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Control of a High-Purity Ethylene Glycol Reactive Distillation Column with Insights of Process Dynamics

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Inventory control is often regarded as less important than product quality control in the operation of reactive and nonreactive distillation columns (i.e., often detuned considerably in control system design). For the high-purity ethylene glycol reactive distillation column, the inventory control of top condenser is, however, an exception and plays actually a crucial role in the stable and effective process operation, reminding the necessity to thoroughly investigate the intricate dynamic mechanism and its complicated implications on control system synthesis and design. In this article, the dynamics of a high-purity ethylene glycol reactive distillation column is examined, and it is found that the complicated dynamics, for example, the nonminimum phase behavior and process nonlinearity, can be suppressed considerably with the tight inventory control of the top condenser. Moreover, an extremely low controllability is detected, implying the potential difficulties in process operation and thus the need of process design modification. In terms of these insights obtained, two control schemes are devised and studied. It is demonstrated that sharp improvement could be acquired in control system performance when the tight inventory control has been implemented in the top condenser. © 2009 American Institute of Chemical Engineers *AIChE J,* 55: 2106–2121, 2009

Keywords: reactive distillation, process design, nonminimum phase dynamics, inventory control, process operation

Introduction

Although an ethylene glycol reactive distillation column appears to be fairly simple in process configuration, it features unique characteristics in the aspect of process synthesis, design, and control, and has been given considerable attention in recent years.[1] There have already been a lot of

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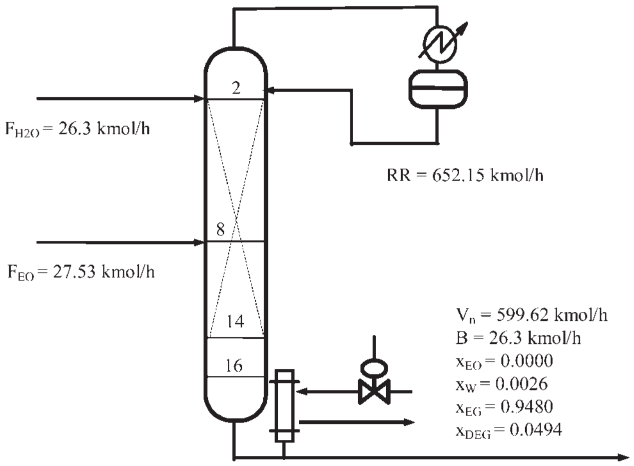


Figure 1. Schematic representation of a high-purity ethylene glycol reactive distillation column.

research articles published on this process. Okasinski and Doherty[2] addressed the synthesis and design of a high-purity ethylene glycol reactive distillation column and presented many useful rules and guidelines for the effective process development. A number of researchers including Ciric and Gu,[3] Cardoso et al.,[4] Jackson and Grossmann,[5] Lima et al.,[6] Papalexandri and Pistikopoulos,[7] and Ismail et al.,[8] conducted the design of a high-purity ethylene glycol reactive distillation column in terms of a mixed-integer nonlinear programming (MINLP) formulation. They demonstrated that adopting multiple feeding locations for the fresh ethylene oxide in the reactive section could lead to larger improvement in the thermodynamic efficiency than the ordinary double-feed arrangement. Huang et al.[9] addressed process synthesis and design by means of reinforcing internal heat integration between the reaction operation and the separation operation involved and found dramatic improvement in the thermodynamic efficiency of the high-purity ethylene glycol reactive distillation column. In a later article, they further demonstrated that such reinforcement of internal heat integration could even be beneficial to process dynamics and operation (Zhu et al.).[32] Steady state input and output multiplicities were found in this system and once sparked considerable interests in the study of their formation mechanism (e.g., Ciric and Miao,[10] Monroy-Loperena and Alvarez-Ramirez,[11] Taylor and Krishna,[1] and Chen et al.[12,13]). To overcome the uncertainties introduced by the input and output multiplicities, a handful of researchers, including, for example, Kumar and Daoutidis[14] and Monroy-Loperena et al.,[15] pointed out the necessity of special effort expended to develop the decentralized control system, for example, by virtue of a nonlinear inversion-based controller and an observer-based input/output linearizing compensator. In terms of a MINLP formulation with an economical objective, Heath et al.[16] attempted to synthesize and design a decentralized proportional-plus-integral (PI) control system for a high-purity ethylene glycol reactive distillation column and showed that the process could be regulated stably with a simple temperature PI control system. Al-Arfaj and Luyben[17] investigated the control system design and operation for a high-purity ethylene glycol reactive distillation column and demonstrated that a simple temperature PI controller plus an appropriate feed-forward compensator between the feed flow rate of fresh ethylene oxide and reflux flow rate could work satisfactorily for the effective regulation of the ethylene glycol composition in the bottom product. Scenna and Benz[18] studied the start-up operation of an ethylene glycol reactive distillation column and indicated that the different branches of steady states could be reached with specific start-up configuration and/or effective operation strategies.

Despite of the continuous effort exerted on the synthesis, design, analysis, and control of the high-purity ethylene glycol reactive distillation column, very little attention has been given to the studies and interpretation of its unique process dynamics and the use of these insights to guiding process development as well as control system synthesis and design. This is actually not a very uncommon problem encountered currently in the control of complicated chemical processes with unique process dynamics. The primary objective of this work is to study the dynamics and its implications on the control of a high-purity ethylene glycol reactive distillation column. The process is found to be of extremely low controllability and possess severe nonminimum phase behavior and strong nonlinearity, which hold close relations with the inventory control of the top condenser. By means of the tight inventory control of the top condenser, the severity of the nonminimum phase behavior and process nonlinearity could

Table 1. Physical Properties and Nominal Operating Conditions of the High-Purity Ethylene Glycol Reactive Distillation Column

Parameters	Values
System pressure (MPa)	0.101
Number of stages	
Reactive section	13
Stripping section	2
Feeding stage of water	2
Feed flow rate of water ($\mathrm{kmol\ h^{-1}}$)	26.3
Feeding stage of EO	8
Feed flow rate of EO ($\mathrm{kmol\ h^{-1}}$)	27.53
The bottom product composition of EG (mole fraction)	0.948
Latent heat ($\mathrm{J\ mol^{-1}}$)	40×10^3
Condenser duty (MW)	7.25
Reboiler duty (MW)	6.66
Distribution of catalyst ($\mathrm{m^3}$)	
Stages 2–8	0.043
Stages 9–14	0.1
Condenser holdup (kmol)	30
Reboiler holdup (kmol)	30
Reaction rate ($\mathrm{mol\ m^{-3}\ s^{-1}}$)	
Main reaction	$3.15 \times 10^{15} \exp(-9547/T) x_{EO} x_W$
Side reaction	$6.3 \times 10^{15} \exp(-9547/T) x_{EO} x_{EG}$
Heat of reaction ($\mathrm{J\ mol^{-1}}$)	
Main reaction	-80.0×10^3
Side reaction	-13.1×10^3
Vapor–liquid equilibrium constants at the atmosphere pressure	
EO	$71.9 \exp[5.720\ (T - 469)/(T - 35.9)]$
Water	$221.2 \exp[6.310\ (T - 467)/(T - 52.9)]$
EG	$77.0 \exp[9.940\ (T - 645)/(T - 71.4)]$
DEG	$47.0 \exp[10.42(T - 681)/(T - 80.6)]$

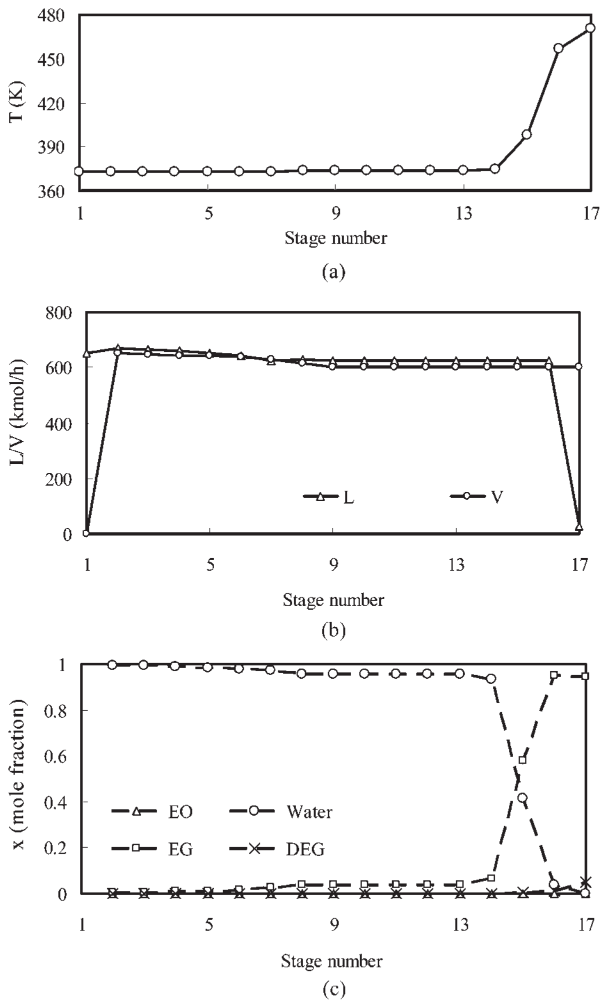


Figure 2. Steady state profiles of temperature, vapor and liquid flow rates, and liquid composition.

(a) Temperature, (b) vapor and liquid flow rates, and (c) liquid composition.

be suppressed considerably, thereby being able to benefit process operation substantially. In terms of the insights gained into the process dynamics, two control schemes are designed and evaluated, and the simulation results obtained confirm the extreme importance of serious dynamics studies prior to the detailed control system synthesis and design.

