

See discussions, stats, and author profiles for this publication at: <https://www.researchgate.net/publication/228368862>

Plantwide Optimal Grade Transition for an Industrial High-Density Polyethylene Plant

ARTICLE *in* INDUSTRIAL & ENGINEERING CHEMISTRY RESEARCH · JANUARY 2003

Impact Factor: 2.59 · DOI: 10.1021/ie0202221

CITATIONS

19

READS

30

5 AUTHORS, INCLUDING:



Heui-Seok Yi

Pohang University of Science and Technology

24 PUBLICATIONS 176 CITATIONS

SEE PROFILE

Plantwide Optimal Grade Transition for an Industrial High-Density Polyethylene Plant

Heui-Seok Yi

School of Environmental Science and Engineering, Pohang University of Science and Technology, San 31, Hyoja, Pohang, Kyungbuk 790-784, Korea

Jeong Hwan Kim and Chonghun Han*

Department of Chemical Engineering, Pohang University of Science and Technology, San 31, Hyoja, Pohang, Kyungbuk 790-784, Korea

Jinsuk Lee and Sang-Seop Na

Polymerization Process Development Team, Samsung General Chemical Company, Ltd., San 222-2, Dokgod, Daesan, Seosan, Chungnam 356-711, Korea

Industrial polymer plants are composed of reactors and downstream processes such as separators, dryers, and pelletizers. The polymer properties are delayed and changed in the downstream processes because of process dynamics. The polymer grade is determined by the properties of both the polymer slurry from the reactors and the polymer pellet from the pelletizer. Therefore, the grade transition strategy must be optimized in a plantwide viewpoint, and the models of downstream processes must be included in the optimization problem. In this study, a plantwide optimal grade transition strategy is proposed using the models of the reactors and the downstream processes, and the results are compared in a plantwide viewpoint. The plantwide grade transition time using the entire set of process models is shorter than that using the reactor model only.

1. Introduction

Polymers have wide application areas, and each area requires different grade specifications. Grade transition operation is essential in continuous polymer plants because many grades of polymers are produced from the same process. The reduction of the amount of off-specification polymer during the grade transition operation is important for the economical operation of continuous polymer plants.

There have been many research works about optimal grade transition operation and control of continuous polymer processes. McAuley and MacGregor¹ implemented the optimal grade transition for the gas-phase polyethylene reactor using melt index (MI) and density as the properties for grade specification. For a series of three polyethylene grades, the optimal manipulated inputs were determined by dynamic optimization. For the estimation of MI and density, an inference model was derived by incorporating the reaction kinetic models with an empirical relationship between MI and the average molecular weight. Takeda and Ray² solved the optimal grade transition problem of polyolefin loop reactors and compared the results with and without the constraints in the state variables. The reactor model based on the reaction kinetics was used to calculate the average molecular weight, density, and polydispersity (PD). Wang et al.³ integrated an offline optimizer and a nonlinear model predictive controller to perform the optimal grade transition operation for the slurry-phase polyethylene reactor. The nonlinear model predictive controller was used to compensate for the error between the controlled outputs and the actual plant outputs. Seki

et al.⁴ developed a nonlinear model predictive control for the polypropylene semibatch reactor and high-density polyethylene (HDPE) reactor. A successive linearization scheme was applied to obtain the linear model of a polymer reactor.

However, all of the previous works are limited to the dynamic optimization of a single unit. On the other hand, industrial polymer processes are composed of reactors and downstream processes such as separators, dryers, and pelletizers. The properties of the polymer slurry from the reactors are changed because of process dynamics when it is separated, dried, and pelletized. The polymer grades are determined by the MIs and the densities of the polymer slurry from the reactors and the polymer pellet from the pelletizers because the polymer slurry is the intermediate product. The slurry MIs in the reactors are used to control the molecular weight distribution (MWD) very roughly. MWD is less important than MI and the density for grade specification, and the wide MWD can be obtained by producing a polymer slurry that has a large difference between the MIs in the first and second reactors in a series of reactors. Therefore, both the properties of the polymer slurry and the polymer pellet must satisfy the grade specification after the grade transition operation, and the models of the reactors and downstream processes must be included in the dynamic optimization problem for grade transition. However, the previous research works used only the properties of the polymer slurry in reactors as the grade specification. The optimal grade transition using the properties of the polymer slurry as the grade specification makes the properties of the final product not satisfy the grade specification and results in an increase of the plantwide grade transition time.

In this paper, the optimal grade transition strategy for the industrial slurry-phase HDPE plant is proposed

* To whom correspondence should be addressed. E-mail: chan@postech.ac.kr. Fax: (+82) 54-279-3499. Tel: (+82) 54-279-2279.

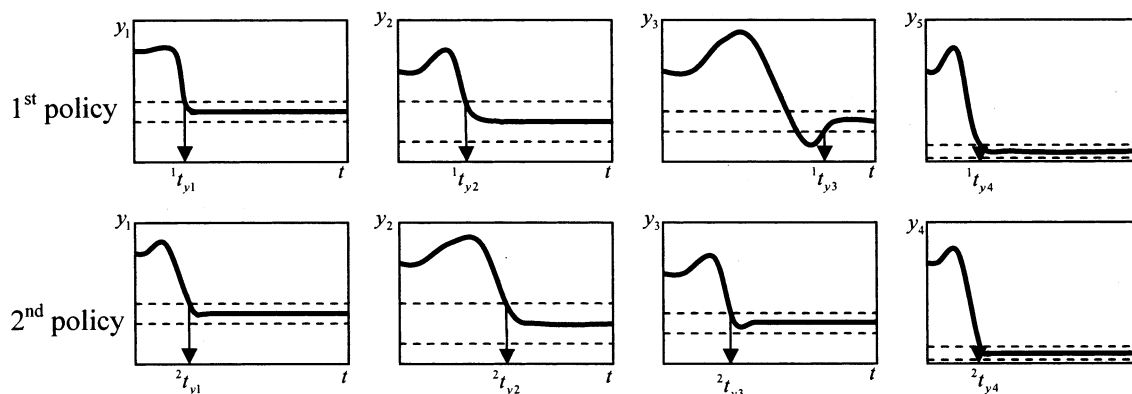


Figure 1. Comparison of the plantwide grade transition time.

