Optimizing Control of CSTR/Distillation Column Processes with One Material Recycle

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Control system design for recycle processes that are composed of a continuous stirred tank reactor (CSTR) and a distillation column is discussed. It is shown that the reflux ratio control, which has been known to serve as a self-optimizing control for the process with the elementary chemistry A → B, can be also applied to processes with more-general types of chemistry that involves side reactions. Using the self-optimizing control as the basis, control systems that seek economically optimal operations are hierarchically constructed for example processes with different types of process chemistry. Particularly, for a process that exhibits tradeoff between product yield and separation cost, a linear model predictive controller design example is given with simulation results.

1. Introduction

A reactor/separator system with a material recycle is one of the very common process configurations found in chemical production. It is well-known that the presence of recycle changes the behavior of the plant and a particular attention should be paid to control system design; control strategies designed for isolated units sometimes fail in the plantwide context.[1]

As a simple example of reactor/separator systems with recycle, several researchers[2–6] studied the process, which is composed of a continuous stirred tank reactor (CSTR), a distillation column, and a single material recycle shown in Figure 1, assuming the most elementary reaction, that is, the first-order irreversible reaction A → B. Luyben[2] suggested that flow in the recycle loop should be kept constant and the reactor holdup be permitted to change, because the recycle flow rate is very sensitive to changes in the feed flow rate and a "snowball" effect may occur. Wu et al.[3] showed that, for a fixed reactor effluent flow, the reactor holdup has a high gain for feed rate changes and they suggested that the reactor composition be controlled. In addition to the conventional control configuration, they proposed using the reactor temperature to control reactor composition. Monroy-Loperena et al.[6] also suggested using the reactor temperature to effectively handle disturbances.

One interesting feature of the reactor/separator system with recycle is that there exists an unconstrained optimum in the energy consumption. More precisely, when the distillation column bottom composition is regulated, a well-defined minimum appears in the vapor boilup for a certain set of manipulated variables. Taking advantage of this feature, Larsson et al.[5] designed a self-optimizing controller[7] by setting the ratio of the reflux flow rate to the column feed at some appropriate value. They used a fixed maximum reactor volume and temperature to maximize the per-pass conversion for economically optimal operations.

A major criticism of the aforementioned studies may be that the reaction A → B is too simple to accommodate the most general case, because no side reaction is considered and the important real-world problem of conversion/selectivity issue never arises. Ward et al.[8] noted the importance of process chemistry in deciding steady-state operation policy for reactor/separator systems with recycle; depending on the kinetics of desired and undesired reactions, manipulation of recycle streams may have different effects on the production rate of the undesired product, so that economically optimal operating policies should be different accordingly. They categorized process chemistry into "bounded" and "nonbounded", and derived heuristics for a plantwide operating policy for processes with one recycle stream.[9] The process chemistry is called "bounded" if the minimum production rate of the undesired product is *bounded* by the reactor volume constraint. In other words, the byproduct production rate is reduced for a decreased recycle flow, so that an operating policy that seeks to minimize the byproduct production rate for a fixed production rate of the desired species will be to minimize the recycle flow rate and keep the reactor holdup as large as possible to keep the per-pass conversion as high as possible. On the other hand, when the byproduct production rate decreases for an increased recycle flow, the chemistry is called "nonbounded".

In deriving the heuristics for the operation policy, Ward et al. assumed that the recycle stream is essentially pure and did not specify the nature of separation system.[8–10] They did not take full advantage of the self-optimizing control by Larsson et al.,[5] which focuses on the operational degree of freedom in the distillation column.

In this paper, the separation system is specified as a distillation column unit and, first, we investigate whether the self-optimizing control configuration, which has been developed for the recycle process with the elementary chemistry A → B, is still applicable to the more-general cases for bounded and nonbounded chemistries. For that purpose, the origin of appearance of the unconstrained minimum in the energy consumption will be identified. Also, the appropriate regulatory control loop configuration that realizes the self-optimizing control will be discussed with a focus on the effect of the recycle flow on the dynamics of the closed loop. Then, in addition to the self-optimizing control, the optimizing control that seeks steady-state economically optimal operation will be hierarchically realized, based on the heuristics derived by Ward et al.;[10] especially for recycle processes with nonbounded chemistry, a supervisory optimizing controller will be designed in the framework of linear model predictive control (MPC) with simulation examples.

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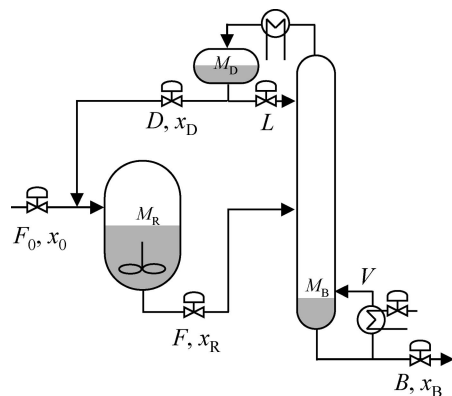
Figure 1. Schematic of a reactor/separator system with one material recycle.

2. Recycle System with the Simple Reaction A → B

As the simplest example of reactor/separator processes with recycle, a binary system that assumes an irreversible first-order reaction A → B is studied in this section. As shown in Figure 1, a fresh feed that contains A is fed to the CSTR and the reactor effluent is sent to the distillation column, where the unreacted A is separated and recycled back from the top of the column.

The following assumptions are made for control system design:

(1) The feed rate F_0 and composition x_0 are given.

(2) The distillation bottom composition x_B is specified as product purity and must be regulated by the vapor boilup (V).

(3) The three inventories—that is, the holdups of the reactor (M_R), the distillation column bottom (M_B), and the reflux drum (M_D)—must be stabilized.

(4) The column pressure is maintained constant, controlled by the overhead condenser.

Then, the operational degrees of freedom for this recycle system is three: the reactor temperature T_R, the reactor holdup M_R, and the remaining variable involves the distillation column. Note that the throughput is assumed to be determined directly by the given feed rate F_0 here; however, this assumption will be lifted later.

As noted by Larsson et al.[5] and Ward et al.,[8] it is economically optimal to seek the maximum per pass conversion and operate the reactor at its full capacity (that is, maximum holdup and reaction temperature); in terms of the reaction temperature, the maximum temperature is optimal when Arrhenius-type reaction kinetics can be assumed. Particularly, Ward et al.[8] noted that this operating policy is valid because there are no side reactions and no need to consider a conversion/selectivity tradeoff. With the reactor temperature and holdup being kept at their upper limits, the remaining operational degree of freedom is that for operation of the distillation column.

2.1. Unconstrained Minimum for Energy Consumption. One interesting feature of this recycle system is that there exists a well-defined minimum in the vapor boilup V when the bottom composition x_B is regulated at some constant value for a fixed feeding condition and reactor operation (F_0, x_0, M_R, and T_R are constant). The origin of appearance of such a minimum can be identified as follows.

The mass balance equations are given as

$$F_0 + D - F = 0$$

$$F_0 x_0 + D x_D - k x_R M_R - F x_R = 0$$

$$F - B - D = 0$$

$$F x_R - B x_B - D x_D = 0$$

where k is the kinetic constant.

The concentrations of species A in the reactor (x_R) and in the recycle stream (x_D) then can be expressed by

$$x_R = \frac{F_0(x_0 - x_B)}{k M_R} \tag{1}$$

$$x_D = \frac{F_0}{D}(x_R - x_B) + x_R \tag{2}$$

Equation 1 implies that the composition x_R is determined independent of x_D; note that the column bottom composition (x_B) is regulated. In other words, specification of x_B imposes the mass balance constraint for the consumption rate of A to be $F_0(x_0 - x_B)$. The consumption rate of A, consequently, x_R, then is determined.

Furthermore, eq 2 implies that the column top composition x_D is a decreasing function of D. Therefore, in the combined system, which is comprised of the reactor and the distillation column, increasing the recycle flow D has competing effects on the column load: an increased column feed $F_0 + D$ and decreased separation requirement for the top composition x_D. This can be interpreted as the origin of the appearance of a minimum in the vapor boilup (V).

In the following discussion, the origin of the appearance of such a minimum is shown more quantitatively, assuming an ideal binary distillation column with constant liquid and vapor molar flows.

As an isolated unit, a binary distillation column with its bottom composition x_B fixed at some constant value generally has the following properties (F and x_D are chosen as independent variables):

$$\left(\frac{\partial V}{\partial x_D}\right)_F > 0, \quad \left(\frac{\partial V}{\partial F}\right)_{x_D} > 0 \tag{3}$$

As a combined system with x_D as the sole operational degree of freedom, because $F = F_0 + D$ and using eq 2, we have

$$\frac{dV}{dx_D} = \left(\frac{\partial V}{\partial x_D}\right)_F - \frac{F_0(x_R - x_B)}{(x_D - x_R)^2}\left(\frac{\partial V}{\partial F}\right)_{x_D} \tag{4}$$

The sign property of eq 3 and inspection of eq 4 imply that operating conditions may certainly exist such that $dV/dx_D = 0$; note that x_B is normally chosen as a small value ($x_B \approx 0$), so that $x_R - x_B > 0$. As $x_D \to 1$ (which means that the distillation column begins to fill up with A),

$$\left(\frac{\partial V}{\partial x_D}\right)_F \to \infty$$

while the second term of the right-hand side (RHS) of eq 4 remains finite. This implies that $dV/dx_D > 0$, as $x_D \to 1$. On the other hand, as $x_D \to x_R$, the first term of the RHS of eq 4 remains finite (the minimum reflux condition), while the second term becomes negative infinity. This implies that $dV/dx_D < 0$. Figure 2 shows the steady-state relationship between x_D and V for the example process; indeed, dV/dx_D changes its sign and a well-defined minimum for V still exists.