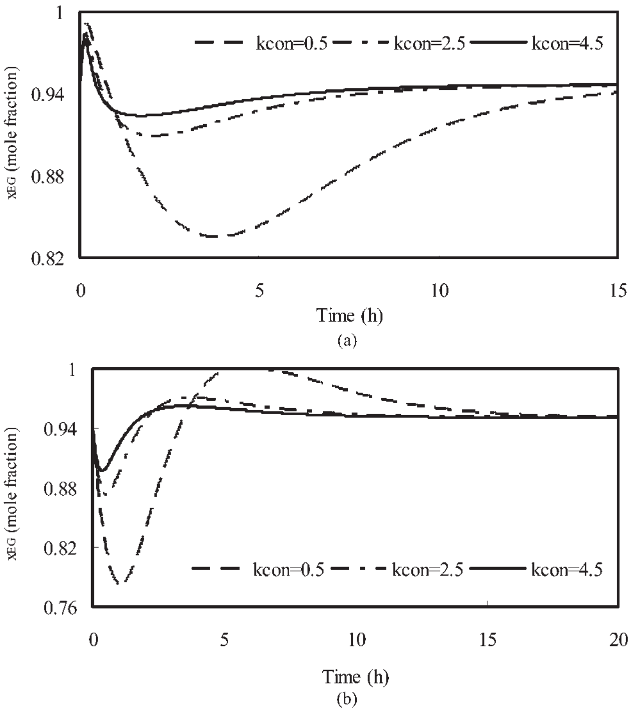


Figure 3. Open-loop transient responses of the high-purity ethylene glycol reactive distillation column when it is subject to a ±2% step change in the boilup flow rate, respectively.

(a) -2%, (b) $+2\%$.

Process Description and Design

Ethylene glycol ($C_2H_6O_2$) is produced from the reaction of ethylene oxide (C_2H_4O) and water:

$$\begin{aligned} &C_2H_4O(\mathrm{EO}) + H_2O \rightarrow C_2H_6O_2(\mathrm{EG}) \\ &\Delta H_R = -80.0 \times 10^3\ \mathrm{kJ/kmol} \end{aligned} \tag{1}$$

$$r(\mathrm{mol\ cm^{-3}s^{-1}}) = 3.15 \times 10^9 \exp[-9547/T] X_{\mathrm{EO}} X_{\mathrm{H_2O}} \tag{2}$$

Ethylene oxide can react further with ethylene glycol to produce the unwanted by-product diethylene glycol ($C_4H_{10}O_3$):

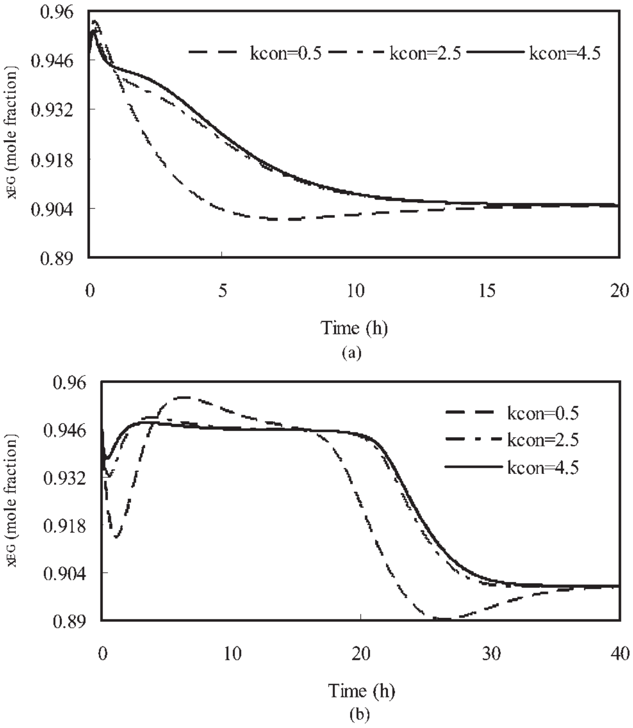
Table 2. Transfer Function Models for the High-Purity Ethylene Glycol Reactive Distillation Column

	Parameters		
Models	$k_{\mathrm{con}} = 0.5$	$k_{\mathrm{con}} = 2.5$	$k_{\mathrm{con}} = 4.5$
Positive responses	$\frac{-0.0026(S+0.0001)(S-0.0014)}{(S+0.0054)(S+0.0054)(S+0.0954)}$	$\frac{-0.0014(S+0.0012)(S-0.0019)}{(S+0.0085)(S+0.0085)(S+0.1487)}$	$\frac{-0.001(S+0.0014)(S-0.0024)}{(S+0.009)(S+0.0095)(S+0.16)}$
Negative responses	$\frac{-0.0013(S+0.0001)(S-0.0175)}{(S+0.0035)(S+0.013)(S+0.14)}$	$\frac{-0.0008(S+0.0002)(S-0.0188)}{(S+0.004)(S+0.03)(S+0.15)}$	$\frac{-0.0007(S+0.0002)(S-0.022)}{(S+0.0044)(S+0.048)(S+0.1528)}$
Balanced responses	$\frac{-0.002(S+0.0001)(S-0.0088)}{(S+0.0045)(S+0.0092)(S+0.1177)}$	$\frac{-0.0011(S+0.0007)(S-0.0104)}{(S+0.0063)(S+0.0193)(S+0.1494)}$	$\frac{-0.0008(S+0.0008)(S-0.0122)}{(S+0.0067)(S+0.0287)(S+0.1564)}$

$$\begin{aligned} &\mathrm{C_2H_4O(EO) + C_2H_6O_2(EG) \rightarrow C_4H_{10}O_3(DEG)} \\ &\Delta H_R = -13.1 \times 10^3\ \mathrm{kJ/kmol} \end{aligned} \tag{3}$$

$$r(\mathrm{mol\ cm^{-3}s^{-1}}) = 6.3 \times 10^9 \exp[-9547/T] X_{EO} X_{EG} \tag{4}$$

Both reactions are highly exothermic and occur at moderate temperatures, allowing the production of ethylene glycol via a reactive distillation column. Figure 1 shows a process design with totally 17 stages including a total condenser at the top and a partial reboiler at the bottom. The process had no rectifying section and was operated in a total reflux mode (i.e., without distillate product) with water and ethylene oxide fed onto Stages 2 and 8 (numbered from the top condenser down to the bottom reboiler), respectively. It involved a high conversion rate of 99.7 mol % (in terms of water), and the relevant physicochemical properties and nominal steady state operating conditions are tabulated in Table 1. An ideal vapor–liquid equilibrium relationship was assumed for the reaction system, and the detailed physicochemical data were taken from Ciric and Gu.[3] In conjunction with the given vapor–liquid equilibrium relationship, a static and a dynamic models were established according to the principle of mass and energy conservation. One may note the unique placement of the reactive section and the special distribution of catalyst, which were actually resulted from the reinforcement of internal heat integration between the reaction operation and the separation operation involved (Zhu et al., submitted).[9] By such internal heat integration, the thermodynamic efficiency could be improved substantially besides an additional reduction in capital investment.



(a)

(b)

Figure 4. Open-loop transient responses of the high-purity ethylene glycol reactive distillation column when it is subject to a ±5% step change in the EO feed flow rate, respectively.

(a) −5%, (b) +5%.

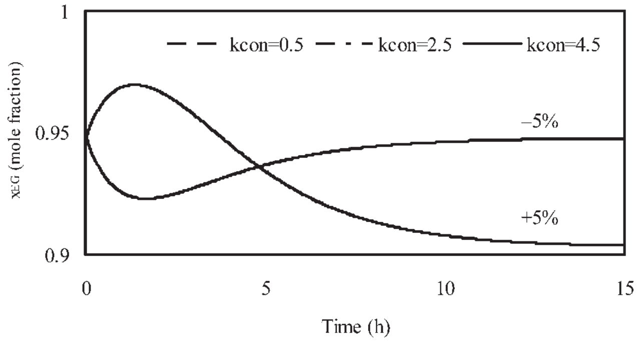


Figure 5. Open-loop transient responses of the high-purity ethylene glycol reactive distillation column when it is subject to a ±5% step change in the water feed flow rate, respectively.

The steady state profiles of temperature, vapor and liquid flow rates, and liquid composition are shown in Figure 2. Similar simulation results were also presented by Ciric and Gu,[3] Baur et al.,[19,20] Chen et al.,[12] Cardoso et al.,[4] Jackson and Grossmann,[5] and Al-Arfaj and Luyben.[17] It was noted that a steep front of composition and temperature was formed at the bottom part of the high-purity ethylene glycol reactive distillation column, which was closely related to the drastic variation of water composition and could be a good candidate controlled variable for the maintenance of the stoichiometric balance between ethylene oxide and water.

Open-Loop Process Dynamics and Its Interpretation

Figure 3 depicts the transient responses of the high-purity ethylene glycol reactive distillation column when it is in the face of a ±2% step change in the heat duty of reboiler, respectively. Three kinds of controller settings (i.e., k_{con} = 0.5, 2.5, and 4.5) were considered for the level of the top condenser. It was noted that severe nonminimum phase behavior (i.e., initial inverse response) was observed in both the positive and the negative responses, which was likely to

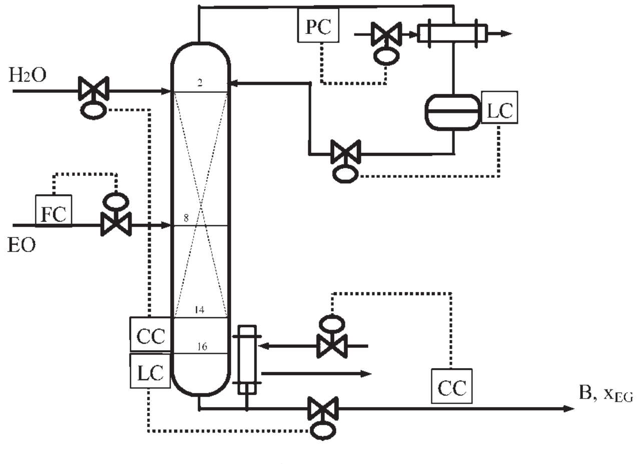


Figure 6. Control scheme I.