in a plantwide viewpoint. The HDPE process models of the reactors, the separator, the dryer, and the pelletizer are used for the dynamic optimization model to include the properties of the polymer slurry and the polymer pellet in the objective function, and the results are compared with those by the reactor model only in a plantwide viewpoint. The empirical model combined with the first principle model is used because MI cannot be calculated by the first principle model and the moment equations. Regression models are used to estimate the MI value from the concentration of the reactants in reactors.^{5,6} A modified hierarchical method is applied to solve the dynamic optimization problem for the grade transition.⁷

2. Plantwide Grade Transition

Continuous polymer processes are composed of reactors and downstream processes such as separators, dryers, and pelletizers. The downstream processes recover the unreacted monomer and produce the polymer slurry as the final product. Polymer grades are determined by the MIs and densities of the polymer slurry from the reactors and the polymer pellet from the pelletizers. In a plantwide viewpoint, grade transition operation is completed if all of the properties satisfy the ranges for the grade specification. The plantwide grade transition time can be defined as the time when all of the properties satisfy the ranges for the grade specifications. It is required in industrial HDPE plants to satisfy all of the grade specifications together. Figure 1 shows the grade transition results for two different grade transition policies where four properties, y_1 (MI in the first reactor), y_2 (MI in the second reactor), y_3 (density of the final product), and y_4 (MI of the final product), determine the polymer grade. The individual grade transition times for properties y_1 , y_2 , and y_4 from the first policy are shorter than those from the second policy.

$${}^1t_{y1} < {}^2t_{y1}, {}^1t_{y2} < {}^2t_{y2}, {}^1t_{y4} < {}^2t_{y4} \quad (1)$$

where t_{yk} is the individual grade transition time of property yk by the i th policy. The individual grade transition time for property y_3 from the first policy is longer than that from the second policy

$${}^1t_{y3} > {}^2t_{y3} \quad (2)$$

The plantwide grade transition time by the first policy is ${}^1t_{y3}$, which is the longest grade transition time of the

individual grade transition times of y_1 , y_2 , y_3 , and y_4 . The plantwide grade transition time by the second policy is ${}^2t_{y2}$. The plantwide grade transition time by the second policy is shorter than that by the first policy, although three individual grade transition times by the first policy are shorter than those by the second policy. Therefore, it is important in industrial grade transition operations to make all properties satisfy grade specifications rather than only one of properties being used for grade specifications.

3. Case Study

3.1. Process Description. Figure 2 shows the simplified schematic diagram of the slurry-phase HDPE process for this study. It is composed of two continuously stirred tank reactors in series, the centrifugal separator, the dryer, and the pelletizer. Polymerization reaction occurs in the slurry phase. Ethylene monomer is the main raw material of the HDPE product, and hexane adjusts the slurry concentration in the reactors. Ethylene, hexane, and the catalyst are continuously fed to the reactors. Propylene or 1-butene is used as the comonomer and hydrogen as the chain-transfer agent. The polymer slurry from the first reactor is transferred to the flash tank for dropping the pressure and fed to the second reactor. The polymer slurry from the second reactor is not the final product but the intermediate product in polymer industries. The polymer slurry from the second reactor is fed to the separator to recycle the unreacted materials. The polymer from the separator is dried to remove the remaining solvent. The polymer powder from the dryer is pelletized as the final product. The polymer pellet from the pelletizer is stored in the silos.

Figure 3 shows the strategy for the grade transition of the HDPE product. The HDPE plant has measuring instruments for temperature, pressure, concentration, MI, and density. Measurements are used for updating the dynamic and steady-state models of the HDPE process to predict polymer properties. MI and density are the major properties of grade specification in polyethylene industries. MI is controlled by the molar ratios of hydrogen to ethylene in the gas phase in the reactors. Because the feed rates of ethylene are determined by the production rate, MI must be controlled by the feed rate of hydrogen to the reactors. Specifically, the polymer slurry MI from the first/second reactors and the polymer pellet MI from the pelletizer are used as the properties for grade specification in industrial HDPE processes. The density of the HDPE product is