2.2. Self-Optimizing Control for Minimum Energy Consumption. In terms of minimizing energy consumption for a fixed feeding condition and product purity, the optimal operation of this recycle process would be to regulate the distillation column top composition around the value that minimizes the vapor boilup, as identified in Figure 2.
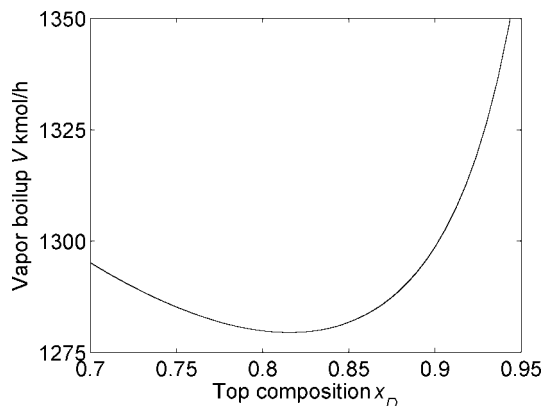
Figure 2. Steady-state relationship between x_D and V for the example process. (The example process is taken from Wu et al.[3])

Table 1. Heuristics for Plantwide Operating Policy Based on Active/Inactive Constraints for One Recycle Stream Processes with Nonbounded Chemistry[a]

| | upper limit of reactor volume, M_R^{max} | upper limit of recycle capacity, V^{max} | Reactor Temperature | |
			lower limit, T_R^{min}	upper limit, T_R^{max}
isothermal	may switch	may switch		
$E' > 1$[b]	may switch	may switch	may switch	inactive
$E' < 1$[b]	inactive	may switch	inactive	active

[a] From Ward et al.[9] [b] E' = ratio of activation energies of the undesired reaction to the desired reaction.
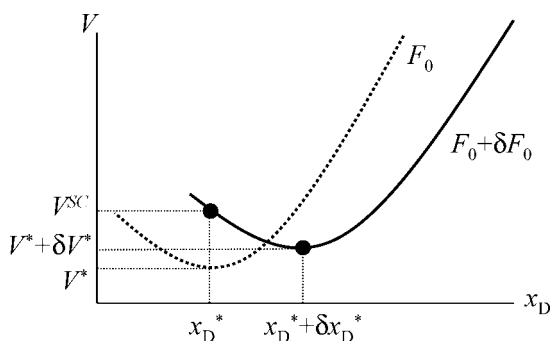

Figure 3. Definition of loss due to a change in operating conditions.

Larsson et al.[5] applied the concept of self-optimizing control[7] and found that the top composition x_D is a good candidate for a self-optimizing control variable. Keeping the distillation column top composition x_D around some constant setpoint, which may be normally selected as the value which minimizes the vapor boilup at the nominal operating condition, yields near-optimal operation for various perturbations such as changes in feed rate, kinetic constant, etc.; the loss of the vapor boilup is almost negligible. Figure 3 defines the loss where the self-optimizing control is designed for the nominal feed condition (F_0) and the setpoint for x_D is designated as x_D^*. When there is a change in the feed rate ($F_0 + \delta F_0$), the minimum shifts to $x_D^* + \delta x_D^*$, so that keeping the composition at x_D^* suffers a loss:

$$\text{Loss (\%)} = 100 \times \frac{V^{SC} - (V^* + \delta V^*)}{V^* + \delta V^*}$$

2.3. Loop Configuration of Regulatory and Self-Optimizing Control. Larsson et al.[5] suggested that ratio control such as L/D or L/F is also a good self-optimizing control configuration, which may be easier to implement than the two-point composition control; in fact, the L/F configuration is their final choice.

The D/V configuration also is possible.[11] Realization of self-optimizing control via top composition control or various ratio control setup are shown in Figure 4.

In Figure 4, the fresh feed rate F_0 is used as the direct production rate control handle. An alternative and popular method of implementation is to use the fresh feed stream to makeup for the reactor inventory.[12] Figure 5 shows the L/D ratio control configuration with the fresh feed stream as the reactor level control handle. In this case, the throughput can be controlled by manipulating the reactor effluent (F).

Note that, in all the cases, the steady-state economics are almost the same, as long as the appropriate ratio is set according to the self-optimizing control design, which minimizes the energy consumption.

The important consideration, which will be elaborated by the case studies in the following sections, is that the magnitude of the recycle flow rate is relative to the fresh feed flow rate (D/F_0) and how it affects the dynamics of the overall plant, depending on the control configurations. It has been known that a large recycle/feed ratio introduces very slow dynamics.[14,15]

In the case studies to follow, it is assumed that the L/D ratio control is used as the self-optimizing control configuration. Depending on the treatment of the fresh feed flow F_0, the control configurations are designated as C1 (see Figure 4A), which manipulates F_0 independently, and C2 (see Figure 5), which uses F_0 as the reactor level control handle. The reason for selecting the L/D configuration as the self-optimizing control is only informal and qualitative here: two-point composition control (see Figure 4B) is known to pose control problems and it requires an additional composition analyzer, whereas the L/F and D/V configurations may suffer tuning problems for the reflux drum level control, because tuning of this loop is dependent on manipulation of F or V. Formally, discrimination of these control configurations should be done more carefully from various aspects, such as evaluation of the implementation errors, dynamic responses, etc.

2.4. Throughput Control Based on Self-Optimizing Control. The advantage of the control loop configuration shown in Figure 4 is that the throughput can be directly specified by the fresh feed flow F_0. For the control loop configuration shown in Figure 5, an additional control loop that manipulates F by examining F_0 can be easily incorporated for throughput control.

When process constraints must be considered (for example, the vapor load of the distillation column) for the purpose of feed maximization, override control such as that shown in Figure 6 is readily applicable. Because the self-optimizing controller keeps the vapor load around its minimum for various disturbances, including the feed increase or decrease, the controller should simply stop increasing the feed when the vapor load hits its upper limit.

3. Recycle System with Nonbounded Chemistry

In this section, control system design for a recycle system with nonbounded chemistry is discussed. For a recycle process with nonbounded chemistry, when the recycle stream is increased, the byproduct production rate decreases. Therefore, economical tradeoff between the yield of the desired product and separation cost exists, which casues the existence of nonzero values of the optimal recycle flow rate in the unconstrained economic optimization. When the process constraints are considered, it is rather complicated to locate where the optimal operating point lies: the active constraints may switch, depending on the operating conditions. It is particularly dependent on the reactor size and recycle capacity whether the reactor holdup
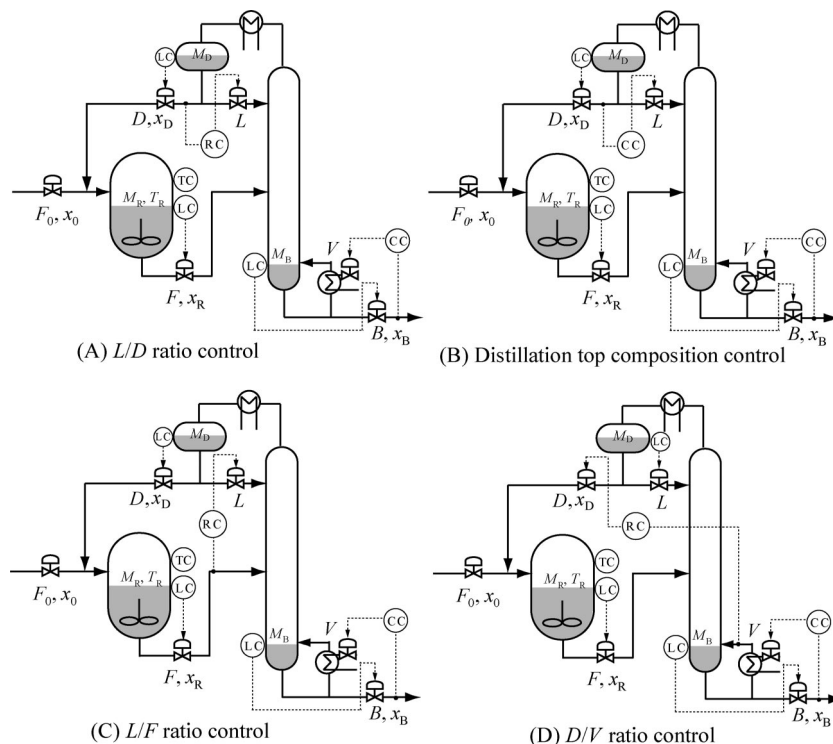
Figure 4. Examples of self-optimizing control configuration for minimum energy consumption; the fresh feed flow is an independent variable.
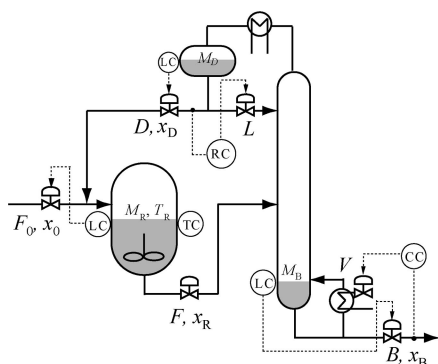

