from the previous section, and derivatives  $\dot{\mathbf{x}}$  to be specified. The variables to solve for are  $\mathbf{x}$ ,  $\mathbf{u}$ , and  $\mathbf{w}$ . Here,  $\mathbf{u}$  is now an algebraic variable instead of input and its size determines how many grade specifications may be used, i.e., size of  $\mathbf{y}$ , if the non-linear equation system should have zero degrees of freedom. For full flexibility, as used here, all variables in  $\mathbf{u}$  will be free. If constructing the extended algebraic variable vector  $\mathbf{z} = [\mathbf{w}, \mathbf{u}]$ , the problem may be formulated as

$$\begin{aligned} 0 &= \tilde{\mathbf{F}}(\dot{\mathbf{x}}, \mathbf{x}, \mathbf{z}) \\ 0 &= \dot{\mathbf{x}} \\ 0 &= \tilde{\mathbf{g}}(\mathbf{x}, \mathbf{z}) - \mathbf{y}_{\text{spec.}}, \end{aligned}$$

where  $\tilde{\mathbf{F}}$  and  $\tilde{\mathbf{g}}$  corresponds to  $\mathbf{F}$  and  $\mathbf{g}$  without input  $\mathbf{u}$ , and  $\mathbf{y}_{\text{spec.}}$  contains the grade definitions in Table 2. The first equation has, as the DAE in Eq. (11),  $n_x + n_w$  equations, and the second and third has  $n_x$  and  $n_u$ , respectively, and thus, there is equally many equations as variables to solve for, yielding zero degrees of freedom.

The problem is encoded with JModelica.org using the Modelica models. Initial guesses may be set for all variables to solve for and upper and lower limits helps the solver. The solution time for each grade is less than 10 seconds.

Defining the two output vectors  $\mathbf{y}_A$  and  $\mathbf{y}_B$ , corresponding to the two grades, and solving the initialization problem, yields the inputs  $\mathbf{u}_A$  and  $\mathbf{u}_B$ , state vectors  $\mathbf{x}_A$  and  $\mathbf{x}_B$ , and algebraic vectors  $\mathbf{w}_A$  and  $\mathbf{w}_B$  for stationary on-specification production that may be used in the dynamic optimization.

### 5.3 Dynamic Optimization of Grade Transition

The dynamic optimization problem solves for optimal trajectories between the two grades A and B, satisfying dynamics and constraints on states, algebraic variables, inflows and outputs.

Initial conditions of the plant is the solution from DAE initialization for grade A, i.e.,  $\mathbf{x}_A$ ,  $\mathbf{w}_A$ , and  $\mathbf{u}_A$ , and a quadratic cost function that includes deviations from grade B specifications in form of  $\mathbf{x}_B$ ,  $\mathbf{w}_B$  and  $\mathbf{u}_B$  is used. Introducing the deviation vectors

$$\Delta \mathbf{y} = \mathbf{y} - \mathbf{y}_B, \quad \Delta \mathbf{u} = \mathbf{u} - \mathbf{u}_B, \quad \Delta \mathbf{w} = \mathbf{w} - \mathbf{w}_B,$$

the dynamic grade transition optimization problem can be formulated as

$$\begin{aligned} \min_{\dot{\mathbf{u}}} \int_{t_1}^{t_2} & \begin{bmatrix} \Delta \mathbf{y} \\ \Delta \mathbf{u} \\ \Delta \mathbf{w} \\ \dot{\mathbf{u}} \end{bmatrix}^T \begin{bmatrix} \mathbf{Q}_{\Delta \mathbf{y}} & 0 & 0 & 0 \\ 0 & \mathbf{Q}_{\Delta \mathbf{u}} & 0 & 0 \\ 0 & 0 & \mathbf{Q}_{\Delta \mathbf{w}} & 0 \\ 0 & 0 & 0 & \mathbf{Q}_{\dot{\mathbf{u}}} \end{bmatrix} \begin{bmatrix} \Delta \mathbf{y} \\ \Delta \mathbf{u} \\ \Delta \mathbf{w} \\ \dot{\mathbf{u}} \end{bmatrix} dt \\ \text{subj. to } & \mathbf{0} = \mathbf{F}(\dot{\mathbf{x}}, \mathbf{x}, \mathbf{w}, \mathbf{u}), \quad \mathbf{y} = \mathbf{g}(\mathbf{x}, \mathbf{w}, \mathbf{u}), \\ & \mathbf{y}_{\min} \leq \mathbf{y} \leq \mathbf{y}_{\max}, \quad \mathbf{u}_{\min} \leq \mathbf{u} \leq \mathbf{u}_{\max}, \\ & \mathbf{w}_{\min} \leq \mathbf{w} \leq \mathbf{w}_{\max}, \quad \dot{\mathbf{u}}_{\min} \leq \dot{\mathbf{u}} \leq \dot{\mathbf{u}}_{\max}, \\ & \mathbf{x}_{\min} \leq \mathbf{x} \leq \mathbf{x}_{\max}, \end{aligned}$$