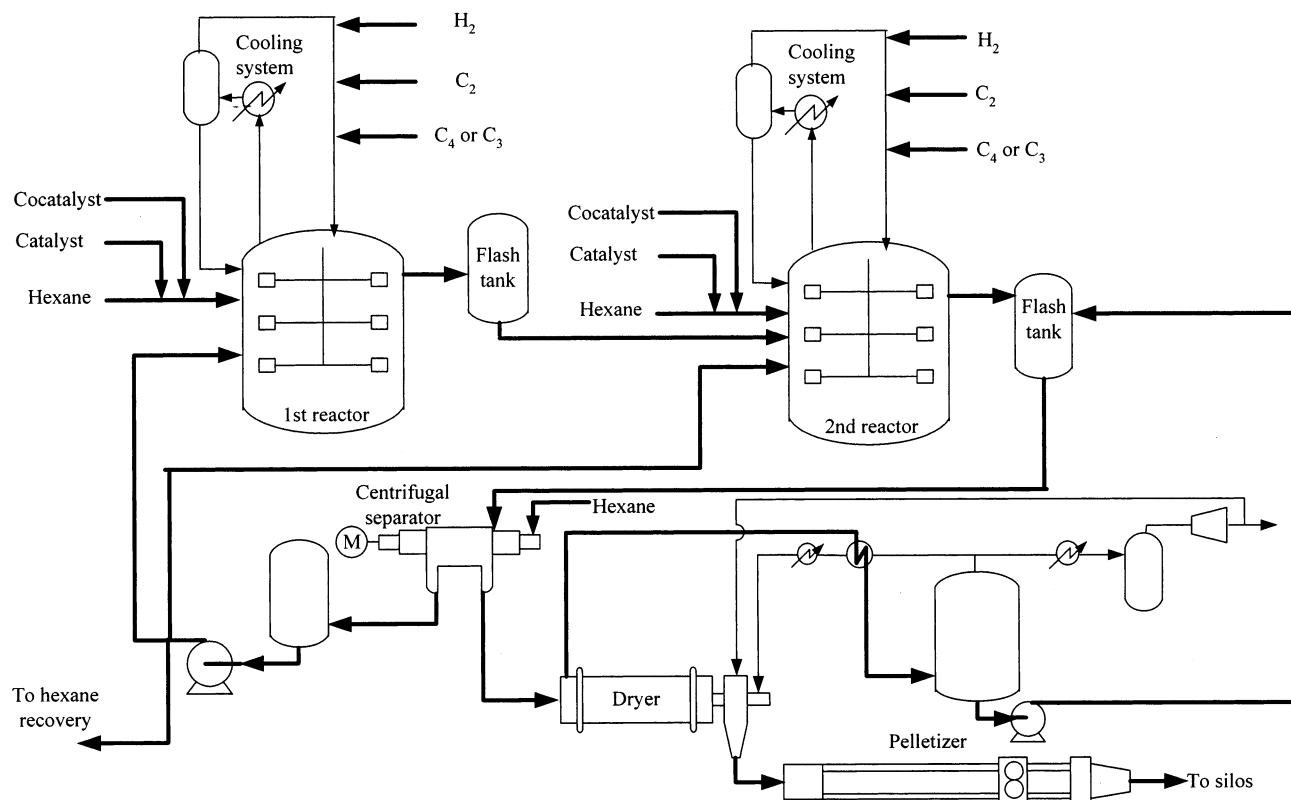


Figure 2. Process flow diagram of the slurry-phase HDPE polymerization process.

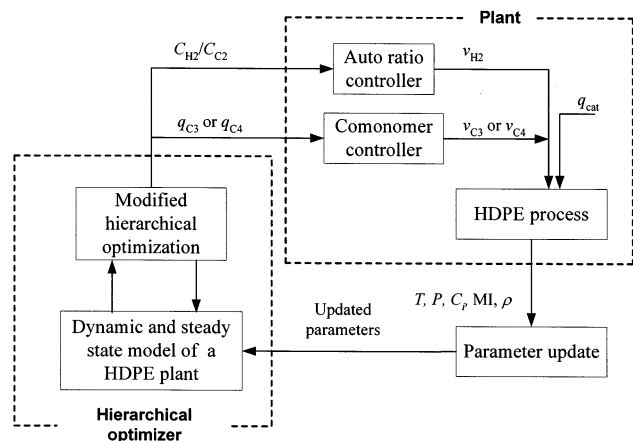


Figure 3. Strategy for grade transition in the industrial HDPE process.

controlled primarily by the feed flow rates of the comonomer. The density of the polymer pellet is obtained only after the pelletizer is used as the property for grade specification in the industrial HDPE process.

3.2. Problem Description and Formulation. The grade specification for the initial and target grades is shown in Table 1. The polymer slurry MI in the first/second reactors, the polymer pellet MI, and the polymer pellet density determine the grade of the HDPE products. Each grade specification has the values of minimum and maximum. The model for the HDPE process, which is available from industry, is used to optimize the grade transition operation.⁸ The brief summary of the HDPE process model is given in the appendix. A modified hierarchical dynamic optimization method is applied to the grade transition problem,⁷ which is the modification of a two-level hierarchical method.^{9,10} Figure 4 shows the procedure of the modified hierarchi-

cal dynamic optimization. The modified hierarchical method is composed of the upper level optimization, dynamic simulation, and the lower level optimization.

In the upper level optimization, the steady-state values of concentration ratios of hydrogen to ethylene in the first/second reactors and the feed flow rate of comonomer to the second reactor are formulated as decision variables and the objective function is formulated to satisfy the grade specification and to minimize the operating cost.

Upper level formulation

$$\min_{\mathbf{u}_{SS}} J = w_1 \left(1 - \frac{MI_1}{MI_{1,tg}} \right)^2 + w_2 \left(1 - \frac{MI_2}{MI_{2,tg}} \right)^2 + w_3 \left(1 - \frac{MI_{\text{pellet}}}{MI_{\text{pellet,tg}}} \right)^2 + w_4 \left(1 - \frac{\rho_{\text{pellet}}}{\rho_{\text{pellet,tg}}} \right)^2 + \mathbf{C}^T \mathbf{u}_{SS} \quad (3)$$

subject to

$$f(\mathbf{y}_{SS}, \mathbf{u}_{SS}) = 0 \quad (4)$$

$$\mathbf{u}_{SS,\min} \leq \mathbf{u}_{SS} \leq \mathbf{u}_{SS,\max} \quad (5)$$

$$\mathbf{y}_{SS,\min} \leq \mathbf{y}_{SS} \leq \mathbf{y}_{SS,\max} \quad (6)$$

