



Process Systems Engineering and Process Safety

Interpreting the dynamic effect of internal heat integration on reactive distillation columns☆

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ABSTRACT

In this work, the impact of internal heat integration upon process dynamics and controllability by superposing reactive section onto stripping section, relocating feed locations, and redistributing catalyst within the reactive section is explored based on a hypothetical ideal reactive distillation system containing an exothermic reaction: $A + B \leftrightarrow C + D$. Steady state operation analysis and closed-loop controllability evaluation are carried out by comparing the process designs with and without the consideration of internal heat integration. For superposing reactive section onto stripping section, favorable effect is aroused due to its low sensitivities to the changes in operating condition. For ascending the lower feed stage, somewhat detrimental effect occurs because of the accompanied adverse internal heat integration and strong sensitivity to the changes in operating condition. For descending the upper feed stage, serious detrimental effect happens because of the introduced adverse internal heat integration and strong sensitivity to the changes in operating condition. For redistributing catalyst in the reactive section, fairly small negative influence is aroused by the sensitivity to the changes in operating condition. When reinforcing internal heat integration with a combinatorial use of these three strategies, the decent of the upper feed stage should be avoided in process development. Although the conclusions are derived based on the hypothetical ideal reactive distillation column studied, they are considered to be of general significance to the design and operation of other reactive distillation columns.

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1. Introduction

Research outcomes have demonstrated that distributing reactive section (RXS) strictly between rectifying section (RS) and stripping section (SS) is not always a good design option for the reactive distillation columns (RDCs) involving reactions with highly thermal effect, because the potentials of internal heat integration (IHI) between the reaction operation (RO) and the separation operation (SO) involved cannot be fully exploited [1–3]. Based on the second law of thermodynamics, we therefore devised three strategies, i.e., extending the RXS onto the RS (for the RDCs containing endothermic reactions) or the SS (for the RDCs containing exothermic reactions), relocating feed locations, and redistributing catalyst within the RXS, for the reinforcement of IHI during process development [4–8]. Further studies have also indicated that these strategies, if cautiously used in a combinatorial way, are not only effective for enhancing the thermodynamic efficiency of the RDCs having

reactions with highly thermal effect but also likely to yield favorable influences to process dynamics and controllability [6–9]. With regard to the detailed impact of each individual strategy on process dynamics and controllability, no systematic studies have been conducted, yet.

Recently, Kumar and Kaistha in a series of two papers addressed the influences of IHI (i.e., through feed stage relocation and catalyst redistribution by superposing the RXS onto the SS) upon the controllability of an ideal RDC (involving a hypothetical exothermic reaction: $A + B \leftrightarrow C + D$) and a methyl acetate RDC [10,11]. They found that while IHI always presented favorable impact upon the controllability of the latter, unfavorable effect could be observed to the former. Based on these results they claimed that the impact of IHI upon the process dynamics and controllability of a RDC was case dependent. Although their interpretation on process dynamics and controllability appeared reasonable, almost no comprehensive explanations were given on the underlying interplay between process development and process dynamics and controllability. Moreover, their reinforcement of IHI in the ideal RDC was actually not well established. In fact, two places could be questionable and they are: (1) No necessary compensation for mass transfer driving forces was made to the SS after the extension of the RXS; and (2) an adverse IHI was introduced into the process design by descending the upper feed stage. For the first issue, the deficiency of

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mass transfer driving forces caused by the IHI between the RXS and the SS can present certain negative influences to the separation of the reacting mixture and consequently the reaction conversion, intensifying the interaction between the RO and the SO involved. For the second issue, the adverse IHI between the RXS and the RS can restrain considerably the attainable quality of the top product due to the pinch effect aroused, intensifying the interaction between the RO and the SO involved. These two issues might have been the main reasons that made IHI unfavorable to process dynamics and controllability. To gain a deep insight into the effect of IHI on process dynamics and operability, one has therefore to conduct correctly the reinforcement of internal heat integration before its impact on process dynamics and operability is studied.