The weight  $\mathbf{Q}_{\Delta \mathbf{y}}$  may be used to emphasize importance of the different grade defining variables, while  $\mathbf{Q}_{\Delta \mathbf{w}}$  and  $\mathbf{Q}_{\Delta \mathbf{u}}$  are used to remove too large over- and undershoots of algebraic variables and inflows. The optimization variables are the inflow derivatives  $\dot{\mathbf{u}}$ , which gives possibility to directly include them in the cost function using  $\mathbf{Q}_{\dot{\mathbf{u}}}$  to control inflow smoothness, and also in the constraints, without any additional filtering of the inflows  $\mathbf{u}$ . All weights are chosen diagonal for simplicity.

Over- and undershoots are accepted up to a certain limit for the instantaneous concentrations and ratios, i.e.,  $X_{ij}$ , set as constraints on  $\mathbf{y}$ . However, for the bed average concentrations and ratios, i.e.,  $\bar{X}_{ij}$ , and the split  $S$ , no over- or undershoots are accepted in the grade change. The constraints on the algebraic variables  $\mathbf{w}$  and states  $\mathbf{x}$  are for instance limits on volumes, pressures, reaction rates, component masses and catalyst properties, while constraints on inflows, both magnitudes and rates of changes, concern physical limits such as e.g., pump capacities.

### 5.4 Optimal Grade Transition Trajectories

For non-convex optimization it is advantageous to have good initial values. Since a simultaneous method is used, all variables at all discretization points are solved for at the same time, hence, initial trajectories should be supplied. Since the stationary points are known, i.e., solved for in DAE initialization problem, one can generate initial trajectories by ramping inflows from  $\mathbf{u}_A$  to  $\mathbf{u}_B$  and simulate the response. This can be performed in JModelica.org where simulation is available through SUNDIALS, see Hindmarsh et al. [2005].

The transition time is normalized to 1 time unit (t.u.) and the dynamic optimization problem have been solved several times for different element lengths approximately in the range 0.015-0.075 t.u. with 3 collocation points in each element. Resulting optimal inflows have been used as inputs in simulations, showing that the discretization is fine enough, i.e., the difference between simulated response and discretized response from optimization is negligible. After discretization, the NLP problem contains approximately 20.000-200.000 variables depending on number of elements. Using an Intel® Core™2 Duo CPU@3.00GHz, a solution is obtained in approximately 5-90 minutes depending on number of variables and initial values.

Figures 2-3 show the resulting optimal inflows to the loop reactor, key grade variables, and mass of polymer in loop reactor, respectively, while figures 4-6 show the corresponding for GPR. Note the scaling, i.e., the transition is 1 time unit (t.u.) and all variables have initial value 1.

Since the production rate  $Q_2$  is higher in grade B than in grade A, the inflow of ethylene is increased in total

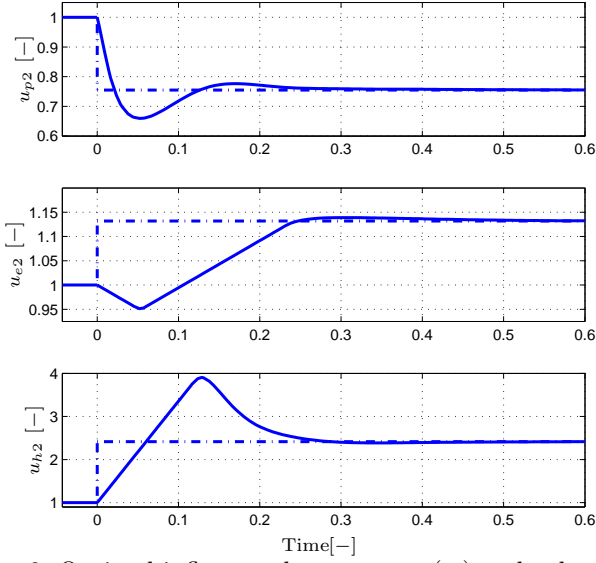


Fig. 2. Optimal inflows to loop reactor (—) and values for grade A and B at stationarity (---).