Equation 4 is obtained by removing the accumulation terms of eqs A1–A9 in the appendix as zeros. MI_1 is the polymer slurry MI in the first reactor, MI_2 the polymer slurry MI in the second reactor, MI_{pellet} the MI of the polymer pellet, and ρ_{pellet} the density of polymer pellet. $MI_{i,tg}$ and $\rho_{i,tg}$ are target values of MI_i and ρ_i , respectively. \mathbf{C} is the vector of the operating cost for the HDPE process. w_1 , w_2 , w_3 , and w_4 are the weighting factors of MI and the density. \mathbf{u}_{SS} is the vector of control inputs at steady state for the next grade, and \mathbf{y}_{SS} is the

Table 1. Grade Specification for the Initial and Target Grades To Compare the Plantwide Grade Transition Time

	initial grade				target grade			
	1st reactor MI	2nd reactor MI	pellet MI	pellet density	1st reactor MI	2nd reactor MI	pellet MI	pellet density
minimum	80.0	0.39	0.5130	0.9500	784.0	0.07	0.0390	0.9535
standard	110.0	0.49	0.4830	0.9590	884.0	0.09	0.0460	0.9543
maximum	140.0	0.59	0.4530	0.9680	984.0	0.11	0.0530	0.9551

vector of algebraic state variables at steady state for the next grade. Dynamic simulation is implemented to calculate the final transition time, t_{ft} , using the optimal solution from the upper level as step inputs.

In the lower level optimization, the optimal trajectories of the concentration ratios of hydrogen to ethylene in the first/second reactors and the comonomer flow rate to the second reactor are formulated as decision variables. The objective function is formulated to minimize the off-specification polymer during the grade transition operation. The operating cost in the lower level optimization is omitted because the reduction of the off-specification polymer is essential or more important than the reduction of the operating cost during the grade transition operations.

Lower level formulation

$$\min_{\mathbf{u}(t_0:t_{fm}), t_{ft}} J = \int_{t_0}^{t_{ft}} \left[w_5 \left(1 - \frac{MI_1(t)}{MI_{1,tg}} \right)^2 + w_6 \left(1 - \frac{MI_2(t)}{MI_{2,tg}} \right)^2 + w_7 \left(1 - \frac{MI_{\text{pellet}}(t)}{MI_{\text{pellet,tg}}} \right)^2 + w_8 \left(1 - \frac{\rho_{\text{pellet}}(t)}{\rho_{\text{pellet,tg}}} \right)^2 \right] dt \quad (7)$$

subject to

$$f(\dot{\mathbf{x}}(t), \mathbf{x}(t), \mathbf{y}(t), \mathbf{u}(t), t) = \mathbf{0} \quad (8)$$

$$\mathbf{u}_{\min}(t) \leq \mathbf{u}(t) \leq \mathbf{u}_{\max}(t) \quad (9)$$

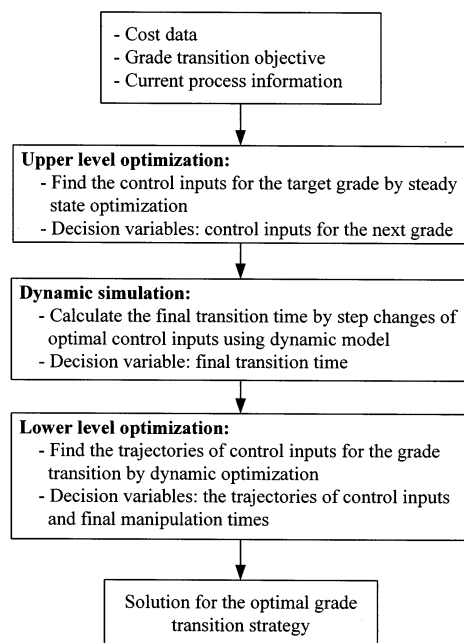
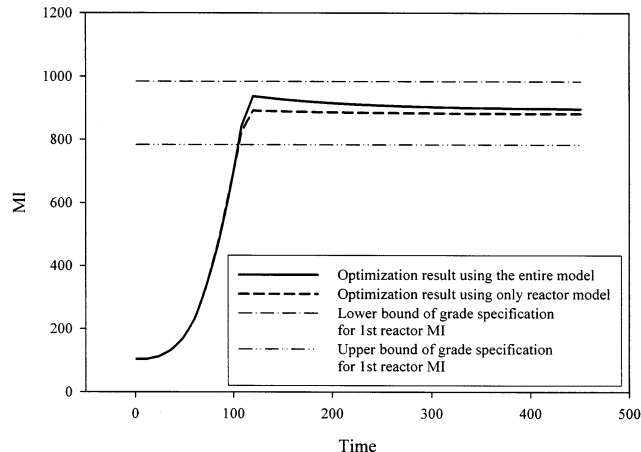
$$\mathbf{x}_{\min}(t) \leq \mathbf{x}(t) \leq \mathbf{x}_{\max}(t) \quad (10)$$

$$\mathbf{y}_{\min}(t) \leq \mathbf{y}(t) \leq \mathbf{y}_{\max}(t) \quad (11)$$

$$\Delta \mathbf{u}_{\min}(t) \leq \Delta \mathbf{u}(t) \leq \Delta \mathbf{u}_{\max}(t) \quad (12)$$

$$\mathbf{u}(t) = \begin{cases} \mathbf{u}(t_0:t_{fm}) & \text{if } 0 \leq t < t_{fm} \\ \mathbf{u}_{SS} & \text{if } t_{fm} \leq t \leq t_{ft} \end{cases} \quad (13)$$