Figure 5. Schematic of an example of self-optimizing control configuration using the fresh feed rate as the reactor level control handle.
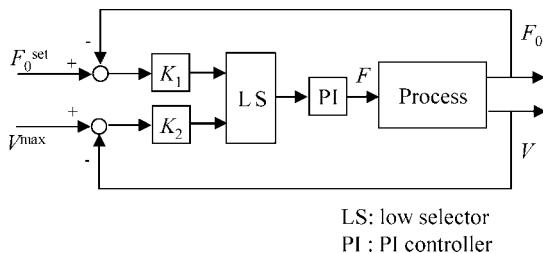

LS: low selector
PI : PI controller

Figure 6. Override controller for feed maximization. F_0^{set} is a feed rate setpoint and V^{max} is an upper limit for the distillation column load.
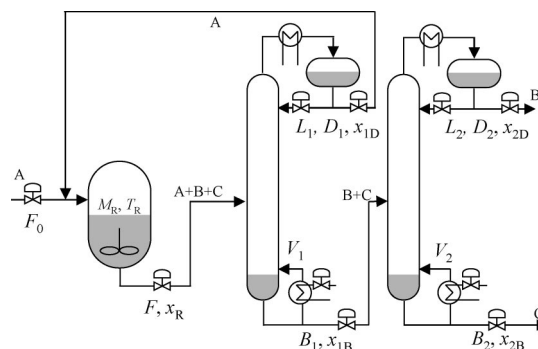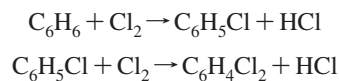

Figure 7. Reactor and two separators with one material recycle.

chlorobenzene from benzene, with monochlorobenzene as the desired product and dichlorobenzene as the undesired side product:

$$C_6H_6 + Cl_2 \rightarrow C_6H_5Cl + HCl$$

$$C_6H_5Cl + Cl_2 \rightarrow C_6H_4Cl_2 + HCl$$

These reactions are performed in a liquid-phase CSTR. Liquid benzene and chlorine gas are introduced into the CSTR, where the concentration of chlorine in the liquid phase is assumed to be determined by mass transfer and solubility limitations and is assumed to be virtually constant, unavailable as an operational degree of freedom. The chemistry then can be considered to be of the type A → B → C (where A is benzene, B monochlorobenzene, and C dichlorobenzene). Hydrogen chloride (HCl) and unreacted chlorine (Cl$_2$) can be removed from the reactor effluent stream through stripping operations, which are not considered in this paper.

The flow diagram of the reactor/separator system for the case study is shown in Figure 7. The pure species A is fed into the CSTR, where the reaction A → B → C occurs, and the reactor effluent is sent to the distillation column sequence. In the first distillation column, the reactor effluent is separated into A and

constraints become active or not. Also, the operating policy for the reactor temperature is dependent on the activation energies of the desired and undesired reactions. Table 1 summarizes the heuristics for the operating policy derived by Ward et al.[9] for processes with nonbounded chemistry when there is one recycle stream and all reactions are of the same order.

The process example considered in this section is from the work of Ward et al.,[10] which assumes the production of

Table 2a. Plant and Design Data for the Process Example with Nonbounded Chemistry

properties of species	A	B	C
Kinetic Parameters: $k_i = k_{i,0} \exp[-E_i/(RT)]^a$			
molecular weight (g/mol)	78.11	112.56	147.00
mass density (g/cm^3)	0.877	1.11	1.29
liquid heat capacity (J mol^{-1} K^{-1})	136.3	150.1	162.4
vapor heat capacity (J mol^{-1} K^{-1})	80.1	97.6	113.9
enthalpy of vaporization (kJ/mol)	30.72	35.19	39.00
Vapor Pressure: $P_i = \exp(A_i - B_i/(C_i + T))$, for i = A, B, C			
Antoine constant A_i	13.886	14.053	14.802
Antoine constant B_i	−2788.5	−3295.1	−4104.1
Antoine constant C_i	−52.4	−55.6	−43.2

^{a} $k_{1,0}$ = 774 min^{-1} and E_1 = 33.9 kJ/mol; $k_{2,0}$ = 3400 min^{-1} and E_2 = 42.7 kJ/mol.

Table 2b. Distillation Column Data and Nominal Operating Conditions

| parameter	value
Distillation Column Data	
number of stages	17
feed stage	7
Nominal Operating Conditions	
F_0	4078 kg/h (52.2 kmol/h)
T_R	333.15 K
M_R	25 m^3 (276 kmol)
F	27399 kg/h (326 kmol/h)
D	21471 kg/h (274 kmol/h)
L	6110 kg/h (78 kmol/h)
$x_{D,A}$	0.980
$x_{D,B}$	0.020
$x_{D,C}$	0
B	5928 kg/h (52.2 kmol/h)
V	323 kmol/h
$x_{B,A}$	0.005
$x_{B,B}$	0.96
$x_{B,C}$	0.035

B + C, and the unreacted A, which is the lightest component, is recycled back from the distillation column top to the reactor and the bottom product is sent to the second column. At the second column, the undesired side product C, which is the heaviest, is withdrawn from the bottom and the desired product B is recovered from the top of the column. It is assumed that the product purity $x_{2D,B}$ at the top of the second column is specified ($x_{i,j}$ denotes the composition of species j at location i).

Control system design is confined to the reactor and the first column. The plant and design data are shown in Tables 2a and 2b. A nonlinear process model that describes the dynamics of the reactor and the first distillation column has been developed, and it is used for control system design and simulations.

3.1. Self-Optimizing Control. This system has three operational degrees of freedom: the reactor temperature, the reactor holdup, and one of the manipulated variables in the distillation column. Instead of designing a control system that simultaneously considers all of the degrees of freedom, the approach used here is hierarchical and we first investigate if the self-optimizing control configuration presented in the previous section, which utilizes the one degree of freedom in the distillation column, is still applicable for this process.

Because the product purity $x_{2D,B}$ at the second column is assumed to be specified and component A is assumed to be the lightest, it is reasonable to put composition control of $x_{1B,A}$ at the bottom of the first column. With the bottom composition $x_{1B,A}$ regulated, the same argument applies to the existence of the minimum for the vapor load V_1: through the overall mass
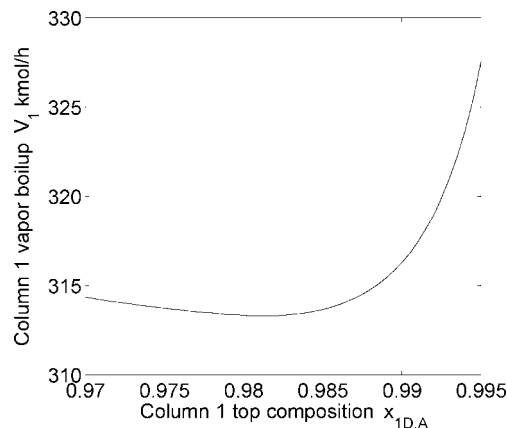
Figure 8. Steady-state relationship between $x_{1D,A}$ and V_1 for the example process.

balance for species A, if $x_{1B,A}$ is specified, $x_{R,A}$ can be determined, regardless of the column operations. If the behavior of the distillation column is similar to that of a binary column, in terms of species A and the other species, competing effects of the increased feed and the decreased separation requirement caused by increasing the recycle flow rate creates the appearance of the minimum in the vapor load. Figure 8 shows the steady-state relationship between the column 1 top composition $x_{1D,A}$ and the vapor boilup V_1; a minimum for V_1 appears at $x_{1D,A} \approx 0.982$.

Self-optimizing control for the minimum energy consumption of the first column then can be designed in the same way as that presented in the previous section. The three candidate configurations—L_1/D_1, L_1/F, and D_1/V_1—are considered and evaluated for various disturbances and operating conditions. Particularly, losses due to manipulations of the reactor holdup and temperature must be evaluated carefully, because these two variables are going to be manipulated by the upper-level optimizing controller, whereas for the process with the simple reaction A → B (considered in the previous section), they have been assumed to be constant. As a result of numerical evaluations, it has been determined that all three candidate configurations are capable of keeping the process near the minimum vapor boilup conditions for a wide range of operating conditions; for changes of ±20% in the feed rate, kinetic constant, and reactor holdup, the losses are almost negligible (<1%).

One important consideration here is the effect of the distillation column operation on the overall product yield; the yield is defined here as the number of moles of B produced divided by the number of moles of A fed to the system, so that it equals the column 1 bottom composition $x_{1B,B}$. In fact, the yield $x_{1B,B}$ has been determined to be quite insensitive to the column operation. This property can be explained as follows (although process operations are performed on the volume and mass basis in reality, the derivations to follow are those on a molar basis, for the ease of theoretical analysis).