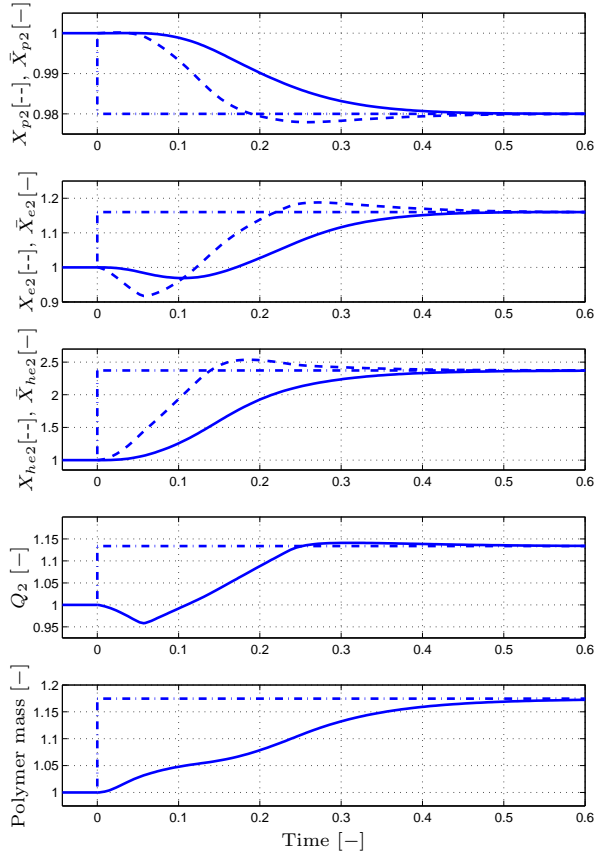


Fig. 3. Instantaneous (---) and bed average conc. and ratios, production rate and polymer mass (—) for loop and values for grade A and B at stationarity (---).

and at the same time inflow of the diluent propane is decreased, making room for more ethylene and hydrogen, as shown in Figure 2. This results in a larger mass of polymer in the loop, see Figure 3. Both  $X_{e2}$  and  $X_{h2}$  are higher in grade B than in grade A and to meet the hydrogen specification, the inflow of hydrogen is increased. To reach the specification of the hydrogen-ethylene ratio  $X_{he2}$  rapidly, the inflow of ethylene is initially decreased and hydrogen overshoot. Note that both the inflow of

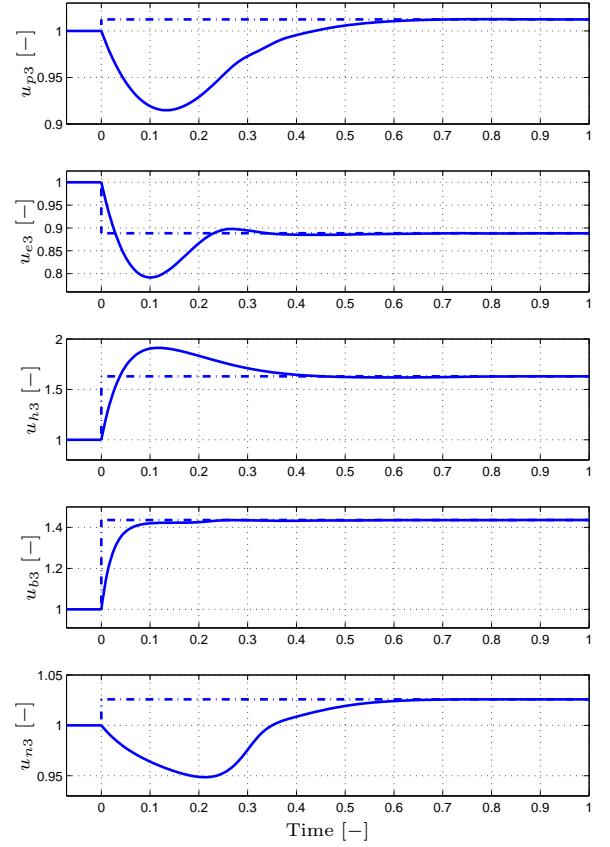


Fig. 4. Optimal inflows to GPR (—) and values for grade A and B at stationarity (---).