Equation 8 is the vector form of the model equations for the HDPE processes such as eqs A1–A9 in the appendix. t_0 is the initial time for grade transition operation, t_{fm} the final manipulation time, and t_{ft} the final transition time. w_5 , w_6 , w_7 , and w_8 are the weighting factors of MI and the density. \mathbf{u} is the vector of control inputs, \mathbf{x} the vector of differential state variables, and \mathbf{y} the vector of algebraic state variables. $\mathbf{u}(t_0:t_{fm})$ means the vector of control inputs from the initial time, t_0 , to the final manipulation time, t_{fm} . The control inputs are optimized from the initial time to the final manipulation time at the lower level optimization. After the final manipulation time, the control inputs are fixed by the optimal solution at steady state obtained from the upper level optimization so that the polymer process can be maintained at the desired steady state for the next grade. Time weighting is used to penalize the long transition time.³ Control vector parametrization is used to solve the solution of the lower level

**Figure 4.** Procedure of modified hierarchical dynamic optimization methodology.**Figure 5.** Result comparison of the slurry MI in the first reactor using the entire model with that using the only reactor model.

problem.¹¹ The step size for control vector parametrization is 12 min, and its size is equal to the control action period of the existing controller in the HDPE process for this study because the smaller step size than the control action period cannot be implemented by the controller.

3.3. Results and Discussion. The results of dynamic optimization for the grade transition operation using the models of the reactors, the separator, the dryer, and the pelletizer are compared with those using only the reactor model in a plantwide viewpoint. Figure 5 compares the polymer slurry MI in the first reactor using the entire model of the HDPE process with that using the reactor model only. The dashed line is the result of the slurry MI in the first reactor by dynamic

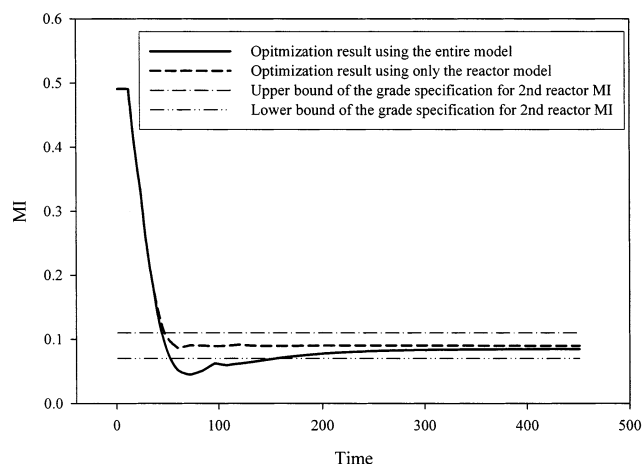


Figure 6. Result comparison of the slurry MI in the second reactor using the entire model with that using the only reactor model.

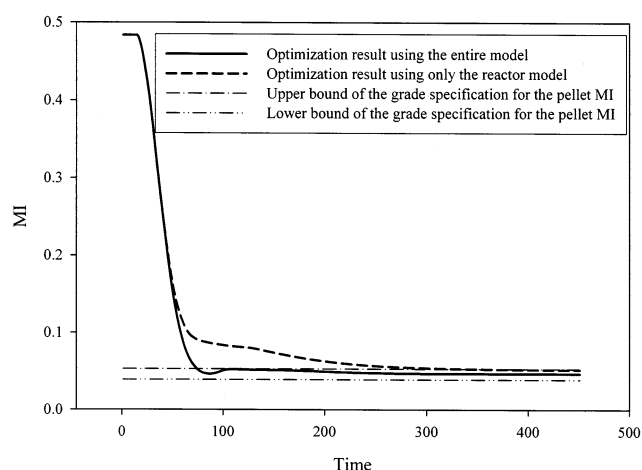


Figure 7. Result comparison of the polymer pellet MI using the entire model with that using the only reactor model.

optimization with the reactor model only. Therefore, the properties of the polymer pellet are not included in the optimization model, and only the properties of the polymer slurry are used to obtain the strategy of the optimal grade transition. The solid line is the result of the slurry MI in the first reactor by the dynamic optimization with the entire model. The results show the different steady-state conditions for the next grade operation. However, the final steady states of the two results satisfy the grade specification for the polymer slurry MI in the first reactor, and the two individual grade transition times have few differences.

Figure 6 shows the results of the polymer slurry MI in the second reactor. The MI profile of the polymer slurry in the second reactor using the entire HDPE process model shows a longer grade transition time than that using only the reactor model. The final steady states of both results satisfy the grade specification for the next target grade. The individual grade transition time using only the reactor model shows a shorter grade transition time than that using the entire process model in Figure 6. However, the plantwide grade transition time using only the reactor model is much longer than that using the entire process model, as shown in Figures 7 and 8. The plantwide grade transition time can be defined as the slowest one among the individual grade transition times of the polymer slurry MI in the first reactor, MI_1 , the polymer slurry MI in the second

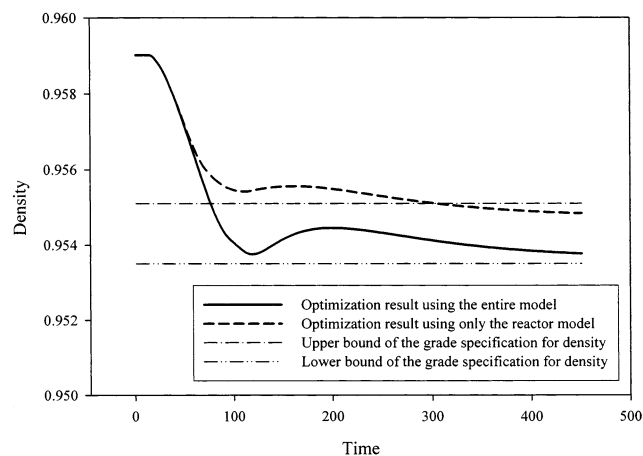


Figure 8. Result comparison of the polymer pellet density using the entire model with that using the only reactor model.