Assuming that $x_{1D,C} = 0$ (the heaviest component C does not reach the top of column 1), mass balances for the components A and B can be written as

$$F_0 + D_1 x_{1D,A} - k_1 x_{R,A} M_R - (F_0 + D_1) x_{R,A} = 0$$

$$(F_0 + D_1) x_{R,A} - D_1 x_{1D,A} - F_0 x_{1B,A} = 0$$

$$D_1 x_{1D,B} + k_1 x_{R,A} M_R - k_2 x_{R,B} M_R - (F_0 + D_1) x_{R,B} = 0$$

$$(F_0 + D_1) x_{R,B} - D_1 x_{1D,B} - F_0 x_{1B,B} = 0$$

$$x_{1D,A} + x_{1D,B} - 1 = 0$$

Table 3. Proportional−Integral (PI) Control Parameters for the Level and Composition Controllers

controller	proportional gain	integral time (h)
reactor holdup[a]	1000 kg h^{-1} m^{-3}	5
distillation column bottom holdup	8000 kg h^{-1} m^{-3}	5
distillation column composition	100 kmol h^{-1} %$^{-1}$	2
reflux drum holdup	5000 kg h^{-1} m^{-3}	5

[a] The sign of the proportional gain is opposite between C1 and C2.

where k_1 and k_2 are the kinetic constants for the desired and undesired reactions, respectively.

The reactor concentration $x_{R,B}$ and the yield $x_{1B,B}$ then can be obtained as

$$x_{R,B} = \frac{F_0 + D_1}{F_0 + D_1 + k_2 M_R}\left[1 - \frac{F_0}{k_1 M_R}(1 - x_{1B,A})\right] \quad (5)$$

$$x_{1B,B} = 1 - x_{1B,A} - \left(\frac{k_2 M_R}{F_0}\right)x_{R,B} \quad (6)$$

From eq 5, the sensitivity of the reactor composition $x_{R,B}$ to changes in the recycle/feed flow ratio (D_1/F_0), under fixed feed conditions, can be calculated as

$$S = \frac{\partial x_{R,B}/x_{R,B}}{\partial (D_1/F_0)/(D_1/F_0)} = -\frac{Da_2}{Da_2 + 1 + (D_1/F_0)}\left[\frac{D_1/F_0}{1 + (D_1/F_0)}\right] \quad (7)$$

where $Da_2 = k_1 M_R/F_0$. In the example process, the sensitivity is very small ($S \approx 0.04$) under the nominal operating conditions with $D_1/F_0 \approx 5$, $Da_2 \approx 0.3$. This property may hold in general cases where D_1/F_0 is large and Da_2 is relatively small; for recycle processes with nonbounded chemistry, the recycle/feed flow ratio D_1/F_0 has a tendency to be large, because an increased recycle flow is favored for higher selectivity. Then, together with eq 6, the yield $x_{1B,B}$ is insensitive to manipulation of D_1.

Consequently, the self-optimizing control, which uses the one degree of freedom in the distillation column to minimize the vapor boilup, can be safely used as the basis for the higher-level optimizing controller that takes care of the remaining two degrees of freedom.

3.2. Implementation of Self-Optimizing Control. Proportional−integral (PI) controllers are used for the level and composition control, whose control parameters are determined considering the dynamics of each *individual isolated* unit, and their parameters are shown in Table 3: the control parameters are the same for the two configurations C1 and C2 (C1 and C2 are defined as shown in Figures 4A and 5, respectively).

Figures 9 and 10 compare the step responses of the two control configurations C1 and C2. In Figure 9, the fresh feed flow rate for C1 is changed from 4078 kg/h to 4266 kg/h, while the reactor effluent flow rate for C2 is changed from 29601 kg/h to 38500 kg/h; both of the changes yield a 5% throughput increase and provide the same steady-state conditions. In Figure 10, the reactor temperature is changed stepwise from 333.15 to 334.15 K. In this case, the steady-state conditions are different for the two configurations.

The response of configuration C1 is sluggish, which is very different from that expected by the dynamics of the isolated units. This can be attributed to the effects of the large recycle/feed ratio ($D_1/F_0 \approx 5$); the large recycle stream acts as positive feedback and introduces very slow dynamics.[14,15] It is possible to tune the controllers of configuration C1 to produce responses comparable with those of configuration C2, but it requires extremely tight PI parameter settings. Consideration for measurement noises and unmodeled time delays would make them impractical.
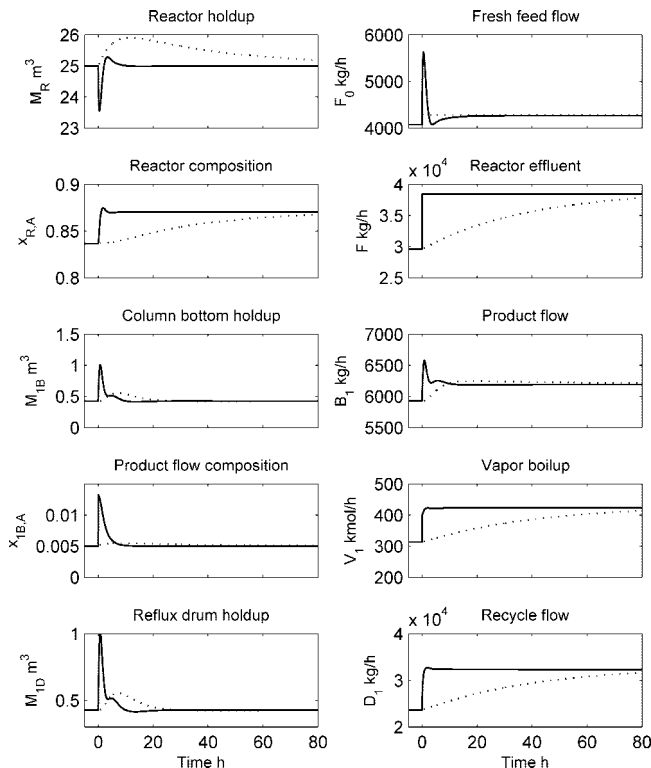

Figure 9. Step responses to a 5% increase in feed: ($\cdots$) control configuration C1 and (—) control configuration C2.
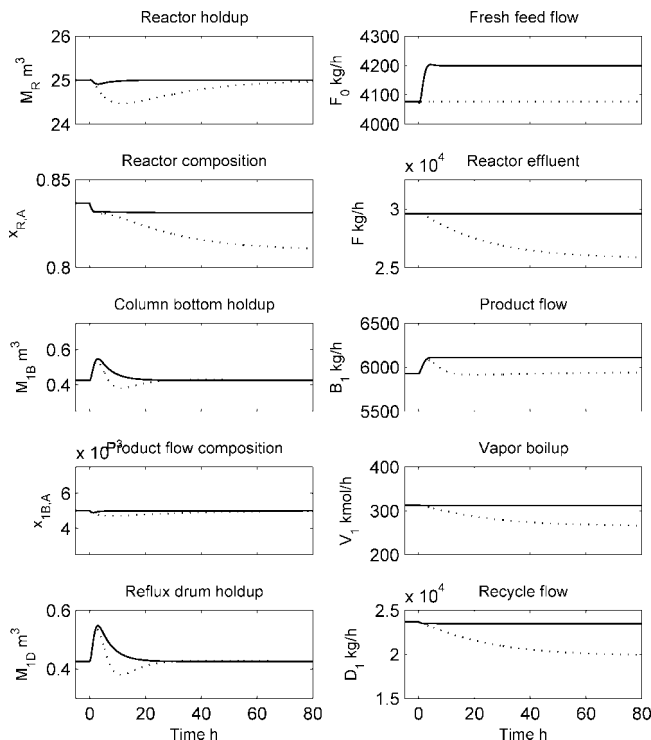

Figure 10. Step responses to a 1 K increase in reactor temperature: ($\cdots$) control configuration C1 and (—) control configuration C2.

On the other hand, with configuration C2, such positive feedback is severed because one of the flows in the recycle path—that is, the reactor effluent flow—is placed under flow control. Luyben's rule[2] applies here, with configuration C2 being clearly more advantageous, with respect to the ease of controller tuning and settling time.
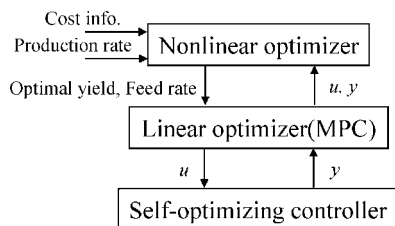
Figure 11. Optimizing control structure for recycle processes with non-bounded chemistry.

The problem with configuration C2 is that the flow rate of the reactor effluent stream (F) is large, compared to the fresh feed flow (F_0) and the product flow (B_1), which are used for inventory control of the reactor and distillation column bottom, so that manipulation of F easily causes large fluctuations in these streams, as can be seen from the large spikes in the initial responses of Figure 9. To prevent such large fluctuations, the bandwidth of the reactor effluent flow manipulation should be limited. Large fluctuations may be still expected, if magnitudes of disturbances in the large recycle flows such as F, V_1, and D_1 are multiplicative. In worst cases, level control capability may be lost by flow rate limitations. Particularly with the reactor level control, because its performance affects the product quality through the reaction residence time fluctuation, its performance should be maintained reasonably well. For this purpose, installation of an additional buffer tank for the introduction of fresh feed[12] may become necessary, although increased holdup of a hazardous material may not be preferred, from a process safety point of view.[13]

3.3. Supervisory Optimizing Control. Using the self-optimizing control configuration C2 as the basis, an optimizing controller is designed that seeks economically optimal operations by manipulating the reactor holdup and temperature. Throughput control capability is also incorporated.