The current article focuses on the dynamic effect of these three strategies, *i.e.*, extending the RXS onto the SS, relocating feed locations, and redistributing catalyst within the RXS, for deepening IHI between the RO and the SO involved in a hypothetical ideal RDC with an exothermic reaction: $A + B \leftrightarrow C + D$. With regard to each of the three strategies, its impact upon process dynamics and operability is examined through steady state operation analysis and close-loop controllability evaluation. Intensive comparison is also made in the aspect of static and dynamic behaviors between the process designs with and without the consideration of IHI. The underlying interplays between the three strategies for IHI and their impact on process dynamics and operability are analyzed, and some useful guidelines are generalized for IHI through the carefully combinatorial application of these three strategies.

2. IHI versus Process Dynamics and Operability

For a simple conventional distillation column, thermodynamic analysis reveals that its RS needs to release a certain amount of heat to approach reverse operation and functions generally as a heat source [12]. On the contrary, its SS needs to absorb a certain amount of heat to approach reverse operation and functions generally as a heat sink. The interpretations have served as useful guidelines for the development of various heat-integrated distillation columns so far [13]. For a RDC involving a reaction with highly thermal effect, the interpretations can still be applied for process synthesis and design and evolve recently into three strategies for process development, *i.e.*, extending the RXS onto the RS (in case of an endothermic reaction) or the SS (in case of an exothermic reaction), relocating feed locations, and redistributing catalyst within the RXS. These three strategies work to strengthen IHI between the RO and the SO involved and secure consequently great improvement in thermodynamic efficiency.

For a RDC, its dynamics and controllability are mainly determined by the combination between the RO and the SO involved. Inappropriate combination between these two operations is the primary reason that gives rise to complicated process dynamics and worsens consequently process controllability. Although IHI is aimed at the enhancement of thermodynamic efficiency, it is likely to modify the inherent process dynamics and controllability. Two reasons can be listed here. One is the resultant combination between the RO and the SO involved. A high thermodynamic efficiency is yielded with the more coordinated relation between the RO and the SO involved. The same can also be true for the resultant process dynamics and operability. The other is the reduction in the size of RDCs because IHI abates vapor and liquid flow rates and thus stage holdups, leading to a smaller time constant. Obviously, the former is much more prominent than the latter.

It should be indicated here that two characteristics of the scheme for IHI could degrade substantially the dynamics and controllability of a RDC. One is the sensitivity of the thermodynamic efficiency to the changes in operating condition, and the other is the occurrence of adverse IHI. For a derived scheme for IHI, if its thermodynamic efficiency appears sensitive to the changes in operating condition (*e.g.*, variations in throughput and/or product specifications), then the process gains are subject to changes, presenting definitely negative influences to process dynamics and controllability. In cases when favorable IHI is aroused

along with adverse IHI (*e.g.*, relocations of the upper and/or lower feed stages), the resultant adverse IHI can restrict the RO and thus intensify the conflict between the RO and the SO involved. Therefore, interpreting the inherent characteristics of the three strategies for IHI is essential to ascertain their impact on process dynamics and controllability.

One fact should be pointed here that IHI (*i.e.*, extending the RXS onto the RS or the SS) reduces actually the mass transfer driving forces. The degradation in process dynamics and operability thus caused should not be attributed to IHI itself. To ensure the resultant process design with a satisfactory redundancy, the process designers have to make