Table 3. Controller Parameters for the High-Purity Ethylene Glycol Reactive Distillation Column

Parameters		Condenser Level Control Loop	Reboiler Level Control Loop	Bottom EG Control Loop	Stoichiometric Balance Control Loop
Scheme I	K_C	0.5/2.5/4.5	0.28	4.0	3.6
	T_I	0	0	0.03	0
Scheme II	K_C	0.5	0.28	4.0	3.6
	T_I	0	0	0.03	0

pose a detrimental effect on the process operation. In particular, the inventory control of the top condenser was found to have a strong impact on the severity of the nonminimum phase behavior. To be more exact, the tight inventory control of the top condenser helped to subdue the severity of the nonminimum phase behavior and should therefore be preferred in the control system synthesis and design. This finding was actually a unique feature of the high-purity ethylene glycol reactive distillation column, and it was sharply different from the inventory control of other reactive and nonreactive distillation columns, where the top condenser worked primarily as a surge tank to prevent the propagation of disturbances to the downstream units. Further tightening the inventory control of the top condenser (i.e., $k_{\mathrm{con}} \gg 4.5$) was also attempted, and almost no progress was found on the suppression of the nonminimum phase behavior. Transfer function models were derived for the positive, negative, and balanced (or averaged) responses and listed in Table 2. As can be seen, with the tight inventory control of the top condenser, the positive zero was moved away from the imaginary axis on the right half of the complex plane, implying a certain degree of alleviation of the nonminimum phase behavior. In addition, all the positive poles were moved away from the imaginary axis on the left half of the

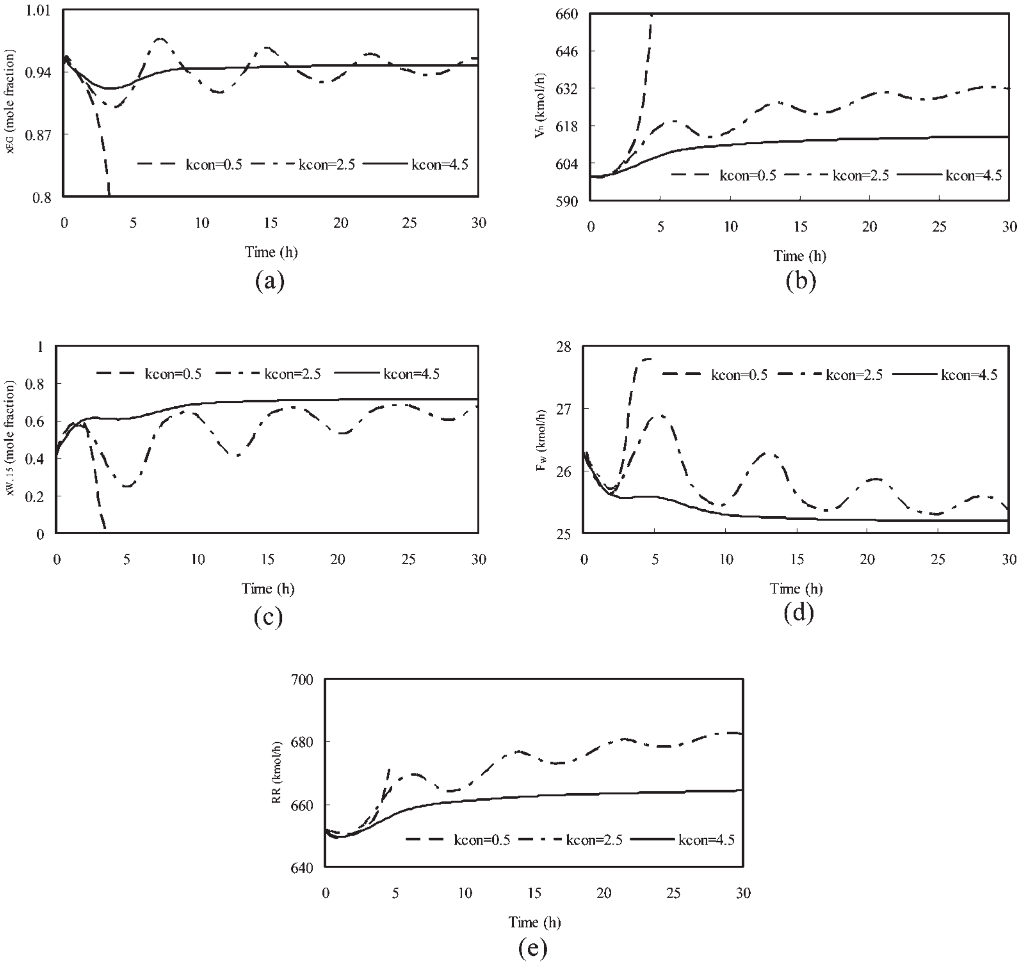


Figure 7. Regulatory responses of the high-purity ethylene glycol reactive distillation column in the face of a −5% step change in the feed flow rate of fresh ethylene oxide.

(a) Bottom EG composition, (b) bottom vapor flow rate, (c) water composition on Stage 15, (d) feed flow rate of fresh water, and (e) reflux flow rate.

complex plane, indicating a certain improvement in process controllability.

The occurrence of the nonminimum phase behavior is frequently caused by the strong interaction between the reaction operation and the separation operation involved in a chemical plant.[21] For the high-purity ethylene glycol reactive distillation column, it was closely related to the variation of the water composition in the reactive section. The increase/decrease of the heat duty of reboiler reduced/enhanced immediately the water composition in the reactive section and thus the conversion rate of ethylene glycol. This was why the bottom composition of ethylene glycol always exhibited an initial inverse response. With the accumulation/depletion of water in the top condenser, the level controller gradually increased/decreased the reflux flow rate and thus the water composition in the reactive section, leading to a relatively slow increase/decrease in the conversion rate of ethylene glycol. The variation caused by the reflux flow rate took a relatively long time and became the dominant one at a certain time after the changes in the reboiler heat duty. The interaction of these two conflicting factors was, in fact, the principal reason for the occurrence of the nonminimum phase behavior. According to this interpretation, it was not difficult to understand why the tight inventory control of the top condenser could help to suppress the severity of the nonminimum phase behavior and, as a result, was likely to lead to improved performance of process operation. Kumar and Daoutidis[14] also noticed that the depletion of water in the reactive section was closely related to the transition of process dynamics from the minimum phase behavior to the nonminimum phase behavior but gave no attempt to confine the nonminimum phase behavior in terms of the tight control of the water composition in the reactive section.

A great degree of asymmetry existed between the positive and the negative responses, reflecting the strong nonlinearity of the high-purity ethylene glycol reactive distillation column. With the tight inventory control of the top condenser, the degree of process asymmetry was also alleviated, substantially, and this reality could also be noticed from the difference among the transfer function models for the positive, negative, and balanced responses. Although the transient responses displayed fairly large variations, the steady state gain of the process appeared to be extremely small (i.e., in the vicinity of 2.3 × 10^{-4} kmol^{-1} h^{-1}, depending on the

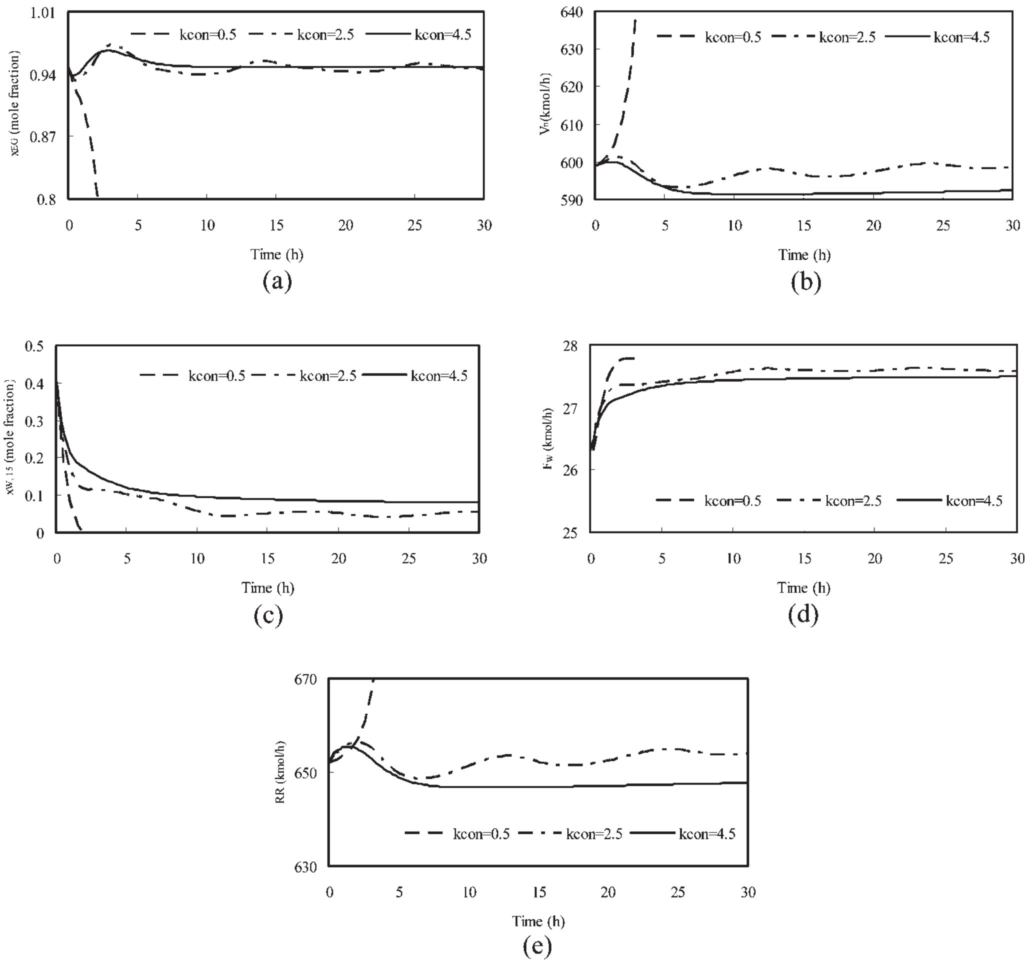


Figure 8. Regulatory responses of the high-purity ethylene glycol reactive distillation column in the face of a +5% step change in the feed flow rate of fresh ethylene oxide.