Because of the complexities in the constrained optimization landscape for processes with nonbounded chemistry, Ward et al.[10] suggested using a supervisory optimizing controller such as a real-time optimizer (RTO): they implemented the operating policy for the reactor volume and temperature obtained for a given feed rate by solving nonlinear optimization problems off-line.

Here, the optimization layer is further divided into two sublayers: the upper sublayer, which determines the feed rate and the corresponding economically optimal yield, and the lower sublayer, which regulates the process around the specified yield and minimizes energy consumption; the nature of the economical optimization can be cast as a multiobjective optimization problem in which the optimal tradeoff between the yield and separation cost is sought, so that the lower sublayer realizes the Pareto optimal operation.

More specifically, the setpoint to the first column bottom composition $x_{1B,B}$ is calculated and the feed rate F_0 is determined at the upper level, and the lower level optimizing controller realizes the specified yield and minimizes the column vapor load V_1 by manipulating the reactor temperature and holdup while respecting process constraints.

The reason why the optimization layer is divided is that, even without the upper sublayer, which requires a nonlinear process model and nonlinear optimization and may be sometimes difficult to maintain, the lower sublayer controller alone could act independently, as long as a reasonable (not necessarily optimal) yield setpoint is given. The lower sublayer optimizing controller can be realized using conventional linear MPC that

can be invoked at a shorter control interval, so that better disturbance rejection could be expected.

The following discussion focuses only on the lower sublayer optimizing controller; particularly, a steady-state target optimizer, based on the LP (linear program), is designed. The resulting hierarchical control structure is shown in Figure 11.

The controlled variable (y) and manipulated variable (u) are defined as

$$y = (F_0 \ x_{1B,B} \ V_1)^T$$

and

$$u = (F \ M_R \ T_R)^T$$

where the manipulated variable u is constructed by the setpoint to the regulatory PI controllers. The linear steady-state model is described as

$$G = \frac{\partial y}{\partial u}$$

where G is the steady-state gain matrix, which can be obtained, for example, by step testing.

The future uncontrolled steady-state responses of the controlled variable and the current manipulated variable can be described as

$$\bar{y} = (\bar{F}_0 \ \bar{x}_{1B,B} \ \bar{V}_1)^T \tag{8a}$$

and

$$\bar{u} = (\bar{F} \ \bar{M}_R \ \bar{T}_R)^T \tag{8b}$$

and the manipulated variable move Δu is given as

$$\Delta u = (\Delta F \Delta M_R \ \Delta T_R)^T$$

The linear program can be posed as follows:

$$\min_{\Delta_{u,\,s_i}} J = \Delta V_1 + M_1 s_1 + M_2 s_2 \tag{9}$$

subject to

$$(\Delta F_0 \ \Delta x_{1B,B} \ \Delta V_1)^T = G\Delta u$$
$$F_0^{set} - s_1 \le \bar{F}_0 + \Delta F_0 \le F_0^{set} + s_1$$
$$x_{1B,B}^{set} - s_2 \le \bar{x}_{1B,B} + \Delta x_{1B,B}$$
$$V_1^{min} \le \bar{V}_1 + \Delta V_1 \le V_1^{max} \tag{10}$$
$$M_R^{min} \le \bar{M}_R + \Delta M_R \le M_R^{max}$$
$$T_R^{min} \le \bar{T}_R + \Delta T_R \le T_R^{max}$$
$$0 \le s_1, s_2$$

where $x_{1B,B}^{set}$ and F_0^{set} are the setpoints of the yield and the feed rate, respectively, which are passed from the upper sublayer; $(\cdot)^{min}$ and $(\cdot)^{max}$ are the lower and upper limits of the manipulated/controlled variables, respectively; s_i ($i = 1, 2$) is a feasibility slack; and M_i is a large positive weight on the slack variables. Other practical considerations, such as constraints on the manipulated variable move Δu, may be readily incorporated.

In formulating the LP, the constraint for the yield $x_{1B,B}$ is posed as the inequality described by eq 10 with the setpoint $x_{1B,B}^{set}$ as the lower bound. The controller tries to decrease the vapor boilup as long as the yield constraint is not violated. Basically, $x_{1B,B}$ and V_1 are in the tradeoff relationship: improvement of the yield results in greater energy consumption. When the controller is unable to meet the constraints, it eases the constraints for the setpoint F_0^{set} or the lower bound of $x_{1B,B}$. Which of the slack variables becomes active first is dependent on the weights M_i; in this case study, M_i is chosen so that the
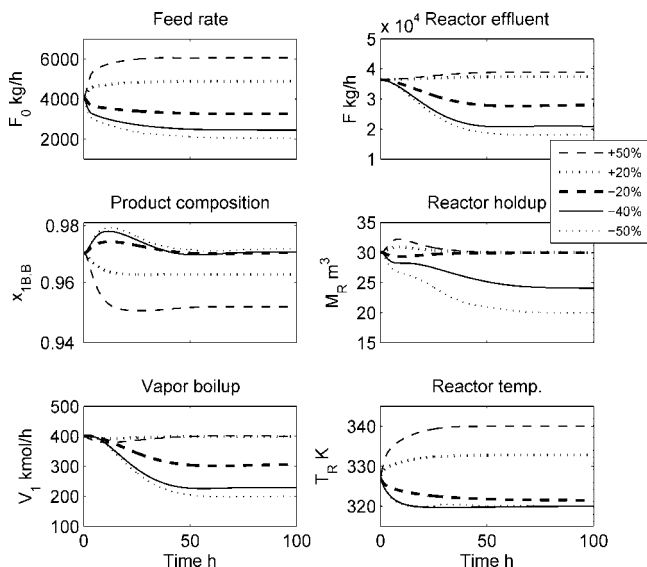
Figure 12. Simulation results: dynamic responses for feed rate changes.

Table 4. Upper and Lower Limits and Setpoints Used for Simulations

parameter	value
vapor boilup	
upper limit, V_1^{max}	400 kmol/h
lower limit, V_1^{min}	200 kmol/h
reactor holdup	
upper limit, M_R^{max}	30 m^3
lower limit, M_R^{min}	20 m^3
reactor temperature	
upper limit, T_R^{max}	340 K
lower limit, T_R^{min}	320 K
bottom composition setpoint	
$x_{1B,A}$	0.005 mole fraction
$x_{1B,B}$	0.97 mole fraction

lower bound for $x_{1B,B}$ may be eased first. Detailed controller behavior will be described in the next section.

3.4. Simulation Study. The optimizing controller discussed so far is implemented for simulation studies. For the supervisory MPC, the linear process model is identified through step responses for each input/output pair around the nominal operating condition, which are then approximated by a first-order lag. Model update and future prediction are done simply by putting output bias, which is updated at each control sampling time from the prediction error. The steady-state target $\bar{u} + \Delta u$ obtained as the LP solution is implemented through LQR (linear quadratic regulator) in the receding horizon fashion. Movement of the reactor effluent F is heavily weighted to prevent excessive response of the fresh feed F_0. Detailed algorithm is given in the Appendix. The control sampling time is selected as 5 min.

Initially, the process is assumed to be at rest, with the reactor holdup and the vapor boilup at their maximum for the nominal feed rate $\tilde{F}_0$. Stepwise changes in the feed setpoint (+50%, +20%, −20%, −40%, −50%) then are given, while the yield setpoint is held constant at $x_{1B,B}^{set} = 0.97$. Other simulation parameters are given in Table 4, and the dynamic responses are shown in Figure 12. Figure 13 and Table 5 summarize the steady-state operating points provided by the designed controller.

The 20% feed increase is achieved by increasing the temperature and the reactor effluent; the reactor effluent increase should be marginal, because the vapor boilup is already at its maximum. At this throughput, the yield constraint cannot be met so that $x_{1B,B} < x_{1B,B}^{set}$ at steady states. For further feed increases, the controller increases the temperature until it reaches

the upper limit. In the +50% feed increase case, the temperature constraint is met before the feed reaches its setpoint.

The 20% feed decrease is achieved by decreasing the temperature. At the same time, the reactor effluent is decreased to reduce the vapor boilup. At this production rate, the yield constraint can be met. For further feed decreases, the controller starts to decrease the reactor holdup after the temperature reaches its lower limit (−40% case). When the feed is decreased even further to −50%, the reactor holdup and the vapor boilup attain their lower limits before the feed setpoint is reached.

In all the cases, the controller keeps the process on the Pareto optimal front, as shown in Figure 13. Note that when the lower limit of the vapor boilup becomes active, the controller loses its optimality, because the self-optimizing control has been designed for minimization of the vapor boilup.