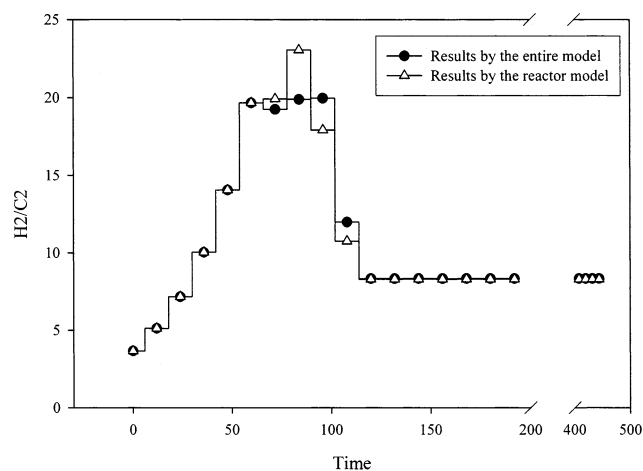


Figure 9. Result comparison of the optimal trajectory ratios of hydrogen to ethylene in the first reactor.

reactor, MI_2 , the MI of polymer pellet, MI_{pellet} , and the density of the polymer pellet, ρ_{pellet} . The grade transition time using only the reactor model is about 310 time units because the slowest grade transition time among the grade transition times of MI_1 , MI_2 , MI_{pellet} , and ρ_{pellet} is about 310 time units, which corresponds to the grade transition time of ρ_{pellet} . However, the grade transition time using the entire HDPE process model is about 150 time units because the slowest grade transition time or the grade transition time of MI_2 is about 150 time units. Therefore, the plantwide grade transition time from the entire HDPE process model is shorter than that by the reactor model only.

The allowable ranges of polymer properties for the dynamic optimization problem such as eq 11 are larger than the allowable ranges of polymer properties for steady-state optimization or grade specifications such as eq 6. The MI change in the second reactor affects the MI change of the pellet. To satisfy the grade specification for pellet MI, the MI profile in the second reactor using the entire model is deviated from the range of grade specification in the initial region of the grade transition time.

Figures 9–11 show the results of the manipulated inputs by dynamic optimization. The lines with dots are the results of the entire process model and the lines with triangles whose of the reactor model only. In general, the manipulated inputs of the first process move more aggressively than the manipulated inputs of the second

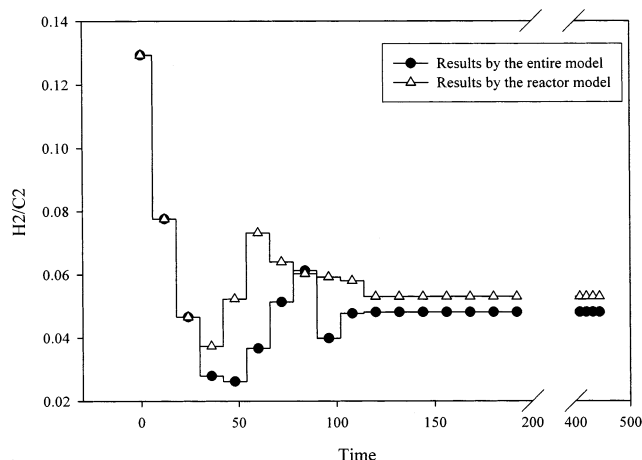


Figure 10. Result comparison of the optimal trajectory ratios of hydrogen to ethylene in the second reactor.

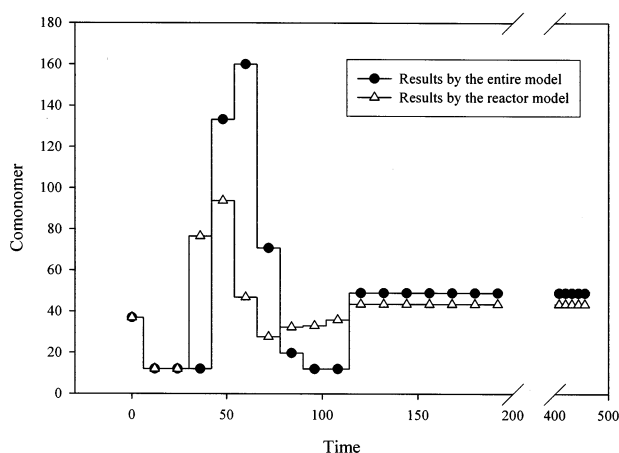


Figure 11. Result comparison of the comonomer flow rate in the second reactor.

process when a series of processes are optimized to the reduced transition time. The trajectory ratios of hydrogen to ethylene in the first reactor move more aggressively than the trajectory ratios in the second reactor, as shown in Figures 9 and 10. However, the dynamic optimization results of the comonomer flow rate in the second reactor show large changes although it is the second reactor in the series of processes because there is no comonomer flow fed into the first reactor. If the comonomer flows fed into the first and second reactors exist, the results of the comonomer flow in the first reactor move more aggressively than those in the second reactor.

4. Conclusions

The optimal grade transition strategy for the industrial HDPE processes is proposed to reduce the grade transition time in a plantwide viewpoint. The process models of the reactors and the downstream processes are included in the optimization problem, and the results are compared with those using the reactor model only. The individual grade transition time for the MI in the first reactor has little difference. The individual grade transition time for the MI in the second reactor using the entire model is longer than that using the reactor model only. However, the plantwide grade transition time using the entire process model is shorter than that using the reactor model only. Therefore, the

dynamic optimization of a single unit, such as reactors, separators, dryers, etc., is not a desirable grade transition policy, and the dynamic optimization for grade transition must be implemented in a plantwide viewpoint.