(a) Bottom EG composition, (b) bottom vapor flow rate, (c) water composition on Stage 15, (d) feed flow rate of fresh water, and (e) reflux flow rate.

magnitude and direction of the changes in the boilup flow rate), implying that the heat duty of reboiler might not be a very effective manipulated variable for the control of the bottom composition of ethylene glycol. The high conversion rate of ethylene glycol was anticipated the main reason that made the process sharply different in dynamics from the case of low conversion rate, worsening the process controllability, substantially.[14,22] Therefore, it is not strongly recommended to operate the high-purity ethylene glycol reactive distillation column in an operating region with a product purity as high as 94.8 mol %.

Figure 4 depicts the transient responses of the high-purity ethylene glycol reactive distillation column when it is in the face of a ±5% step change in the feed flow rate of fresh ethylene oxide, respectively. As can be seen, with the increasingly tight inventory control of the top condenser, the process could become less influenced by the abrupt changes in the operating condition with decreasingly small initial deviations. In the case of the negative responses, the transition to the new steady state was retarded, exhibiting an increasingly slow departure speed. In the case of the positive responses, the process could operate near the nominal steady state in a relatively long time period before the new steady state was reached. These findings indicated evidently that the tight inventory control of the top condenser could also be beneficial to the rejection of the disturbances from the feed flow rate of fresh ethylene oxide (i.e., throughput variations), thereby being favorable to the smooth operation of the high-purity ethylene glycol reactive distillation column. Comparing the positive and the negative responses, one could, again, notice the sharp asymmetrical behavior, which reflected the strong nonlinearity and thus the necessity of special consideration in the control system synthesis and design for the high-purity ethylene glycol reactive distillation column.

As for the transient responses to the step changes in the feed flow rate of fresh water (c.f., Figure 5), almost no influences could be observed with respect to the inventory control of the top condenser. Because a less degree of asymmetry was observed between the positive and the negative responses than the case of the step changes in the feed flow rate of fresh ethylene oxide, it appeared reasonable to use the feed flow rate of fresh water as the manipulated variable for the maintenance of the stoichiometric balance between the reactants.

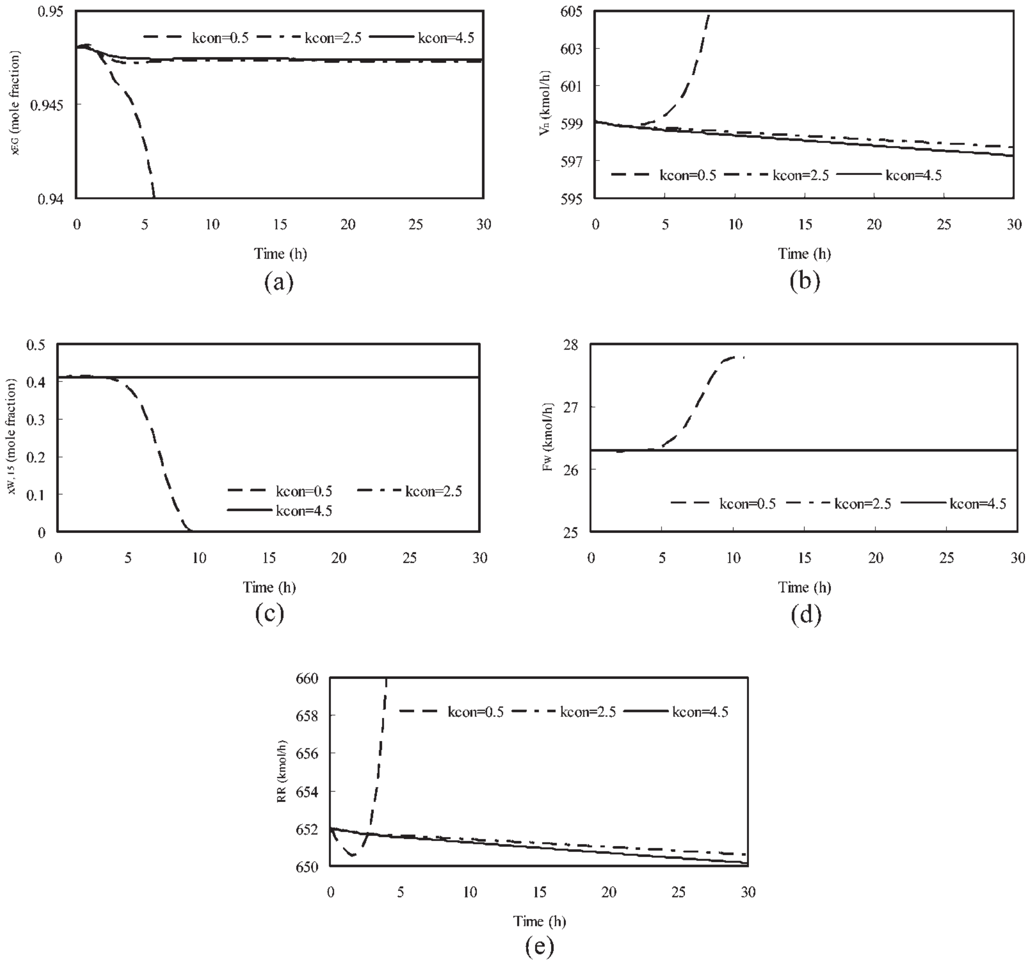


Figure 9. Servo responses of the high-purity ethylene glycol reactive distillation column in the face of a −0.001 step change in the set-point of bottom control loop.

(a) Bottom EG composition, (b) bottom vapor flow rate, (c) water composition on Stage 15, (d) feed flow rate of fresh water, and (e) reflux flow rate.

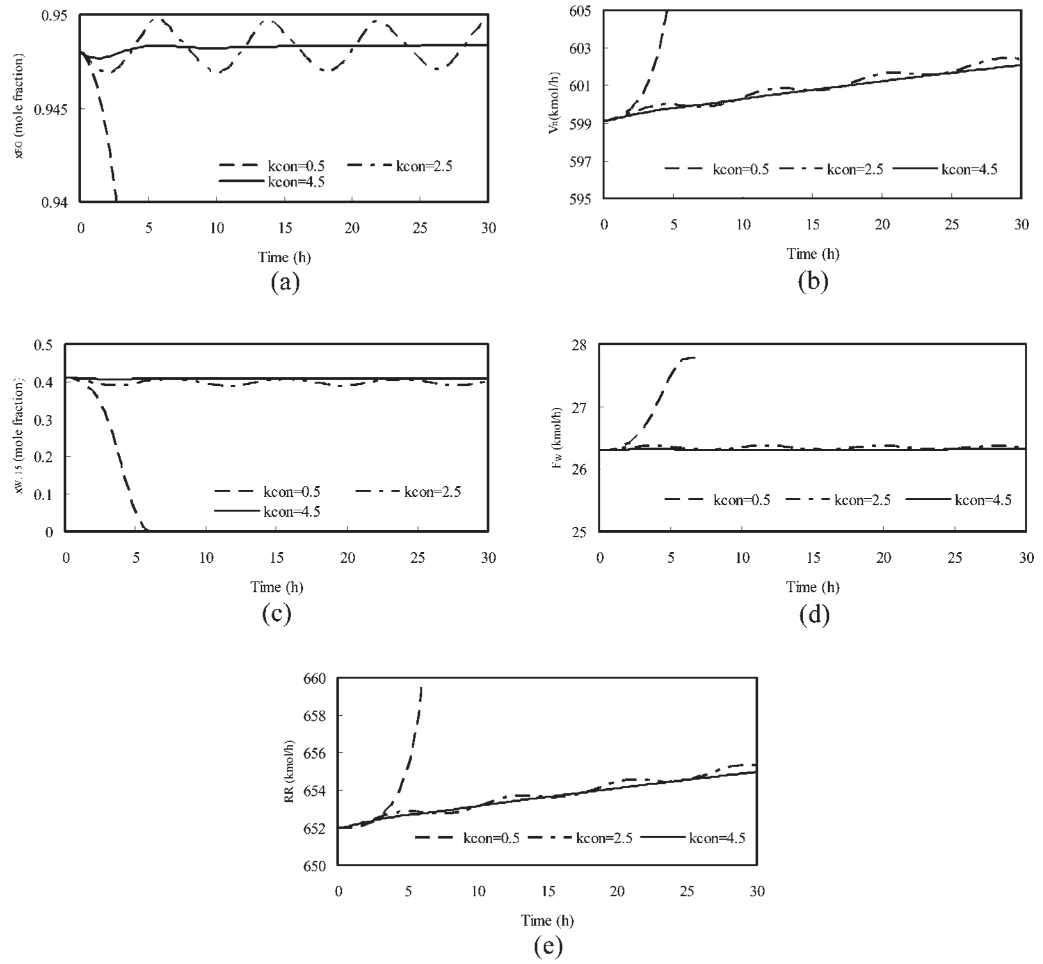


Figure 10. Servo responses of the high-purity ethylene glycol reactive distillation column in the face of a +0.001 step change in the set-point of bottom control loop.