4. Recycle System with Bounded Chemistry

For a recycle process with bounded chemistry, when the recycle stream is increased, the byproduct production rate increases. The optimal operation policy for a prescribed production rate of the desired species is to minimize the recycle flow rate and maximize the per-pass conversion by setting the reactor holdup at its maximum. In most cases, the reactor temperature also should be kept at its maximum, although there is an exception for the case where the kinetic energy of the undesired reaction is far greater than that of the desired reaction.[9,10] There is no tradeoff between the yield and separation cost, and the control system design approach is the same as in the case for the simple reaction A → B.

Here, we have investigated whether the self-optimizing control configuration is also applicable, using an example process in which the chemistry A → B, A + A → C (where B is the desired product and C is the undesired product) is considered.

The same process configuration as that shown in Figure 7 is used. The example process is hypothetical, and its data are shown in Tables 6a and 6b. The reaction kinetics is assumed to be

$$\text{Desired:} \quad r_1 = k_1 x_{R,A}, \quad k_1 = k_{10} \exp\left(-\frac{E_1}{RT_R}\right) \quad (11)$$

$$\text{Undesired:} \quad r_2 = k_2 x_{R,A}^2 \quad k_2 = k_{20} \exp\left(-\frac{E_2}{RT_R}\right) \quad (12)$$

where the reaction rates r_1 and r_2 are the rates of generation of the moles of the desired and undesired products, respectively. As stated previously, it is assumed that the product purity $x_{2D,B}$ at the top of the second distillation column is specified and the first column bottom composition $x_{1B,A}$ should be regulated to keep the product purity specification. Detailed analysis on the steady-state optimal operation regarding this process is given in the Appendix.

4.1. Self-Optimizing Control. Through the mass balance equations, the concentration of A in the reactor ($x_{R,A}$) is given as the solution to the following equation:

$$(x_{1B,A} - 2)k_2 M_R x_{R,A}^2 - k_1 M_R x_{R,A} + F_0(1 - x_{1B,A}) = 0 \quad (13)$$

which implies that $x_{R,A}$ is independent of the distillation column operation, such as manipulation of D_1 or regulation of $x_{1D,A}$, as long as $x_{1B,A}$ is regulated. The distillation column top composition $x_{1D,A}$ can be expressed as

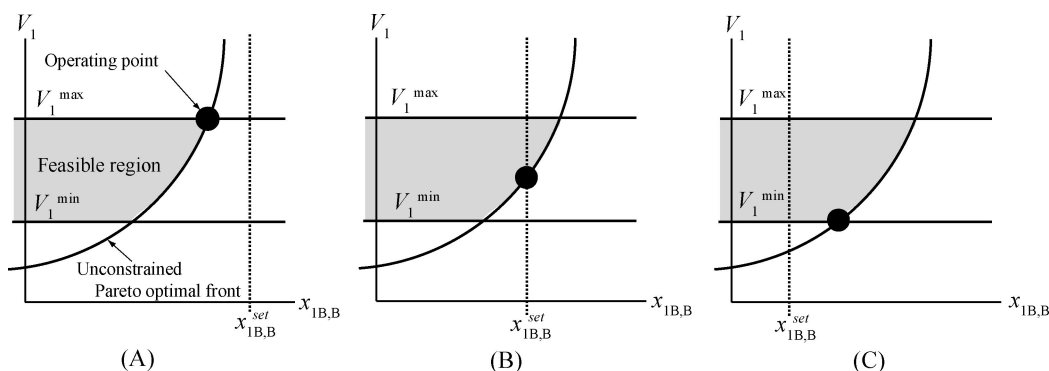$$x_{1D,A} = x_{R,A} + \frac{F_0 - k_2 x_{R,A}^2 M_R}{D_1}(x_{R,A} - x_{1B,A})$$

Figure 13. Optimization landscape for the supervisory optimizing control.

Table 5. Steady-State Operating Conditions Obtained by the Supervisory Optimizing Controller

feed setpoint[a]	active constraints	operating point[b]
$F_0 = 0.5\tilde{F}_0$	T_R^{min}, M_R^{min}, V_1^{min}	C
$F_0 = 0.6\tilde{F}_0$	T_R^{min}, F_0, $x_{1B,B}^{set}$	B
$F_0 = 0.8\tilde{F}_0$	M_R^{max}, F_0, $x_{1B,B}^{set}$	B
$F_0 = 1.0\tilde{F}_0$	M_R^{max}, F_0, V_1^{max}	A
$F_0 = 1.2\tilde{F}_0$	M_R^{max}, F_0, V_1^{max}	A
$F_0 = 1.5\tilde{F}_0$	T_R^{max}, M_R^{max}, V_1^{max}	A

[a] $\tilde{F}_0$ denotes the nominal feed rate. [b] Here, A, B, and C correspond to the operating conditions depicted in Figure 13.

Table 6a. Plant and Design Data for the Process Example with Bounded Chemistry

| properties of species	A	B	C
Kinetic Parameters: $k_i = k_{i,0} \exp[-E_i/(RT)]^a$			
molecular weight (g/mol)	78.11	112.56	156.22
mass density (g/cm³)	0.877	1.11	1.29
liquid heat capacity (J mol⁻¹ K⁻¹)	136.3	150.1	162.4
vapor heat capacity (J mol⁻¹ K⁻¹)	80.1	97.6	113.9
enthalpy of vaporization (kJ/mol)	30.72	35.19	39.00
Vapor Pressure: $P_i = \exp[A_i - B_i/(C_i + T)]$, for i = A, B, C			
Antoine constant A_i	13.886	14.053	14.802
Antoine constant B_i	−2788.5	−3295.1	−4104.1
Antoine constant C_i	−52.4	−55.6	−43.2

[a] $k_{1,0}$ = 360 min⁻¹ and E_1 = 30 kJ/mol; $k_{2,0}$ = 75 (kmol/m³)⁻¹ min⁻¹ and E_2 = 40 kJ/mol.

Table 6b. Distillation Column Data for the Process Example with Bounded Chemistry

parameter	value
number of stages	16
feed stage	9
nominal operating conditions	
F_0	4062 kg/h (52 kmol/h)
T_R	343.15 K
M_R	25 m³ (256 kmol)
F	7827 kg/h (76.1 kmol/h)
D	2055 kg/h (25.1 kmol/h)
L	1917 kg/h (23.4 kmol/h)
$x_{D,A}$	0.97
$x_{D,B}$	0.03
$x_{D,C}$	0
B	5772 kg/h (51.0 kmol/h)
V	59.1 kmol/h
$x_{B,A}$	0.005
$x_{B,B}$	0.975
$x_{B,C}$	0.02

which is a decreasing function of D_1. Following the same argument as in the previous sections, the appearance of a minimum in V_1 then can be expected. Figure 14 shows the steady-state relationship between $x_{1D,A}$ and V_1 for the example process.
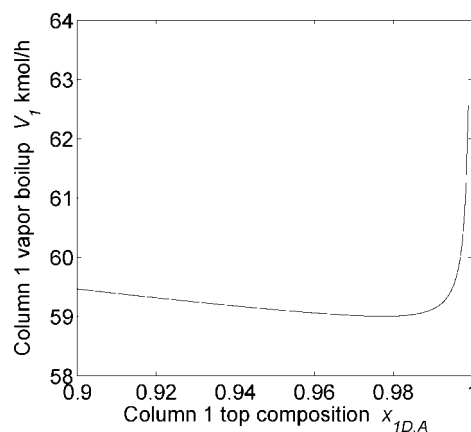
Figure 14. Steady-state relationship between $x_{1D,A}$ and V_1 for the example process.

Assuming that the heaviest species C does not reach the distillation column top, the concentrations of the undesired product C in the reactor ($x_{R,C}$) and at the distillation column bottom ($x_{1B,C}$) are expressed as

$$x_{R,C} = \frac{k_2 M_R x_{R,A}^2}{F_0 + D_1 - k_2 M_R x_{R,A}^2} \tag{14}$$

$$x_{1B,C} = \frac{k_2 M_R x_{R,A}^2}{F_0 - k_2 M_R x_{R,A}^2} \tag{15}$$

which can be easily derived from the overall mole balance. Equations 13 and 15 imply that the overall yield of the desired product B from the fresh feed A, which is defined as the number of moles of B produced divided by the number of moles of A fed, is independent of the column operation D_1. Therefore, the self-optimizing control also can be safely applied as the basis of a supervisory optimizing controller in this case.

4.2. Implementation of Self-Optimizing Control and Throughput Control. Characteristics of recycle processes with bounded chemistry is that a smaller recycle flow rate is advantageous, in terms of undesired species production. When a process is designed with a small recycle/feed flow ratio (D_1/F_0), control configuration C1 can be safely applied, because changes in the dynamics that are due to the recycle flow may be neglected.

For configuration C2, an additional loop is configured between the reactor effluent F and the fresh feed flow F_0, as shown in Figure 6, to specify the production rate explicitly.