Acknowledgment

This work was supported by the National Research Laboratory project (2000-N-NL-01-C-18) and Brain Korea 21 project.

Appendix: Summary of the HDPE Process Models

The HDPE process models for this study are composed of four submodels: (1) the mass balance models to calculate the concentration of monomer, comonomer, hydrogen, and polymer from the feed rates and the reaction rates of each component, (2) the instantaneous property models to calculate the instantaneous properties of the polymer from the concentration of each component, (3) the cumulative property models to calculate the cumulative properties of the polymer from the instantaneous properties, and (4) the empirical model to calculate the change of properties from the polymer slurry to the polymer pellet in the centrifugal separator, the dryer, and the pelletizer. The detailed description of the process models and parameters is found elsewhere.⁸

Mass Balance Equations for Each Component.

The HDPE process has two reactors in series followed by two flash tanks for controlling pressure. Because the polymerization reactions occur in flash tanks during a significant amount of time, the flash tanks are assumed as reactors. The total number of reactors is regarded as four.

Mass balance of polymer

$$\frac{dV_{w,i}C_{sl,i}}{dt} = q_{sl,f,i}C_{sl,f,i} - q_{sl,o,i}C_{sl,i} + W_{p,i} \quad (i = 1, 2, \dots, 4) \quad (A1)$$

where $C_{sl,i}$ = slurry concentration in the i th reactor (kg/m^3), $C_{sl,f,i}$ = slurry concentration fed into the i th reactor (kg/m^3), $q_{sl,f,i}$ = slurry flow rate fed into the i th reactor (m^3/h), $q_{sl,o,i}$ = output slurry flow rate from the i th reactor (m^3/h), $V_{w,i}$ = working volume of the i th reactor (m^3), and $W_{p,i}$ = polymer generation rate in the i th reactor (kg/h).

Mass balance of monomers

$$\frac{dM_{m,i}}{dt} = \dot{M}_{m,f,i} - \dot{M}_{m,o,i} - W_{m,i} \quad (i = 1, 2, \dots, 4) \quad (A2)$$

where $M_{m,i}$ = amount of monomer in the i th reactor (kg), $\dot{M}_{m,f,i}$ = monomer flow rate fed into the i th reactor (kg/h), $\dot{M}_{m,o,i}$ = output monomer flow rate from the i th reactor (kg/h), and $W_{m,i}$ = monomer consumption rate in the i th reactor (kg/h).

Mass balance of catalyst

$$\frac{dM_{cat,i}}{dt} = \dot{M}_{cat,f,i} - \dot{M}_{cat,o,i} - V_{w,i}k_{dsp,i}C_{cat,i}^{a_{dsp,i}} \quad (i = 1, 2, \dots, 4) \quad (A3)$$

where $C_{cat,i}$ = catalyst concentration in the i th reactor

(kg/m³), $k_{\text{dsp},i}$ = spontaneous deactivation rate constant of the catalyst (h⁻¹), $M_{\text{cat},i}$ = amount of catalyst in the i th reactor (kg), $\dot{M}_{\text{cat},f,i}$ = catalyst flow rate fed into the i th reactor (kg/h), and $\dot{M}_{\text{cat},o,i}$ = output flow rate of the catalyst from the i th reactor (kg).

Mass balance of hydrogen

$$\frac{dM_{\text{H}_2,i}}{dt} = \dot{M}_{\text{H}_2,f,i} - \dot{M}_{\text{H}_2,o,i} - V_{w,i} k_{\text{H}_2,i} C_{\text{H}_2,i} C_{\text{cat},i} \quad (i = 1, 2, \dots, 4) \quad (\text{A4})$$

where $C_{\text{H}_2,i}$ = hydrogen concentration in the i th reactor (kg/m³), k_{H_2} = rate constant of chain transfer to hydrogen (m³/kg·h), $\dot{M}_{\text{cat},o,i}$ = output hydrogen flow from the i th reactor (kg/h), $M_{\text{H}_2,i}$ = amount of hydrogen in the i th reactor (kg), and $\dot{M}_{\text{H}_2,f,i}$ = hydrogen flow rate fed into the i th reactor (kg/h).

Instantaneous Polymer Properties in Reactors. The instantaneous properties of the polymer are determined by the empirical equations from the reaction temperature and the concentrations of hydrogen, monomer, and comonomer.

Instantaneous MI equation

$$\log \text{MI}_{\text{inst}} = a_0 + a_1 \log T + a_2 \log C_{\text{H}_2/\text{C}_2} + a_3 M_{\text{C}_3/\text{C}_2} + a_4 M_{\text{C}_4/\text{C}_2} \quad (\text{A5})$$

where $C_{\text{H}_2/\text{C}_2}$ = concentration ratio of hydrogen to ethylene in a reactor (mol %/mol %), MI_{inst} = instantaneous MI (g/10 min), $M_{\text{C}_3/\text{C}_2}$ = mass ratio of propylene to ethylene in a reactor (kg/kg), $M_{\text{C}_4/\text{C}_2}$ = mass ratio of 1-butene to ethylene in a reactor (kg/kg), and T = reaction temperature (°C).

Instantaneous density equation

$$\rho_{\text{inst}} = b_0 + b_1 \log T + b_2 \log C_{\text{H}_2/\text{C}_2} + b_3 M_{\text{C}_3/\text{C}_2} + b_4 M_{\text{C}_4/\text{C}_2} \quad (\text{A6})$$

where ρ_{inst} = instantaneous density of the polymer (kg/m³).

Cumulative Polymer Properties in Reactors. The cumulative properties of the polymer are calculated from the instantaneous properties of the polymer by empirical equations.