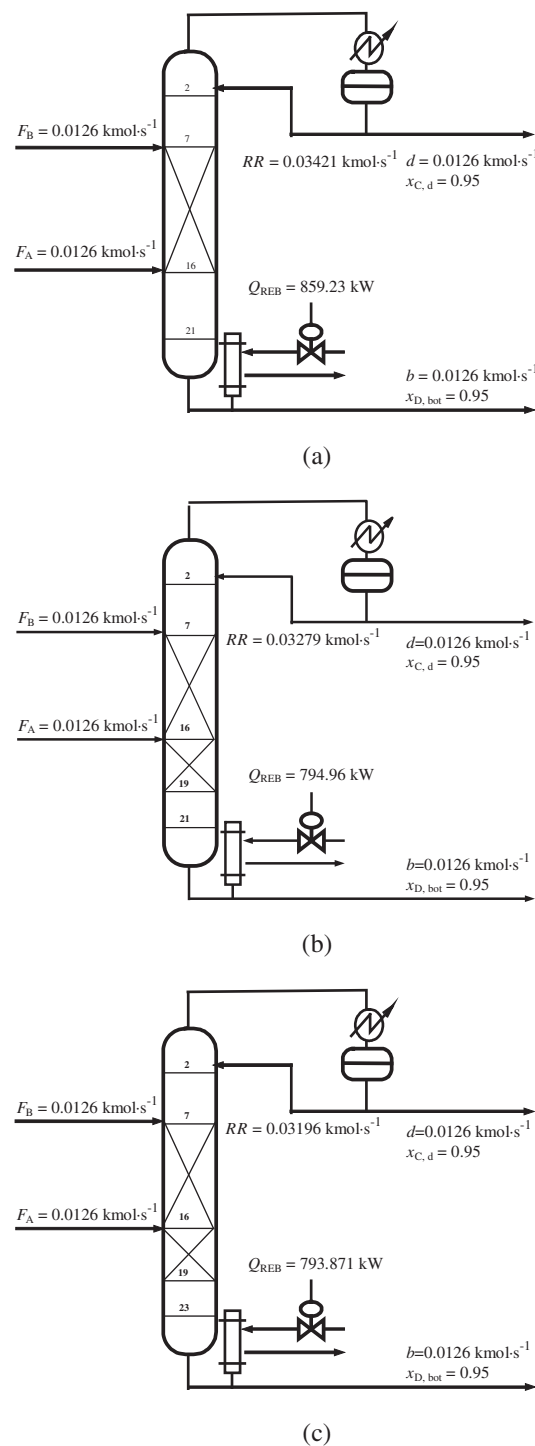


Fig. 1. Schematic of the ideal reactive distillation columns: (a) 5/10/5, (b) 5/10/5(3), (c) 5/10/7(3).

Table 1

Physicochemical properties and operating conditions of the ideal reactive distillation column

Parameter		Value
Number of stages	Rectifying section	5
	Reactive section	10
	Stripping section	5
Pressure $\times 10^{-5}/\text{Pa}$		8.5
Stage holdup/kmol		1
Catalyst loading/kmol $\cdot \text{stage}^{-1}$		0.7
Reaction kinetics	Forward	2.4260×10^{16}
		$\text{Exp}[15098.1/T]$
	Backward	2.11768×10^{-6}
		$\text{Exp}[5032.47/T]$
Feed flow rate of reactant A/mol $\cdot \text{s}^{-1}$		12.6
Feed flow rate of reactant B/mol $\cdot \text{s}^{-1}$		12.6
Feed location of reactant A		15
Feed location of reactant B		6
Thermal condition of F_A		1.0
Thermal condition of F_B		1.0
Relative volatility A:B:C:D		4:2:8:1
Heat of reaction/kJ $\cdot \text{kmol}^{-1}$		-41,480
Latent heat of vaporization/kJ $\cdot \text{kmol}^{-1}$		29053.7
Overhead product composition, C/mol %		95
Bottom product composition, D/mol %		95
Vapor pressure constants	A(A_{vp}/B_{vp})	12.3463/3862
	B(A_{vp}/B_{vp})	11.6531/3862
	C(A_{vp}/B_{vp})	13.0394/3862
	D(A_{vp}/B_{vp})	10.96/3862

necessary compensation by adding some separating stages to the heat-integrated RS or SS. Two kinds of effects are given by such a compensation philosophy. One is certainly the compensation for the mass transfer driving forces, providing redundancy in process operation and lessening the possible conflicts between the RO and the SO involved. The other is the increase in total liquid holdups, which has somewhat negative influences on process dynamics and controllability. The former is generally much greater than the latter especially after the reinforcement of IHI between the RO and the SO involved.

A unified notation, $N_r/N_{rx}(n1, -n2)/N_s(n3)^*$, is defined to represent different process designs with and without the consideration of IHI between the RO and the SO involved in this article. N_r , N_{rx} , and N_s denote the number of stages in the RS, RXS, and SS, respectively. $n1$ and $n2$ signify the movement of the upper and lower feed locations in the RXS, respectively, $n3$ stands for the superimposition of additional reactive stages onto the SS, and the hyphen “-” signifies the movement of the lower feed location upwards from the bottom of the RXS, and the asterisk (*) indicates redistributing catalyst within the RXS. Black lines represent the positive perturbations to operating condition and gray lines the negative perturbations to operating condition.