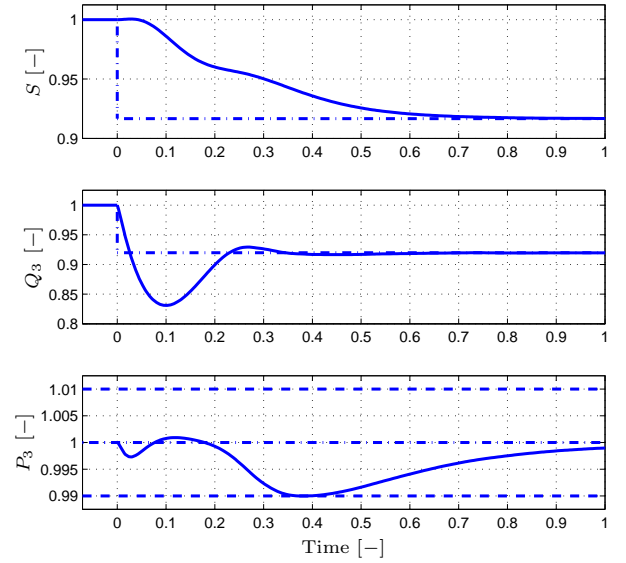


Fig. 5. Split, production rate and pressure for GPR (—) and values for grade A and B at stationarity (---), and pressure limits (---).

ethylene and hydrogen have their derivative constraints active in the beginning, seen by the linear decrease and increase, performing the transition of  $X_{he2}$  as fast as possible.

From Figure 3 it is seen that the under- or overshoot constraints on the averaged concentrations and ratios are followed and the instantaneous measures have over- or undershoots. The transition in the loop reactor is completed after 0.5 t.u.

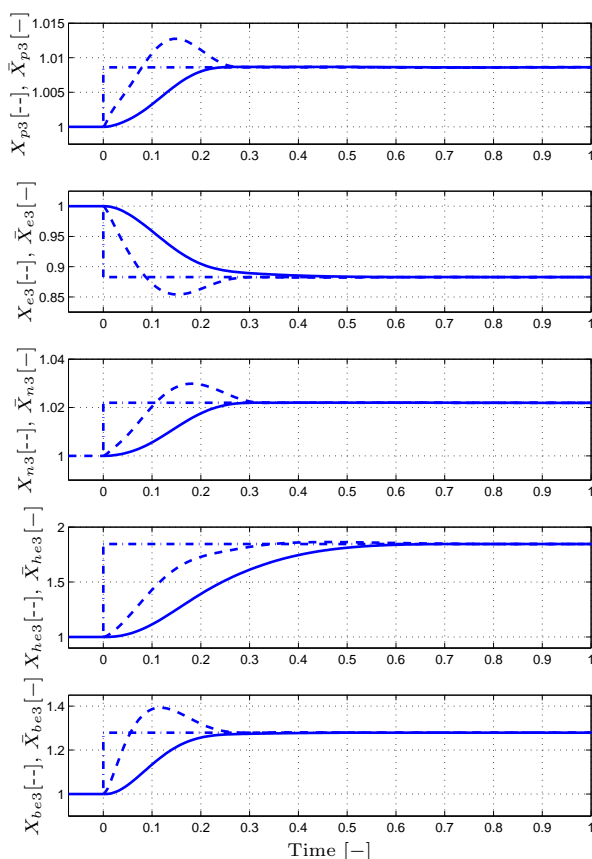


Fig. 6. Instantaneous (--) and bed average concentrations and ratios (—) for GPR and values for grade A and B at stationarity (---).

The split  $S$ , see Figure 5, which indirectly depends on the production rates, is decreased by increasing the production rate  $Q_2$  but also lowering production rate  $Q_3$ . This is performed by decreasing the ethylene inflow  $u_{e3}$ , see Figure 4, and thus also the ethylene concentration  $X_{e3}$ . The ethylene inflow has an undershoot giving a more rapid decrease of production rate. The change in  $u_{e3}$  is not enough for the specification on  $X_{be3}$  to be fulfilled and an increase of butene inflow,  $u_{b3}$ , is required, see figures 4 and 6. Analogously, inflow of hydrogen is increased. Propane and nitrogen are changed such that e.g., reactor pressure  $P_3$  is inside desired limits, specified to  $\pm 1\%$  from nominal pressure, see Figure 5.