(a) Bottom EG composition, (b) bottom vapor flow rate, (c) water composition on Stage 15, (d) feed flow rate of fresh water, and (e) reflux flow rate.

Remarks

The unique process dynamics and its close relation with the inventory control of the top condenser implied the strong necessity of serious consideration of its impacts on the control system synthesis and design for the high-purity ethylene glycol reactive distillation column. Not only could some useful clues be found for the synthesis of decentralized control structure but also some viable policies could be developed for controller tuning. The extremely small steady state gain between the reboiler heat duty and the bottom composition of ethylene glycol reminded that the operating point chosen could be somewhat problematic for the high-purity ethylene glycol reactive distillation column.

Process Control with Insights of Process Dynamics

In the light of the specific characteristics of the process dynamics, two control systems were devised and studied for the high-purity ethylene glycol reactive distillation column in the sequel. They were all direct composition control schemes but with different control configurations, especially in the aspect of the inventory control of the top condenser. The pressure of the process was assumed to be perfectly

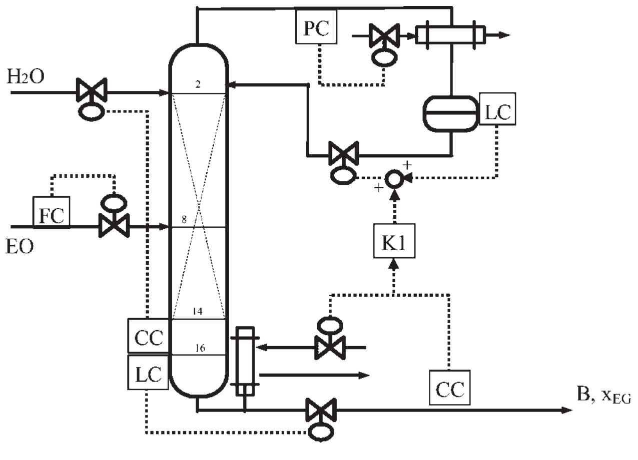


Figure 11. Control scheme II.

controlled at the atmosphere pressure with the cooling medium to the top condenser, and the feed flow rate of fresh ethylene oxide was taken as the throughput handle in all situations. A first-order lag with a time constant of 5.0 min was assumed in all concentration measurements, and all control valves were designed to be half open at the nominal steady state. All composition controllers were tuned in terms of an experimental design approach proposed by Finco et al.[23] The ultimate gain and the ultimate frequency were found by increasing the gain of a proportional-only (P) controller until sustained oscillations occurred. Then, the Ziegler-Nichols settings were calculated for each control loop. Finally, a detuning factor *f* was searched that made all the composition control loops had appropriate damping coefficients.

$$K_C = K_{ZN}/f \tag{5}$$

$$T_I = T_{ZN} \times f \tag{6}$$

where K_C and T_I represent the proportional gain and integral time, respectively, and K_{ZN} and T_{ZN} denote the Ziegler-Nichols settings.

Control Scheme I

The detailed control scheme is depicted in Figure 6. As can be seen, the levels of the condenser and reboiler were controlled, respectively, with the reflux flow rate and bottom product flow rate, and two P controllers were adopted, here. The bottom composition of ethylene glycol was controlled with the reboiler heat duty, and a PI controller was used. According to the steady state profiles of temperature or liquid composition (c.f., Figure 2a or c), the water composition on Stage 15 was chosen and controlled with the feed flow rate of fresh water to keep the stoichiometric balance between the reactants, and a P controller was used. All the controller parameters are listed in Table 3. It was noted that the P controller for the level of reboiler was set to be 0.28, whereas the one for the level of the top condenser was chosen to be 0.5/2.5/4.5, respectively. The latter was much larger than the former and involved a wide range of variations, hence being able to reflect the influences of the inventory control of the top condenser on the process operation.

In Figures 7 and 8, the regulatory responses of the high-purity ethylene glycol reactive distillation column are presented when a ±5% step change is introduced to the feed

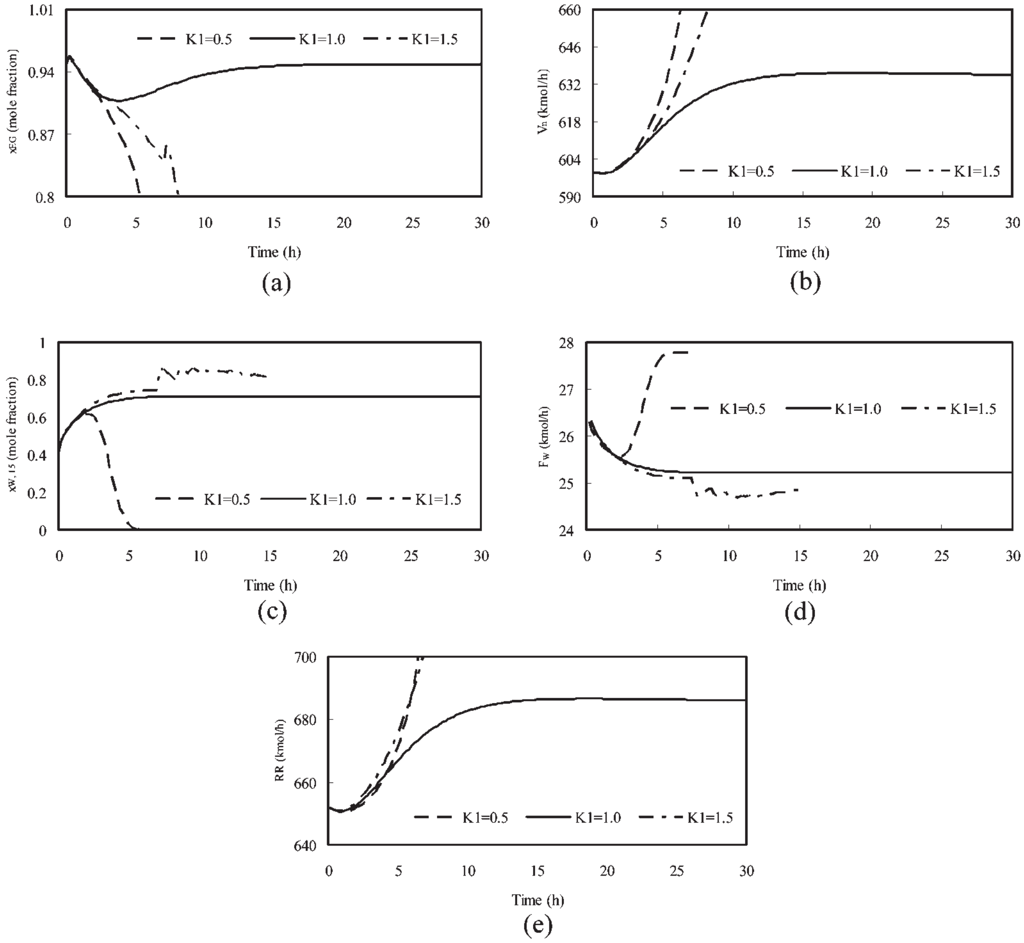
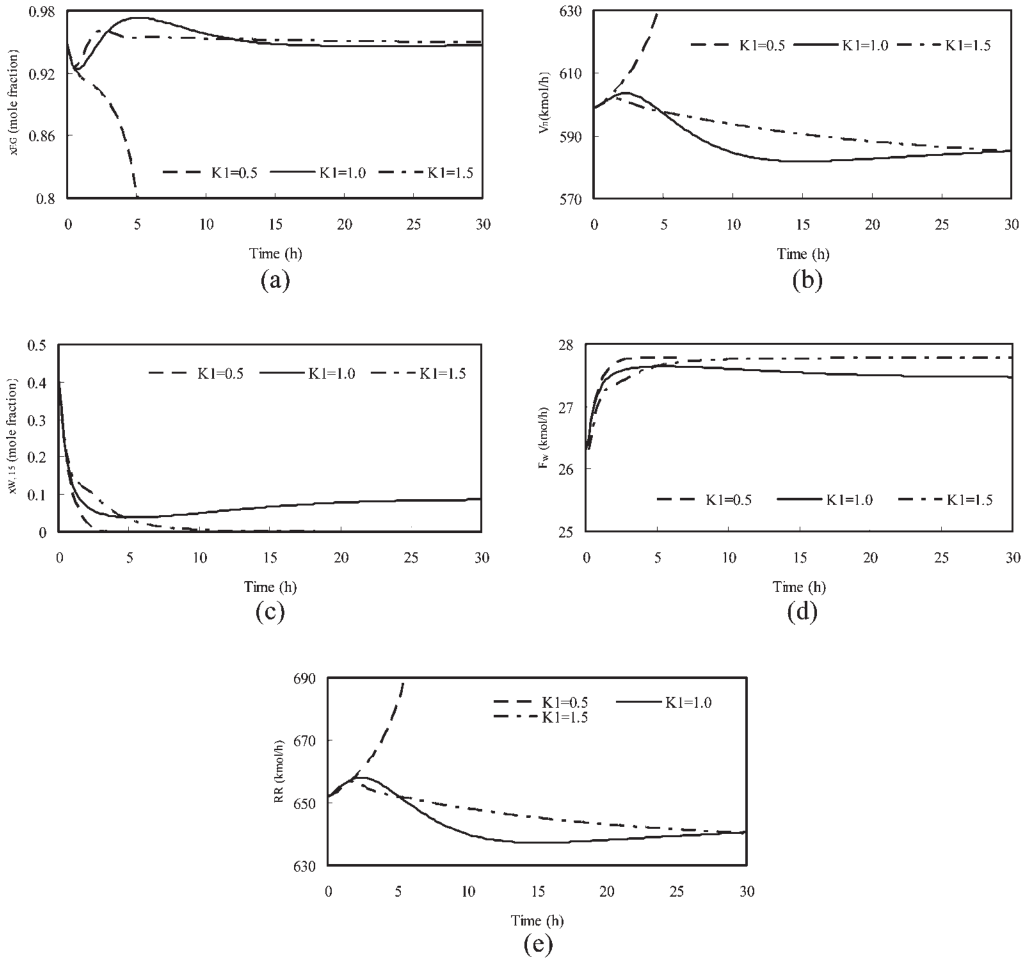


Figure 12. Regulatory responses of the high-purity ethylene glycol reactive distillation column in the face of a −5% step change in the feed flow rate of fresh ethylene oxide.