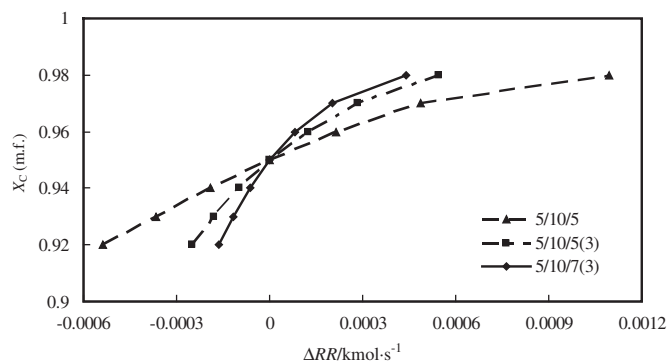
3. A Hypothetical Ideal RDC

Luyben and his co-workers originally defined the hypothetical ideal RDC, and it is then widely used to study process design and operation by

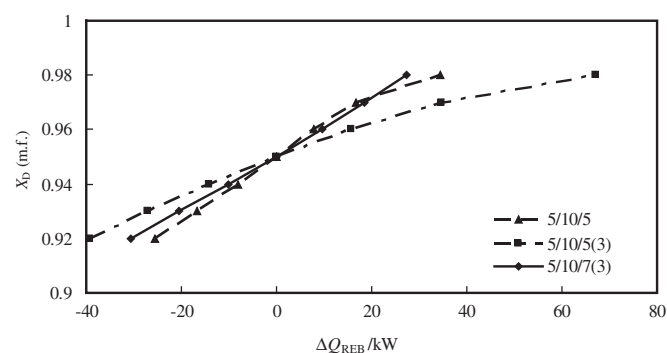
Table 2

Static effect of internal heat integration between reaction operation and separation operation

Design strategy	Process design		
	Process configuration	Reboiler heat duty/kW	Comparison %
Basic process design	5/10/5	859.23	100
Superimposing reactive stages onto the stripping section	5/10/5(3)	794.96	92.52
	5/10/7(3)	793.87	92.39
Relocating the lower feed stage	5/10(0, -4)/5	735.71	85.62
Relocating the upper and lower feed stages simultaneously	5/10(3, -3)/5	700.92	81.58
Redistributing catalyst	5/10/5*	819.77	95.41
Combinatorial use of the three strategies	5/10(0, -3)/7(3)*	710.03	82.64



(a)

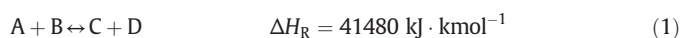


(b)

Fig. 2. Static operation analysis of the ideal reactive distillation columns.

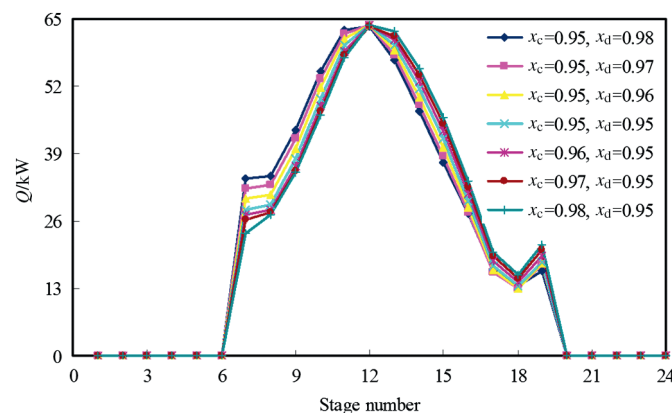
a large number of researchers [14–17]. Fig. 1a reproduces the process design for the hypothetical ideal system, 5/10/5. The light reactant feed, F_A , is fed into the RDC at the bottom of RXS and the heavy reactant feed, F_B , at the top of the RXS. Table 1 lists the physicochemical properties of the reacting mixture and nominal steady-state operating conditions of the RDC.