Cumulative MI equation

$$\frac{dM_{p,i} \log \text{MI}_{\text{cum},i}}{dt} = W_{p,f,i} \log \text{MI}_{\text{cum},i-1} + W_{p,i} \log \text{MI}_{\text{inst},i} - W_{p,o,i} \log \text{MI}_{\text{cum},i} \quad (\text{A7})$$

where $\text{MI}_{\text{cum},i}$ = cumulative MI of the polymer in the i th reactor (g/10 min), $M_{p,i}$ = amount of the polymer in the i th reactor (kg), $W_{p,f,i}$ = polymer feed rate into the i th reactor (kg/h), and $W_{p,o,i}$ = output polymer flow rate from the i th reactor (kg/h).

Cumulative density equation

$$\frac{dM_{p,i} \rho_{\text{cum},i}}{dt} = \frac{W_{p,f,i}}{\rho_{\text{cum},i-1}} + \frac{W_{p,i}}{\rho_{\text{inst},i}} - \frac{W_{p,o,i}}{\rho_{\text{cum},i}} \quad (\text{A8})$$

where $\rho_{\text{cum},i}$ = cumulative density of the polymer in the i th reactor (kg/m³).

Property Change Model in the Downstream Processes. The change of the cumulative MI during pelletizing is modeled empirically using the density, MI, blending ratio, and amount of antioxidant. Antioxidant prevents polymer from the oxidation that makes the MI of the polymer increase in the downstream processes. A simple time delay model is also used to represent the dynamics in the centrifugal separator, the dryer, and the pelletizer.

MI change in the downstream processes

$$\text{MI}_{\text{plt}} = d_0 + d_1 \rho_{\text{cum},2} + d_2 \text{BR} \log \text{MI}_{\text{inst},1} + d_3 (1 - \text{BR}) \log \text{MI}_{\text{inst},2} + d_4 \text{MI}_{\text{cum},2} + d_5 C_{\text{AO}} \quad (\text{A9})$$

where BR = ratio of the ethylene feed in the first reactor to the ethylene feed in the second reactor [(kg/h)/(kg/h)], C_{AO} = concentration of antioxidant (ppm), and MI_{plt} = pellet MI (g/10 min).

Nomenclature

C = vector of the operating cost

J = objective function

MI_1 = polymer slurry MI in the first reactor

MI_2 = polymer slurry MI in the second reactor

$\text{MI}_{\text{pellet}}$ = pellet MI of HDPE

t_0 = initial time for the grade transition operation

t_{fm} = final manipulation time for the grade transition operation

t_{ft} = final transition time for the grade transition operation

t_{yk} = grade transition time of property yk by the i th policy

u = vector of control input variables

$w_1 - w_8$ = weighting factor in the formulation of optimization

x = vector of differential state variables

y = vector of algebraic state variables

Subscripts

max = maximum

min = minimum

SS = steady state

tg = target value

Greek Letter

ρ_{pellet} = pellet density of HDPE

Literature Cited

- (1) McAuley, K. B.; MacGregor, J. F. Optimal Grade Transitions in a Gas-Phase Polyethylene Reactor. *AIChE J.* **1992**, *38*, 1564.
- (2) Takeda, M.; Ray, W. H. Optimal-Grade Transition Strategies for Multistage Polyolefin Reactors. *AIChE J.* **1999**, *45*, 1776.
- (3) Wang, Y.; Seki, H.; Ohyama, S.; Akamatsu, K.; Ogawa, M.; Ohshima, M. Optimal Grade Transition Control for Polymerization Reactors. *Comput. Chem. Eng.* **2000**, *24*, 1555.
- (4) Seki, H.; Ogawa, M.; Ooyama, S.; Akamatsu, K.; Ohshima, M.; Yang, W. Industrial Application of a Nonlinear Model Predictive Control to Polymerization Reactors. *Control Eng. Pract.* **2001**, *9*, 819.
- (5) Kim, J. Y.; Yi, H.-S. Prediction of Overall Heat Transfer Coefficient for Low-Density Polyethylene Tubular Reactors. *Hwahak Konghak* **1999**, *37*, 494.
- (6) Kim, J. Y.; Yi, H.-S. Analysis of Polyethylene Tubular Reactors with Multiple Reaction Zone (I): The Effect on Production Polymer Properties. *Polymer (Korea)* **1999**, *23*, 348.
- (7) Yi, H.-S.; Han, C.; Na, S.-S.; Lee, J. Optimal Grade Transition with Partially Structured Model in a Slurry-Phased HDPE Reactor by Modified Hierarchical Dynamic Optimization. International Conference on Control, Automation and Systems, Cheju, Korea, 2001.

(8) Lee, J.; Kim, W. K.; Kim, S. H. Method of Estimating Product Polymer Properties. Korea Patent Application No. 10-2000-0049232, 2000.

(9) Lee, M. H.; Han, C.; Chang, K. S. Hierarchical Time-Optimal Control of a Continuous Copolymerization Reactor during Start-Up or Grade Change Operation Using Genetic Algorithms. *Comput. Chem. Eng.* **1997**, S1037.

(10) Lee, M. H.; Han, C.; Chang, K. S. Dynamic Optimization of a Continuous Polymer Reactor Using a Modified Differential Evolution Algorithm. *Ind. Eng. Chem. Res.* **1999**, 38, 4825.

(11) Sargent, R. W. H.; Sullivan, G. R. The Development of an Efficient Optimal Control Package. In *Proceedings of the 8th IFIP Conference on Optimization Technique*; Stoer, J., Ed.; Springer-Verlag: Berlin, 1978.

Received for review March 26, 2002

Revised manuscript received October 4, 2002

Accepted November 1, 2002

IE0202221