(a) Bottom EG composition, (b) bottom vapor flow rate, (c) water composition on Stage 15, (d) feed flow rate of fresh water, and (e) reflux flow rate.

flow rate of fresh ethylene oxide, respectively. When the proportional gain of the condenser level controller, k_{con}, was set to be 0.5, the nonminimum phase behavior of the high-purity ethylene glycol reactive distillation column appeared to be much severe, and the bottom composition controller failed to deal with it, resulting in much large deviations from the nominal operating point. Around the time of 4.4 and 3.2 h, the closed-loop simulation was terminated by the numerical errors of overflows, respectively, for the negative and the positive changes in the feed flow rate of fresh ethylene oxide. When the proportional gain of the condenser level controller, k_{con}, was enhanced to be 2.5, the situation was improved substantially. In addition to the elimination of the numerical errors of overflows, the bottom composition of ethylene glycol could be maintained around the desired value. However, a certain degree of oscillation was experienced. When the proportional gain of the condenser level controller, k_{con}, was raised to be 4.5, the situation was improved further, and the process could now be controlled very smoothly, exhibiting fairly small offsets in the bottom composition of ethylene glycol and the feed flow rate of fresh water. It was also noted that the water composition on Stage 15 experienced a dramatic change after the throughput variations; however, a satisfactory maintenance of the stoichiometric balance was still achieved between the two reactants.

In Figures 9 and 10, the servo responses of the high-purity ethylene glycol reactive distillation column are illustrated when a ±0.001 step change is given to the set-point of the bottom control loop, respectively. As can be seen, when the proportional gain of the condenser level controller, k_{con}, was 0.5, the process was driven far away from the nominal steady state (i.e., because of the inability to cope with the nonminimum phase behavior), and the closed-loop simulation was finally terminated (in the vicinity of 4.6 and 7.0 h, respectively) because of the numerical errors of overflows. When the proportional gain of the condenser level controller, k_{con}, was 2.5, the process could operate around the desired steady state, but strong oscillations occurred in the case of the positive change in the set-point of the bottom control loop. Very long time was needed to reach the desired steady state in the case of the negative change in the set-point of the bottom control loop. When the proportional gain of the condenser level controller, k_{con}, was 4.5, stable operation



(a)

(b)

(c)

(d)

(e)

Figure 13. Regulatory responses of the high-purity ethylene glycol reactive distillation column in the face of a +5% step change in the feed flow rate of fresh ethylene oxide.

(a) Bottom EG composition, (b) bottom vapor flow rate, (c) water composition on Stage 15, (d) feed flow rate of fresh water, and (e) reflux flow rate.

could be maintained, but still a very long time was necessary to reach the desired steady states. The slow transition speeds between the different operating points were due to the improper selection of operation region (i.e., too high a conversion rate for ethylene glycol), and this was actually in excellent accordance with our insights gained from the studies of the open-loop process dynamics in the last section.

Control Scheme II

The detailed control scheme is shown in Figure 11. Compared with the control scheme I, one can readily find that it has been reinforced with an additional feed-forward compensator, K_1, from the reboiler heat duty to the reflux flow rate. In terms of the open-loop dynamic analysis in the preceding section, the feed-forward compensator, K_1, is considered helpful to the suppression of the nonminimum phase behavior and process nonlinearity and thus favorable to the operation of the high-purity ethylene glycol reactive distillation column. The exact value of the feed-forward compensator, K_1, should be 1.0, because of the total reflux operation mode. However, because of the sharp difference in the moving rapidity of the vapor and liquid overflows inside the process (including also the slow dynamics of the top condenser), it might be advantageous to set it somehow above 1.0 to exercise a certain degree of compensation for the difference in their moving speeds. Here, three values were assigned, respectively, to the feed-forward compensator, K_1 (i.e., $K_1 = 0.5$, 1.0, and 1.5), and their effect was to be examined through intensive closed-loop simulation. Other controller parameters are also tabulated in Table 3, and one can readily note the fact that the inventory control is in a loose mode for both the condenser and reboiler under this circumstance.

In Figures 12 and 13, the regulatory responses of the high-purity ethylene glycol reactive distillation column are presented when a ±5% step change is introduced to the feed flow rate of fresh ethylene oxide, respectively. When the feed-forward compensator, K_1, was set equal to 0.5, stable operation could not be maintained at all, implying that the function of the feed-forward compensator, K_1, was too weak in this situation. When the feed-forward compensator, K_1,

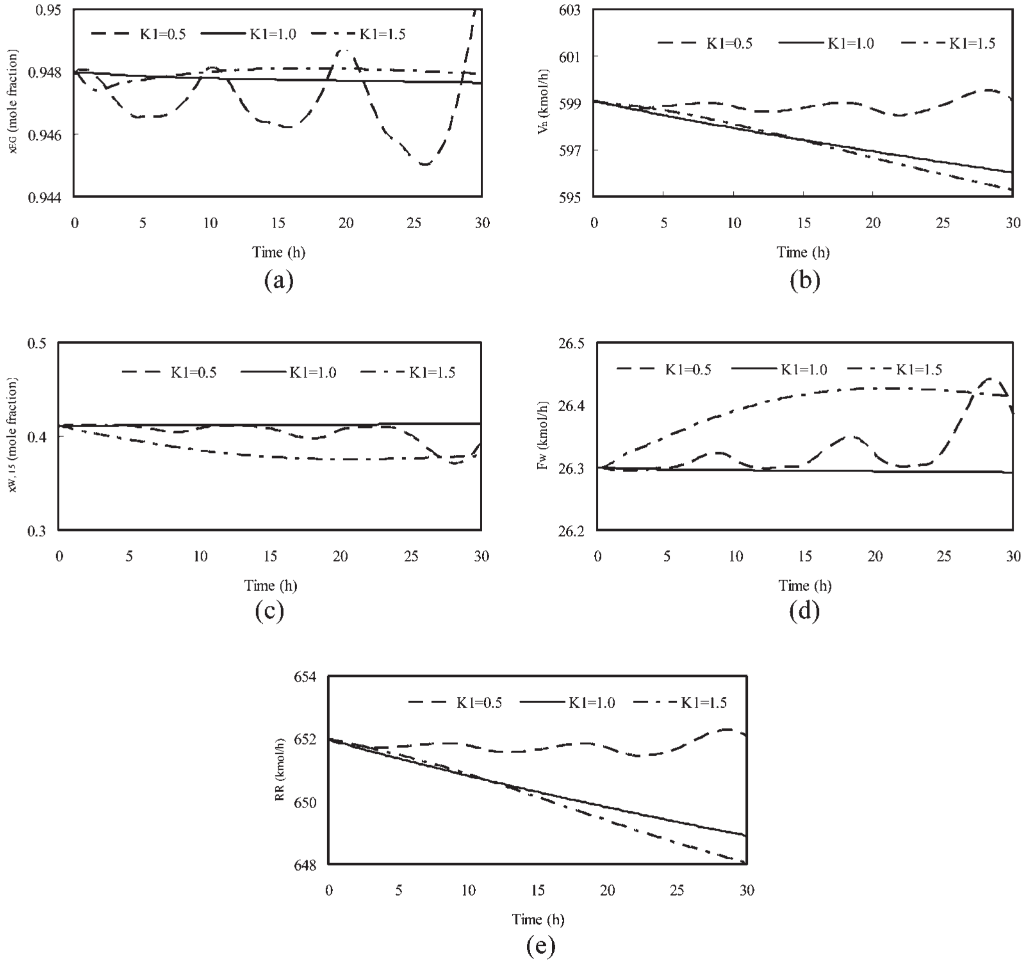


Figure 14. Servo responses of the high-purity ethylene glycol reactive distillation column in the face of a −0.001 step change in the set-point of bottom control loop.

(a) Bottom EG composition, (b) bottom vapor flow rate, (c) water composition on Stage 15, (d) feed flow rate of fresh water, and (e) reflux flow rate.

was set equal to 1.5, stable operation could not be maintained in the case of the negative change in the feed flow rate of fresh ethylene oxide. Although stable operation could be guaranteed in the case of the positive change in the feed flow rate of fresh ethylene oxide, a relatively large discrepancy and a long-settling time were exhibited. These phenomena indicated that the effect of the feed-forward compensator, K_1, was too strong in this situation. When the feed-forward compensator, K_1, was set equal to 1.0, fairly smooth operation could be maintained in both the positive and the negative changes in the feed flow rate of fresh ethylene oxide. These realities indicated that the inventory control of the top condenser was extremely important to the stable operation of the high-purity ethylene glycol reactive distillation column.