The hypothetical reversible reaction within the RDC is



The relative volatility ranking of the four components in the reacting mixture is $\alpha_C > \alpha_A > \alpha_B > \alpha_D$. The net reaction rate for component i on stage j in the RXS is given by

$$R_{i,j} = v_i H_j (K_{f,j} x_{A,j} x_{B,j} - K_{b,j} x_{C,j} x_{D,j}) \quad (2)$$

**Fig. 3.** Sensitivity analysis of the profile of reaction heat loads.

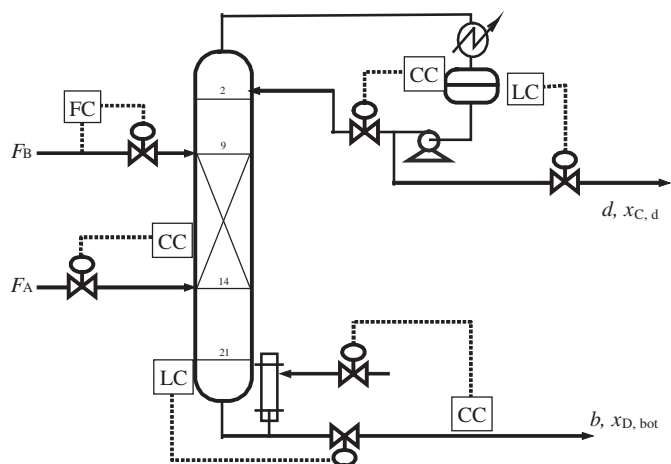


Fig. 4. Control scheme for the ideal reactive distillation column.

where $K_{f,j}$ and $K_{b,j}$ are the forward and backward specific reaction rates and given by

$$K_{f,j} = \alpha_f e^{-E_f/(RT_j)} \quad (3.1)$$

$$K_{b,j} = \alpha_b e^{-E_b/(RT_j)} \quad (3.2)$$

Here, H stands for the liquid holdup, and it can be used to reflect the amount of catalyst installed on a reactive stage. The larger the value of H , the more the amount of catalyst installed on a reactive stage, and vice versa.

Ideal vapor and liquid phase behavior is assumed for the reaction system and the vapor–liquid equilibrium relationship can be expressed as