In the simulation study, the same PI control parameters as those listed in Table 3 are used for both configurations. For the
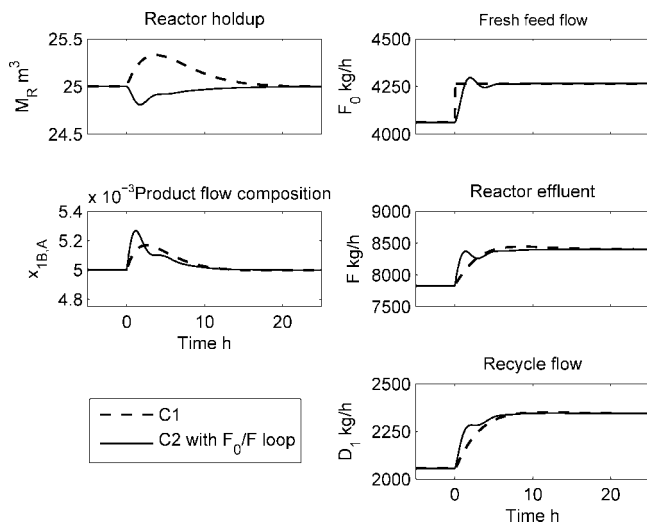
Figure 15. Step responses to a +5% increase in feed around the nominal feed conditions: (- - -) control configuration C1 and (—) control configuration C2.

Figure 16. Step responses to a +5% increase in feed around the higher feed rate (+40%): (- - -) control configuration C1, (—) control configuration C2, and (· · ·) configuration C2 with a retuned F_0/F loop.

F_0/F loop in configuration C2, an integral control with the integral gain 3.6 h^{-1} is applied.

Figure 15 compares step responses to a +5% increase in the throughput around the nominal feed conditions. For configuration C1, the fresh feed flow F_0 is directly manipulated, whereas the setpoint to F_0 is changed stepwise for configuration C2. Because the recycle/feed ratio is small ($D_1/F_0 \approx 0.5$), the dynamic responses are comparable. Although not shown, step responses to the same amount of feed increase around the increased feed condition (+40% of the nominal feed) are also comparable. In this case, the recycle/feed ratio is still small ($D_1/F_0 \approx 0.9$).

The aforementioned heuristics can be applied only when the recycle/feed ratio is kept small throughout the operation range. For this purpose, the Damköhler number (which is defined as $Da = k_1 M_R/F_0$) must be sufficiently large: $Da > 3$ is one criterion.[12] Otherwise, the "snowball" effect has a tendency to occur with the control configuration, which keeps the reactor holdup and temperature constant: the recycle flow rate grows exponentially with increases in the feed rate.[2]

Although a large Da value is advantageous for recycle processes with bounded chemistry, in terms of minimizing the production rate of undesired products, for some reason, the reactor may be designed with a smaller Da value (a smaller maximum holdup and a lower maximum temperature).

Let us assume that the process should be operated at a smaller holdup ($M_R = 20$ m^3) and a lower temperature ($T_R = 330$ K), with $Da \approx 1.5$; in the aforementioned case study, the holdup and temperature are $M_R = 25$ m^3 and $T_R = 330$ K, with $Da \approx 3$.

With the smaller Da value, the recycle feed/ratio is $D_1/F_0 = 1.4$ at a nominal feed rate of $F_0 = 4062$ kg/h (52 kmol/h), whereas it grows to $D_1/F_0 = 3.5$ for the +40% feed rate. At the nominal feed rate, both configurations C1 and C2 still work, providing responses similar to those shown in Figure 15 with the same control parameters. However, at the +40% feed rate, both of the responses with configurations C1 and C2 become slower: the step responses to a feed change around the increased feed rate are shown in Figure 16 With configuration C1, the slowdown in the dynamic responses can be attributed to the enhanced positive feedback effect by the larger recycle flow, whereas the nonlinearity between F_0 and F may be the major cause for configuration C2. Extensive retuning of the PI
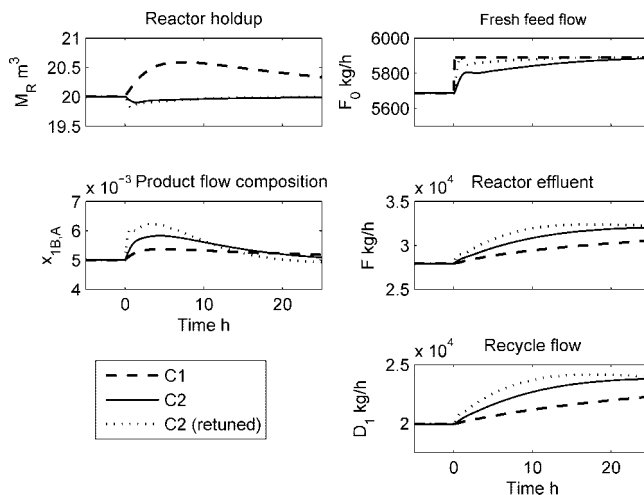
controllers would be needed with configuration C1 for improved response, while retuning of the F_0/F loop would be helpful to some extent for configuration C2; the integral gain for the F_0/F loop is made four times as large in the simulation result shown in Figure 16. For better performance, nonlinear control techniques such as gain scheduling are readily applicable for configuration C2 to compensate for the nonlinearity between F_0 and F.

In practice, such a large change in the recycle flow (250% increase) would be highly unlikely; distillation columns could not be designed for such a wide operation range. Hence, manipulation of the reactor holdup and temperature must be considered for processes with a small Da value, to alleviate the snowball effect.[2,3,6]

However, manipulation of the reactor holdup and temperature for bounded chemistry is necessarily accompanied by economical losses: increased separation cost and increased production of undesired species at lower production rates. Note that the operation with the maximum reactor holdup and temperature minimizes the recycle flow rate and undesired product. The control system design problem for recycle processes with bounded chemistry whose Da value is small then should focus on manipulation of the reactor holdup and temperature, to respect the lower limit of the vapor boilup at a smaller throughput, while minimizing the vapor boilup by maximizing the reactor holdup and temperature at a larger throughput. When the lower limit of the vapor boilup becomes an issue, the control system design approach discussed so far, based on the self-optimizing control, is no longer valid, because the self-optimizing control is designed for *minimization* of the vapor boilup.

5. Discussion

5.1. Applicability of Self-Optimizing Control. As a prerequisite for the self-optimizing controller design, regulation of the distillation column bottom composition $x_{B,A}$ by the vapor boilup is assumed, which originally comes from the requirement to control the product purity. In fact, regulation of the bottom composition can be regarded as equivalent to specifying the overall conversion of feed species A, because of the global mass balance imposed on species A. With the process examples considered, the consumption rate of A is not influenced by the concentrations of the other species, so that the conversion

specification automatically determines the reactor concentration of A for fixed reactor holdup and temperature, independent of the column top composition $x_{D,A}$.

In fact, these properties simplify the arguments for the existence of the unconstrained minimum in the vapor boilup, namely, the derivation of eq 4, in which the column feed concentration $x_{R,A}$ can be assumed to be constant. For systems in which the recycled species is the heaviest and recycled from the column bottom, the same argument can be applied, and an unconstrained minimum certainly exists.

However, there are more-general cases where this assumption does not hold: for example, (a) the order of the volatilities of the species is different, hence, the different distillation column sequence and more than one distillation columns are involved, or (b) the consumption rate of species A is not dependent only on the concentration of A.

For case (a), in which more than one columns are involved, the operational degree of freedom increases so that more-extensive analysis may be required. Furthermore, if the volatility of recycle species A is between the desired and undesired products—that is, (B > A > C) or (C > A > B)—species A may leave the system from two locations, as shown in Figure 17; then, composition control of A at one place alone cannot specify the overall conversion of A.

For case (b), miscellaneous types of chemistry can be considered; the chemistry of the type A → B, A + B → C is such an example. In this case, it should be evaluated numerically regardless of whether the unconstrained minimum appears. Moreover, depending on the type of process chemistry, a change in the recycle flow rate may have a large effect on the byproduct production rate, although it has negligible effects in the given process examples. If they are large, the hierarchical control system design approach that has been used cannot be applied; the three operational degrees of freedom should be handled simultaneously.

An important property for the self-optimizing control configuration to be valid as the basis for the supervisory optimizing control is that the losses due to the reactor operations (manipulation of the reactor holdup and temperature) should be negligible, especially for processes with nonbounded chemistry. In other words, the unconstrained minimum should be flat.[7] In this regard, eq 4 implies the following. If x_D^* is close to 1 (x_D^* is depicted in Figure 3), it can be expected that $(\partial^2 V/\partial x_D^2)_F$ becomes very large, so that the minimum may be sharp. Also, if x_D^* is close to x_R, the second term in eq 4 renders a sharp minimum. Therefore, x_D^* should be located away from 1 and x_R, to make a flat minimum and ensure the existence of self-optimizing control; a distillation column that is designed for unnecessarily high-purity separation would fail to satisfy this condition.

5.2. Regulatory Loop Configurations. As the regulatory control loop configuration, two candidates have been studied, in terms of the reactor level controller handle: configuration C1 (Figure 4A) uses the reactor effluent flow, whereas configuration C2 (Figure 5) uses the fresh feed flow. It has been known that configuration C1 is only applicable to processes with relatively large per-pass conversions, whereas configuration C2 works for both small and large per-pass conversions.[12] Configuration C1, with the reactor holdup and temperature being held constant, suffers from the snowball effect when the per-pass conversion is small, such that a balanced control structure that manipulates the reactor holdup and temperature has been proposed.[2,4,6]

In terms of the process chemistries, the process should be designed and operated for large per-pass conversions when the chemistry is bounded, to minimize the production of undesired species, whereas the process should be designed and operated for small per-pass conversions when the chemistry is non-bounded. In this regard, configuration C1 may be applicable only to processes with bounded chemistry, whereas configuration C2 is applicable to both of the chemistries.