In Figures 14 and 15, the servo responses of the high-purity ethylene glycol reactive distillation column are illustrated when a ±0.001 step change is given to the set-point of the bottom control loop, respectively. When the feed-forward compensator, K_1, was set to be 0.5 and 1.5, respectively, smooth operation could not be achieved at all. When the feed-forward compensator, K_1, was set to be 1.0, smooth operation was achieved in both the negative and the positive responses, but still with a very long settling time, implying the severe bottleneck imposed by the process design.

Discussion

The synthesis and design of the control schemes I and II can also be well explained in the context of partial control principle.[24,25] Although the inventory control of the top condenser is not the primary objective for the operation of the high-purity ethylene glycol reactive distillation column, it is actually an important controlled variable that can exert a significant impact on the control of the composition of ethylene glycol in the bottom product. To facilitate the maintenance of the bottom product composition on its specification, it is apparently beneficial to tightly control the inventory of the top condenser, and this in fact differs sharply from the conventional operation philosophy for distillation columns.

The employment of temperature measurements to facilitate direct composition control (i.e., by means of a composition-temperature cascade control configuration) could improve system performance, for example, in the aspect of settling

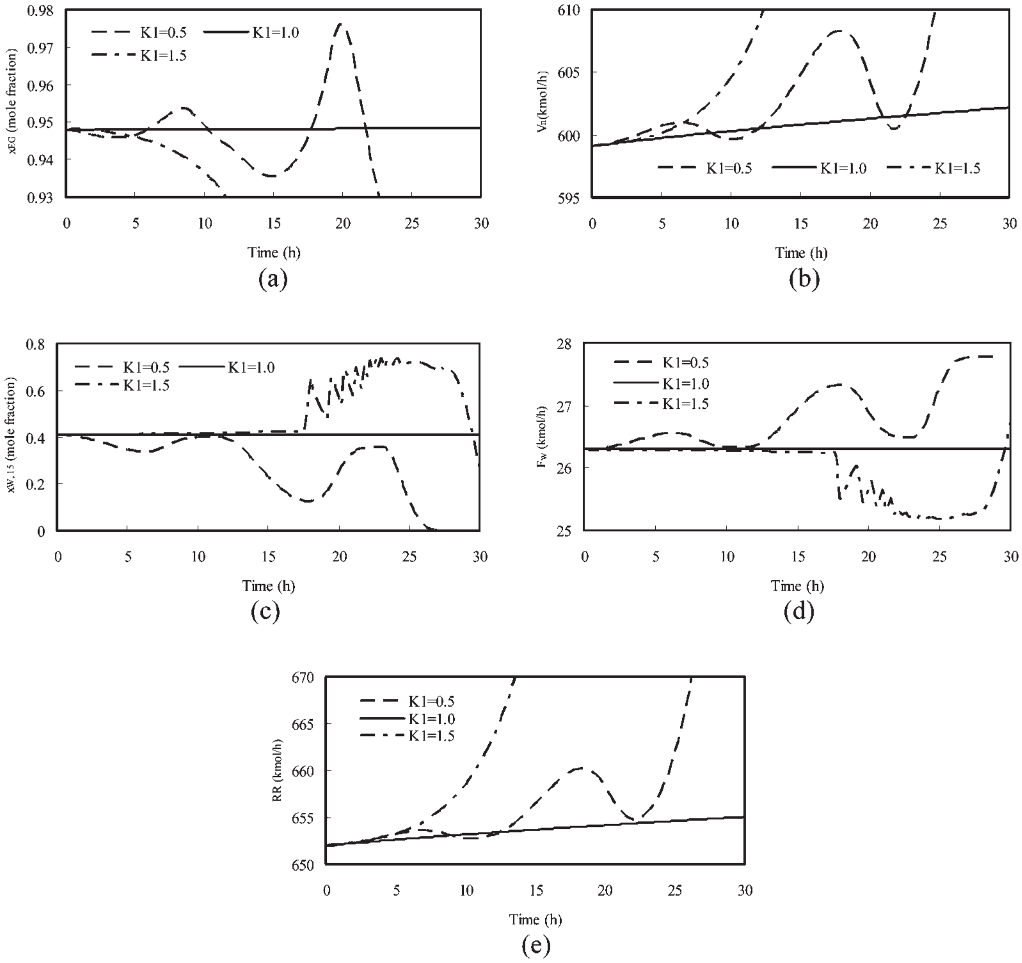


Figure 15. Servo responses of the high-purity ethylene glycol reactive distillation column in the face of a +0.001 step change in the set-point of bottom control loop.

(a) Bottom EG composition, (b) bottom vapor flow rate, (c) water composition on Stage 15, (d) feed flow rate of fresh water, and (e) reflux flow rate.

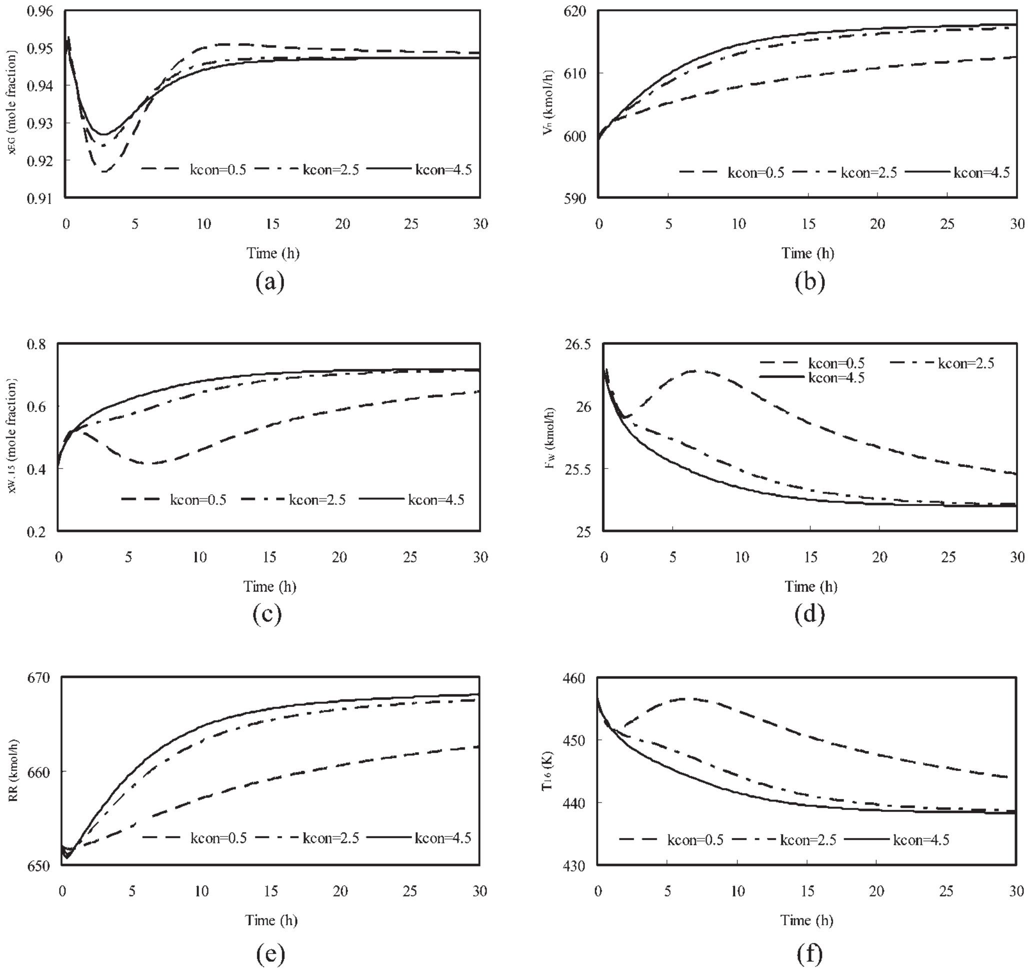


Figure 16. Regulatory responses of the high-purity ethylene glycol reactive distillation column in the face of a −5% step change in the feed flow rate of fresh ethylene oxide (under composition-temperature cascade control in scheme I).

(a) Bottom EG composition, (b) bottom vapor flow rate, (c) water composition on Stage 15, (d) feed flow rate of fresh water, (e) reflux flow rate, and (f) temperature of Stage 16.

time and overshoot. For the high-purity ethylene glycol reactive distillation column, it is reasonable to modify the bottom composition control loop into a composition-temperature cascade one. In Figures 16 and 17, the regulatory responses of the high-purity ethylene glycol reactive distillation column with the modified control schemes I and II are presented, respectively, when a −5% step change is introduced to the feed flow rate of fresh ethylene oxide. The temperature of Stage 16 was selected as the controlled variable in the inner control loop, and a P controller with a proportional gain of 1.6 was used. For the modified Control Scheme I (c.f., Figure 16), although stable operation was achieved for all the three cases examined (i.e., $k_{\mathrm{con}} = 0.5$, 2.5, and 4.5), the increasingly favorable effect, for example, in the aspect of the maximum deviation, settling time, and overshoot, could be readily identified with the increasingly tight inventory control of the top condenser. For the modified Control Scheme II (c.f., Figure 17), whereas stable operation was achieved when K_1 equals to 0.5 and 1.0, instability occurred when K_1 is 1.5, leading to the valve saturation for the reflux flow rate and the heating steam to the bottom reboiler. Again, the tight inventory control of the top condenser presented a favorable effect toward the process operation. As for the servo responses to the set-point changes in the bottom control loop (although not shown here), neither of the two control schemes could exhibit satisfactory tracking performance, implying again the severe bottleneck of the process design.