$$P_j = x_{A,j}P_A^S + x_{B,j}P_B^S + x_{C,j}P_C^S + x_{D,j}P_D^S \quad (4.1)$$

$$y_{i,j} = x_{i,j}P_i^S/P_j \quad (4.2)$$

The vapor saturation pressure is calculated as

$$\ln P_i^S = A_{vp,i} - B_{vp,i}/T_j \quad (5)$$

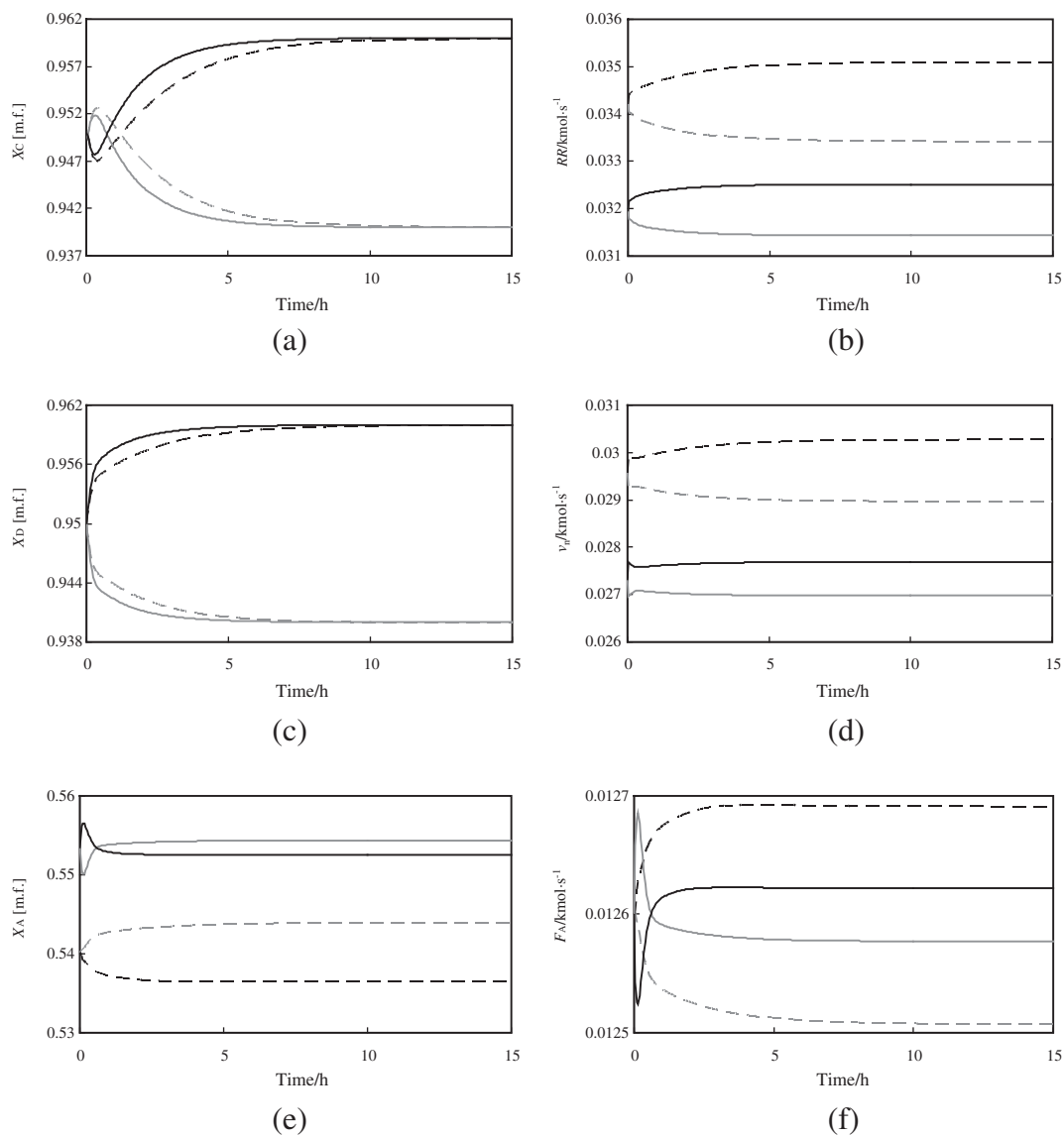


Fig. 5. Servo responses of the ideal reactive distillation columns (solid lines: 5/10/7(3); dashed lines: 5/10/5).

Static and dynamic models are developed in terms of the principle of mass and energy conservation in conjunction with the above vapor–liquid equilibrium relationship. Constant overflow is assumed for the RS and the SS of the hypothetical ideal RDC. In the RXS, the overflow changes from stage to stage because the thermal heat of reaction vaporizes some liquid on each stage. The mass balance equations can be represented as follows:

$$V_j = V_{j+1} - r_{j,c} \Delta H_R / \Delta H_V \quad (6)$$

$$L_j = L_{j-1} + r_{j,c} \Delta H_R / \Delta H_V \quad (7)$$

4. Static and Dynamic Effects of Superimposing the RXS onto the SS

4.1. Static effect of superimposing the RXS onto the SS

With the developed principle for IHI, a process design, 5/10/5(3), as shown in Fig. 1b, is derived through superimposing the RXS onto the SS. In spite of the reduction in the number of stages in the SS, the superimposition of the RXS onto the SS still enhances the thermodynamic efficiency of the ideal RDC. Here, two stages are specially added to the SS

in order to compensate for the reduced mass transfer driving forces, giving rise to a process design, 5/10/7(3), as shown in Fig. 1c. Table 2 compares the process designs with and without the consideration of IHI via superimposing the RXS onto the SS. The reboiler heat duty in the process design, 5/10/5, is regarded as 100%, serving here as a baseline for the comparative studies. The two resultant process designs, 5/10/5(3) and 5/10/7(3), cut the reboiler heat duty by 7.48% and 7.61%, respectively. Note also that the addition of two stages in the SS leads only to a marginal improvement in thermodynamic efficiency.

4.2. Steady state operation analysis

Fig. 2a displays the steady state relationship between the reflux flow rate and the composition of top product when the composition of bottom product has been maintained at its nominal steady state value. The two resultant process designs, 5/10/5(3) and 5/10/7(3), have greater slopes (i.e., static process gains) than 5/10/5, implying that IHI between the RXS and the SS causes clear improvement in process dynamics and controllability. Similarly, Fig. 2b displays the steady state relationship between the reboiler heat duty and the composition of bottom product when the composition of top product has been held at its nominal steady state value. The process design, 5/10/5(3),