When the recycle/feed flow ratio is large, considerable difficulties would be expected in controller tuning with configuration C1, because the positive feedback effect of the recycle flow significantly changes the overall dynamics and introduces very slow responses; the controller setting that is based on the isolated units does not provide satisfactory performance in the plantwide context and extensive retuning would be necessary. Also, configuration C1 may encounter a robustness problem when the snowball effect occurs, because its performance is very sensitive to the recycle/feed flow ratio.

Although configuration C1 has the advantage of being capable of directly specifying the throughput, the conclusion to be drawn here is that configuration C2 is easier and safer to apply, with respect to the ease of controller tuning and the settling time.

5.3. Supervisory Optimizing Control. As already noted Ward et al.,[10] in terms of steady-state economics, a sophisticated supervisory optimizing control may be needed only for processes with nonbounded chemistry. For processes with bounded chemistry, because the steady-state optimal operation usually lies on the process constraints, a simple override control on top of the self-optimizing control would suffice, for the purpose of feed maximization, and there would be little justification to introduce MPC or RTO; they would become necessary for processes with added complexities in which several process constraints must be considered simultaneously.

The nature of the economical optimization for processes with nonbounded chemistry is to determine an optimal tradeoff between the yield and the separation cost. In many cases, the economics of the yield dominates and the optimal operating point lies at the highest possible recycle flow rate. This is mostly true when the throughput is high, but for a lower throughput, the economics of the separation cost sometimes begins to dominate: in the benzene chlorination case study, Ward et al.[8] has shown, using reasonable cost data, that the economical optimum at a lower throughput may become unconstrained.

Formally, *online* usage of a nonlinear process model would be required to identify the optimal tradeoff, because it is dependent on the price of the raw material, as well as the product and energy costs, which may change occasionally, and the relationship between these two variables is nonlinear. However, the quantitative relation between the yield and the separation cost could be available from plant operating data: by changing the yield setpoint occasionally, the resulting vapor load would be recorded and a simple quantitative relation could be obtained empirically. The online usage of a nonlinear process model then could be avoided. In this respect, subdivision of the optimizing layer could be considered as a practical alternative to a nonlinear-optimization-based RTO.

6. Conclusion

Through the case studies, it has been shown that the simple reflux ratio control, which has been known to serve as self-optimizing control for the recycle process with the elementary reaction A → B, is also applicable to processes with bounded chemistry and nonbounded chemistry. The origin of appearance of the unconstrained minimum for the distillation column energy consumption has been identified, and applicability of the self-

optimizing control configuration for more-general cases has been also discussed.

Appropriate regulatory loop configurations that realize the self-optimizing control have been identified, with a particular focus on the effect of the recycle flow on the dynamics of the entire process.

Using the self-optimizing control as the basis, control systems that seek economically optimal operations have been constructed. Particularly, for the example process with nonbounded chemistry, a linear model predictive controller design example has been shown, with simulation results, as a practical alternative to a real-time optimizer, which resorts to nonlinear optimization.

Appendix

(A) Model Predictive Control Algorithm Used for the Simulation Study. A discrete-time version of the linear state space model is described by

$$x(n+1) = Ax(n) + Bu(n)$$
$$y(n) = Cx(n) + d(n)$$

where $x(n)$ is the state, $u(n)$ the input, $y(n)$ the measurement, and $d(n)$ the output bias.

The state $x(n)$ and the output bias $d(n)$ are updated at every control sampling time, using $u(n)$ and $y(n)$. The uncontrolled steady-state response $\bar{y}$ in eqs 8a and 8b can be obtained as

$$\bar{y} = C(I-A)^{-1}B\bar{u} + d(n), \quad \bar{u} = u(n)$$

and they are passed to the linear programming (LP) optimizer.

Following the conventions with industrial model predictive control (MPC), the process model is augmented with an integrator at its input:

$$\begin{bmatrix} x(n+1) \\ u(n+1) \end{bmatrix} = \begin{bmatrix} A & B \\ 0 & I \end{bmatrix}\begin{bmatrix} x(n) \\ u(n) \end{bmatrix} + \begin{bmatrix} B \\ I \end{bmatrix}v(n) \quad (A1)$$

$$\begin{bmatrix} y(n) \\ u(n) \end{bmatrix} = \begin{bmatrix} C & 0 \\ 0 & I \end{bmatrix}\begin{bmatrix} x(n) \\ u(n) \end{bmatrix} + \begin{bmatrix} I \\ 0 \end{bmatrix}d(n) \quad (A2)$$

where $v(n)$ is defined as the manipulated variable increment:

$$v(n) = u(n+1) - u(n)$$

For the augmented process (eqs A1 and A2), an infinite time horizon linear quadratic regulator, which minimizes the performance index J, is designed:

$$J = \sum_{n=0}^{\infty}(\tilde{y}^T(n)Q\tilde{y}(n) + v^T(n)Rv(n))$$

where $\tilde{y}(n) = [y(n)^T \ u(n)^T]^T$, and Q and R are the weight matrices. By solving the Riccati equation, the state feedback

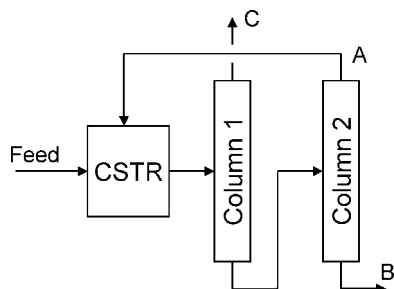$$v(n) = -K\begin{bmatrix} x(n) \\ u(n) \end{bmatrix}$$

Figure 17. Possible distillation column sequence for the volatility order (C > A > B).

can be obtained. Note that the state feedback gain matrix K is calculated off-line.

Denoting the steady-state target for the states and inputs passed from the LP optimizer as $\hat{x}(n)$ and $\hat{u}(n)$, respectively, the control input increment $v(n)$ can be calculated as

$$v(n) = -K\begin{bmatrix} x(n) - \hat{x}(n) \\ u(n) - \hat{u}(n) \end{bmatrix}$$

(B) Optimal Operation of the Process with the Chemistry A → B, A + A → C. It is investigated here whether the process chemistry A → B, A + A → C with the reaction kinetics that is described by eqs 11 and 12 is categorized as bounded. Assuming an ideal separation where the recycle stream contains pure A and assuming that all the unreacted A is recycled, the mass balance constraints yield

$$P_B = k_1 M_R x_{R,A} \quad (B1a)$$

$$P_C = k_2 M_R x_{R,A}^2 \quad (B1b)$$

$$D = \frac{x_{R,A}}{1 - x_{R,A}}(F_0 - P_C) \quad (B1c)$$

where P_B and P_C are the production rates of the desired and undesired species, respectively, and D is the recycle flow rate. For simplicity, the derivation is on a molar basis. Note that the term P_C is introduced in eqs B1a−B1c), because the reaction orders are different in the desired and undesired reactions.

Let us assume a prescribed value of the production rate of the desired product as $P_B = \bar{P}_B$. We then have

$$x_{R,A} = \frac{\bar{P}_B}{k_1/M_R}$$

$$P_C = \left(\frac{k_2}{k_1^2 M_R}\right)\bar{P}_B^2 = \frac{k_{20}}{k_{10}^2}\exp\left(\frac{2E_1 - E_2}{RT_R}\right)\left(\frac{1}{M_R}\right)\bar{P}_B^2 \quad (B2)$$

$$D = \frac{\bar{P}_B}{k_1 M_R - \bar{P}_B}(F_0 - P_C) \quad (B3)$$

Equation B2 implies that, in terms of the operational variables M_R and T_R,

$$\frac{\partial P_C}{\partial M_R} < 0, \quad \frac{\partial P_C}{\partial T_R} < 0 \quad \text{(for } E_2 < 2E_1) \quad (B4)$$

from which it can be concluded that P_C is minimized when M_R and T_R are maximized. The condition $E_2 < 2E_1$ holds in most cases.

Regarding the recycle flow D, eq B3 yields

$$\frac{\partial D}{\partial M_R} = \frac{\partial \alpha}{\partial M_R}(F_0 - P_C) - \alpha\frac{\partial P_C}{\partial M_R}, \quad \alpha = \frac{\bar{P}_B}{k_1 M_R - \bar{P}_B}$$

The first term of the right-hand side of the aforementioned equation is negative ($\partial\alpha/\partial M_R < 0$, $F_0 - P_C > 0$), while the second term $-\alpha\partial P_C/\partial M_R$ is positive, which implies that the sign property of $\partial D/\partial M_R$ should be numerically evaluated. In the numerical range of interest, $\partial D/\partial M_R$ and $\partial D/\partial T_R$ all are determined to be negative. Therefore, together with eq B4, the recycle flow increase is accompanied by increased production of the undesired species.

Note that the heuristics derived by Ward et al.,[9] a portion of which is shown in Table 1, applies to the cases where all the reaction orders are the same and the reaction kinetics reflect the reaction stoichiometry. In more-general cases, where reaction orders are different and reaction kinetics are in different forms,

numerical evaluation would be necessary to determine the operating policy.

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