The extremely low controllability of the high-purity ethylene glycol reactive distillation column indicates the necessity of reconsidering the process design. Figure 18 depicts a process flow sheet involving a combination of one reactive and two nonreactive distillation columns. The first reactive distillation column conducts the hydration of ethylene oxide in a relatively low conversion rate (i.e., $\mathrm{CR_{EG}} < 94.8$ mol %), and the last two distillation columns serve to purify the product of ethylene glycol and meanwhile recycle the unconverted reactants. Although a relatively low selectivity of the reactions is unavoidable in addition to an increased capital investment and operating cost, great improvement in process controllability could be expected.

Because the unique dynamics of the high-purity ethylene glycol reactive distillation column poses additional

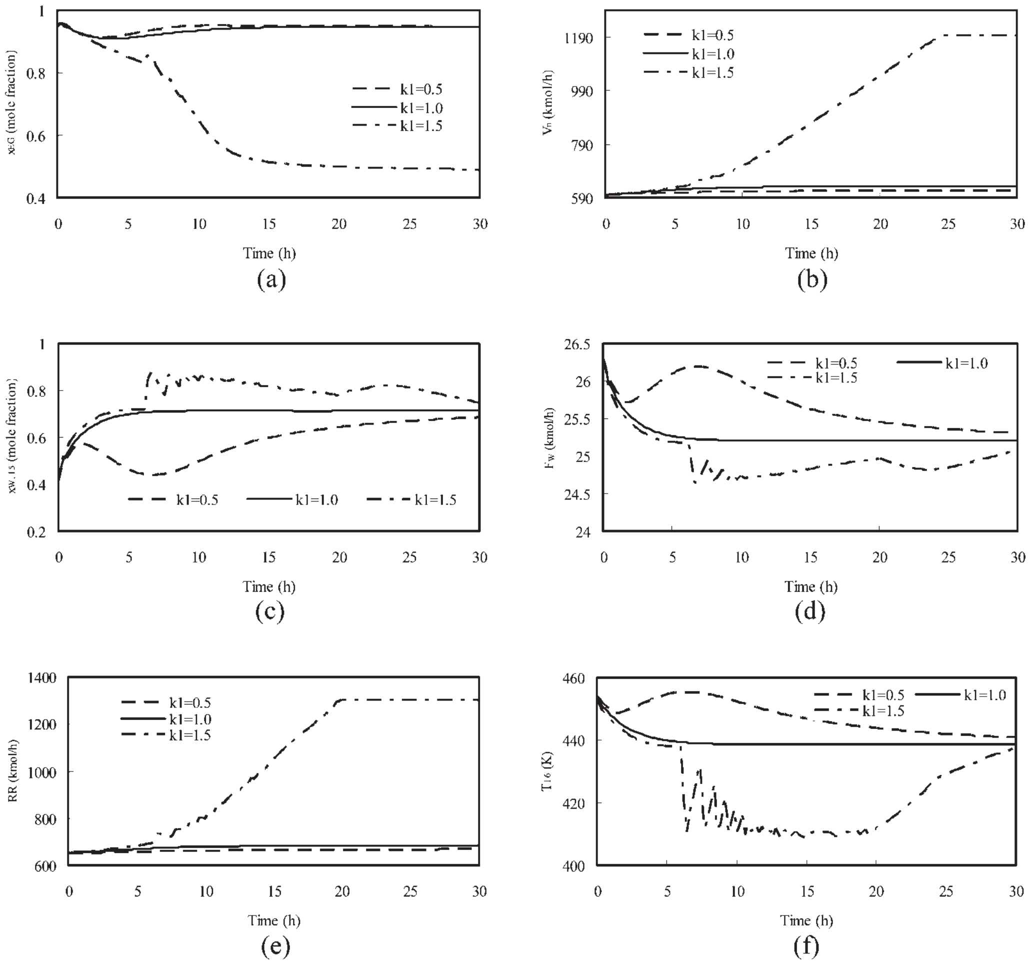


Figure 17. Regulatory responses of the high-purity ethylene glycol reactive distillation column in the face of a −5% step change in the feed flow rate of fresh ethylene oxide (under composition-temperature cascade control in scheme II).

(a) Bottom EG composition, (b) bottom vapor flow rate, (c) water composition on Stage 15, (d) feed flow rate of fresh water, and (e) reflux flow rate, (f) temperature of Stage 16.

requirements on the control system synthesis and design, the dynamic performance of the process could be restricted, accordingly. To ease the restriction, one can rely on refining the design of the high-purity ethylene glycol reactive distillation column. Because liquid hydraulics is one of the major factors that can affect the nonminimum phase behavior and process nonlinearity, selecting stages with quick liquid hydraulics represents therefore a potential way. Decreasing the hydraulic time constant through the reduction of stage holdups constitutes another method. The reinforcement of internal heat integration between the reaction operation and the separation operation involved abates the liquid and vapor flow rates and thus the stage holdups. The striking outcomes obtained so far on the internal heat integration have already demonstrated the great potential.[27,29]

As has been mentioned earlier, Al-Arfaj and Luyben[15] suggested controlling the high-purity ethylene glycol reactive distillation column with the application of a feed-forward compensator between the feed flow rate of fresh ethylene

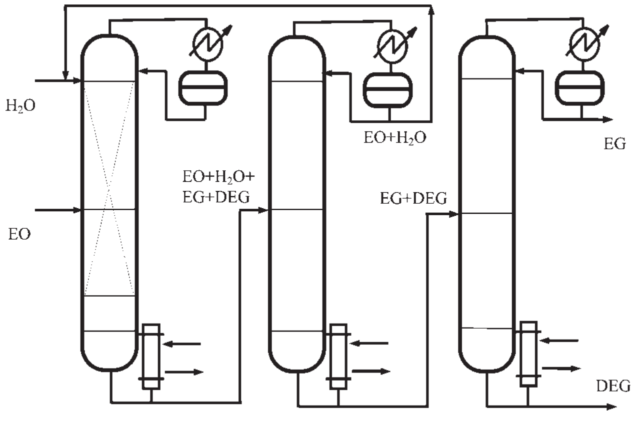


Figure 18. A suggested flow sheet for the production of ethylene glycol by means of combination of reactive and nonreactive distillation columns.

oxide and reflux flow rate. Although it is different in the topological structure from the compensation strategy (i.e., coupling of the reboiler heat duty with the reflux flow rate) proposed in this work, they actually share the same operation mechanism, which are well rooted from the insights gained into the intricate process dynamics of the high-purity ethylene glycol reactive distillation column. In the case of the variations in the feed flow rate of fresh ethylene oxide, immediate changes will occur to the water composition in the reactive section, thus influencing the ethylene glycol conversion rate and its composition in the bottom product. To facilitate the control of the bottom composition of ethylene glycol, one has to counteract the variations of water composition in the reactive section, and the reflux flow rate is apparently the most effective manipulated variable. It should be pointed out here that the compensation policy by Al-Arfaj and Luyben is only effective for the variations in the feed flow rate of fresh ethylene oxide. When changes occur to other operating variables, for example, the set-point in the bottom control loop, it will display no function at all. Under these circumstances, however, the proposed strategy to coupling the reboiler heat duty with the reflux flow rate still works as usual.

Although this work studied exclusively the dynamics and control of the high-purity ethylene glycol reactive distillation column, the results obtained are considered to be of general significance to other reactive distillation systems that contain a reacting mixture with similar physicochemical properties. In fact, a number of practical examples can be found whose operation can be benefited from the insights gained in this work.[29] These include, for example, the synthesis of isopropanol from propylene and water[30] and the esterifications of adipic acid and glutaric acid with methanol.[31]

Conclusions

Although the high-purity ethylene glycol reactive distillation column appears fairly simple in process configuration, it displays unique process dynamics, that is, the extremely low controllability, the strong process nonlinearity, and the severe nonminimum phase behavior with close relation with the inventory control of the top condenser, thus demanding special consideration in process development and delicate strategies in control system synthesis and design. In the aspect of process development, choosing a relatively low composition of ethylene glycol in the bottom product is strongly suggested (c.f., Figure 18), which leads to not only the improvement in process controllability but also the alleviation of the nonminimum phase behavior and process nonlinearity. Unfortunately, a number of side-effects of this modification are incurred, including the relatively low reaction selectivity, augmented investment, and increased operating cost. In the aspect of control system synthesis and design, two alternative strategies have been suggested in this article. One is the tight inventory control of the top condenser with the reflux flow rate as the manipulated variable, and this is actually in sharp contrast to the control strategy for conventional reactive and nonreactive distillation columns. The other is the addition of a feed-forward compensator from the reboiler heat duty to the reflux flow rate. Through intensive open-loop and closed-loop simulation studies, it has been demonstrated evidently that great improvement could be secured in process dynamics and control system performance, for example, in the aspect of set-point tracking and disturbance rejection. This outcome manifests again that gaining deep insight into the inherent process dynamics is an extremely important step towards the effective control of complicated chemical processes, for example, the high-purity ethylene glycol reactive distillation column studied in this work.

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Notation

B = bottom product flow rate, kmol s^{-1}
CC = composition controller
CR = conversion rate
f = detuning factor
F = feed flow rate of reactants, kmol s^{-1}
FC = flow controller
ΔH_R = heat of reaction, kJ $kmol^{-1}$
K_1 = feed-forward compensator between reboiler heat duty and reflux flow rate
K_C = proportional gain
k_{con} = controller gain for the top condenser
L = liquid flow rate, kmol s^{-1}
LC = level controller
n = number of stages
P = pressure, Pa
PC = pressure controller
r = reaction rate, kmol m^{-3} s^{-1}
RR = reflux flow rate, kmol s^{-1}
T = temperature, K
T_I = integral time, s
V = vapor flow rate, kmol s^{-1}
x = liquid composition
y = vapor composition

Subscripts

EO = ethylene oxide
DEG = diethylene glycol
EG = ethylene glycol
W = water
ZN = Ziegler-Nichols settings

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