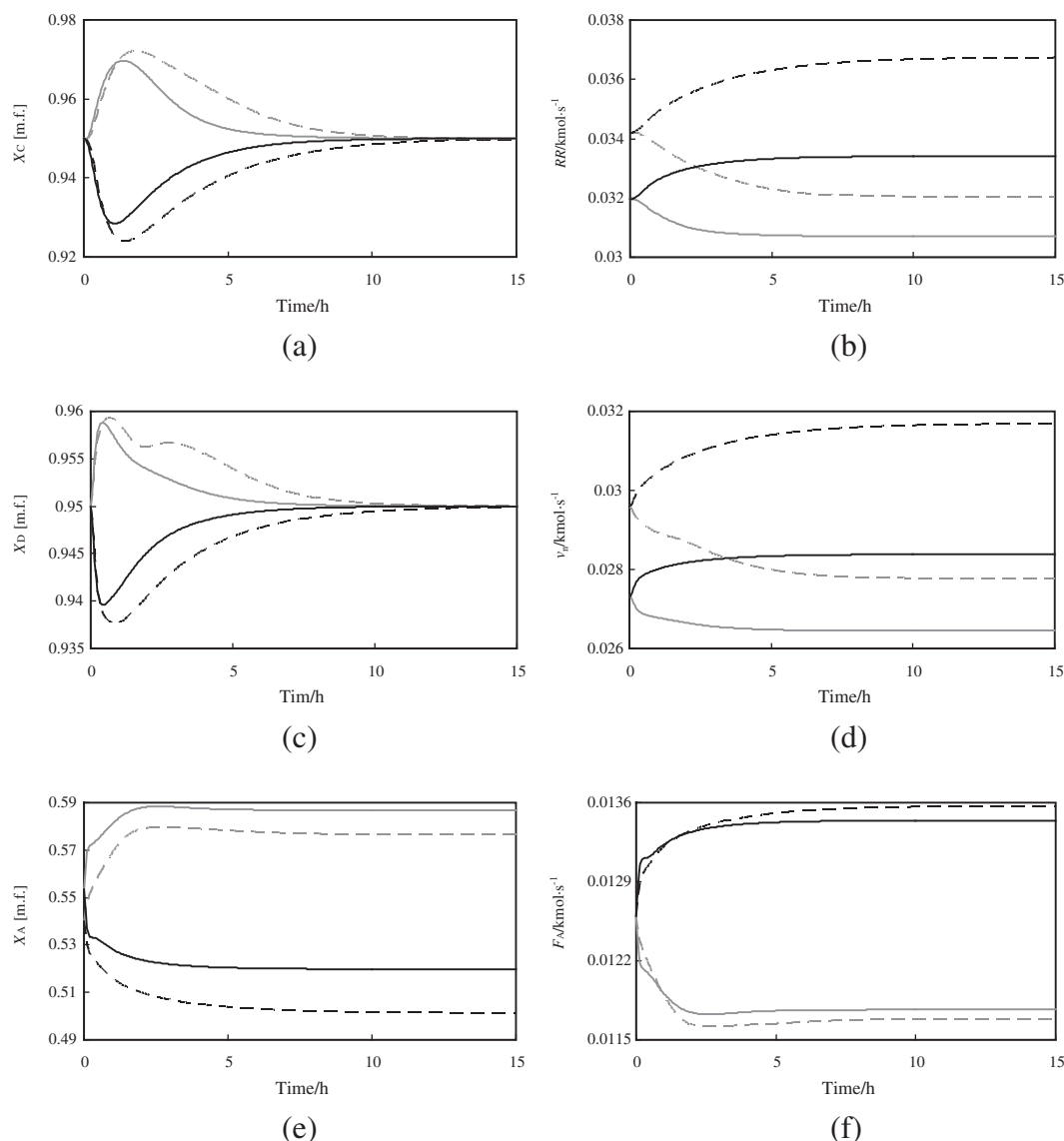


Fig. 6. Regulatory responses of the ideal reactive distillation columns (solid lines: 5/10/7(3); dashed lines: 5/10/5).