All averaged concentrations and ratios in the GPR, and also in the pre-polymerization reactor although not shown here, together with the split, follow constraints of no under- or overshoot. Corresponding instantaneous values do have under- and overshoots, yielding faster transition.

## 6. SUMMARY AND FUTURE WORK

In this paper, modeling of multistage polyethylene reactors has been performed in the high level language Modelica. Optimal trajectories for transition between grades currently used at Borealis AB have been found by formulating an optimization problem using Optimica constructs and solved in the framework of JModelica.org.

Future work includes modeling of additional polymer specifications such as melt indices and densities and also economic objectives in the optimization formulation.

## REFERENCES

- J. Åkesson. Optimica—An Extension of Modelica Supporting Dynamic Optimization. In *6th Int. Modelica Conference 2008*. Modelica Assoc., March 2008.
- J. Åkesson, K.E. Årzén, M. Gäfvert, T. Bergdahl, and H. Tummescheit. Modeling and Optimization with Optimica and JModelica.org—Languages and Tools for Solving Large-Scale Dynamic Optimization Problem. *Computers and Chemical Engineering*, January 2010.
- N. Andersson, P. Larsson, J. Åkesson, S. Haugwitz, and B. Nilsson. Calibration of a polyethylene plant for grade change optimizations. In *21st European Symposium on Computer-Aided Process Engineering*, May 2011.
- L.T. Biegler, A.M. Cervantes, and A. Wächter. Advances in simultaneous strategies for dynamic optimization. *Chemical Engineering Science*, 57:575–593, 2002.
- A. Gísnas, B. Srinivasan, and D. Bonvin. Optimal grade transition for polyethylene reactors. In *Process Systems Engineering*, pages 463–468, Kunming, China, 2003.
- A. Hindmarsh, P. Brown, K. Grant, S. Lee, R. Serban, D. Shumaker, and C. Woodward. Sundials: Suite of nonlinear and differential/algebraic equation solvers. *ACM Trans. Math. Softw.*, 31:363–396, September 2005.
- P. Larsson, N. Andersson, J. Åkesson, and S. Haugwitz. Modeling and Opt. of Grade Changes for a Polyethylene Reactor. 9th Int. Symp. on Dynamics and Control of Process Systems, Leuven, Belgium, July 2010.
- K. B. McAuley and J. F. MacGregor. On-line inference of polymer properties in an industrial polyethylene reactor. *AIChE Journal*, 37(6):825–835, 1991.
- K.B. McAuley and J.F. MacGregor. Optimal grade transitions in a gas phase polyethylene reactor. *AIChE Journal*, 38(10):1564–1576, 1992.
- A. Prata, J. Oldenburg, and A. Kroll. Integrated scheduling and dynamic optimization of grade transitions for a continuous polymerization reactor. *Computers & Chemical Engineering*, 32(3):463–477, 2008.
- A. S. Reginato, J. J. Zacca, and A. R. Secchi. Modeling and simulation of propylene polymerization in nonideal loop reactors. *AIChE Journal*, 49(10):2642–2654, 2003.
- M. Takeda and W. Harmon Ray. Optimal-grade transition strategies for multistage polyolefin reactors. *AIChE Journal*, 45(8):1776–1793, 1999.
- V. Touloupides, V. Kanellopoulos, P. Pladis, C. Kiparisides, D. Mignon, and P. Van-Grambezen. Modeling and simulation of an industrial slurry-phase catalytic olefin polymerization reactor series. *Chemical Engineering Science*, 65(10):3208–3222, 2010.
- W. van Brempt, P. van Overschee, T. Backx, and O. Moen. Plantwide economical dynamic optimization: Application on a Borealis Borstar process model. In *AD-CHEM2003*, Hong Kong, 2004.
- A. Wächter and L. T. Biegler. On the implementation of an interior-point filter line-search algorithm for large-scale nonlinear programming. *Mathematical Programming*, 106(1):25–58, 2006.
- T. Xie, K. B. McAuley, C. C. Hsu, and D. W. Bacon. Gas phase ethylene polymerization: Production processes, polymer properties, and reactor modeling. *Ind. and Eng. Chemistry Research*, 33(3):449–479, 1994.
- J.J. Zacca and W. Ray. Modelling of the liquid phase polymerization of olefins in loop reactors. *Chemical Engineering Science*, 48(22):3743–3765, 1993.