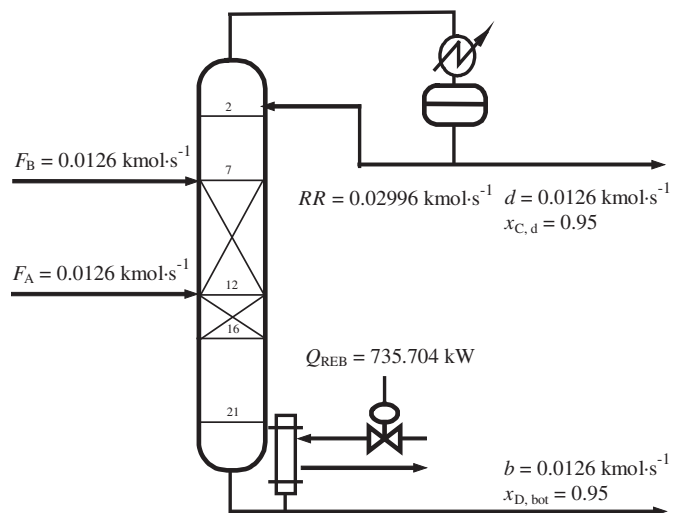
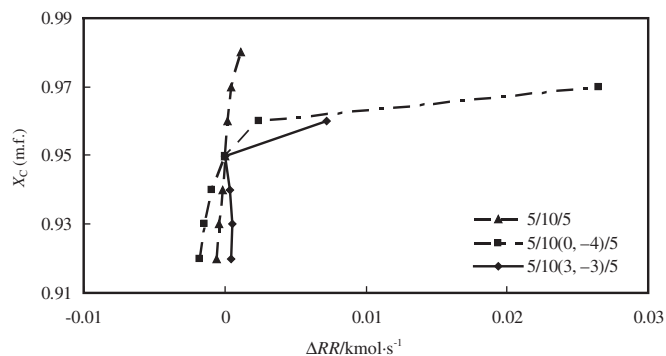


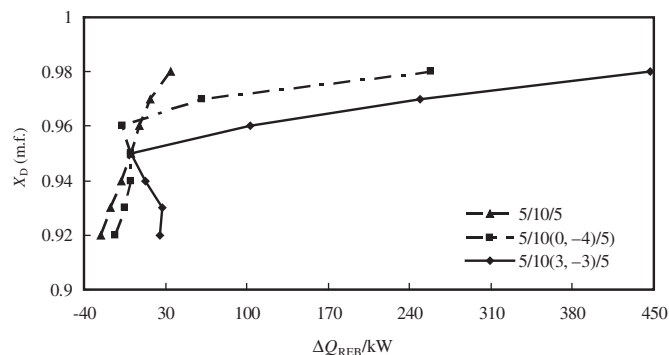
Fig. 7. Schematic of the ideal reactive distillation column, 5/10(0, -4)/5.

has a smaller slope than 5/10/5, indicating that IHI reduces the mass transfer driving forces. With the addition of two stages in the SS, the process design, 5/10/7(3), has now a comparable slope with 5/10/5.

Fig. 3 shows the sensitivities of the profile of reaction heat loads to the variations in the top and bottom product specification. Despite the great variations in operating condition, the profile of reaction heat loads displays no essential changes in shape, implying that IHI between the RXS and the SS can work consistently in a wide operating region. This is why the superimposition of the RXS onto the SS presents actually a favorable impact to the dynamics and controllability of the ideal RDC.



(a)



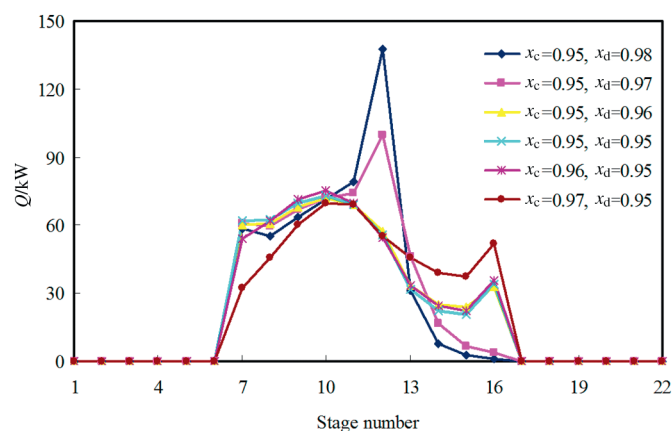
(b)

Fig. 8. Static operation analysis of the ideal reactive distillation column.

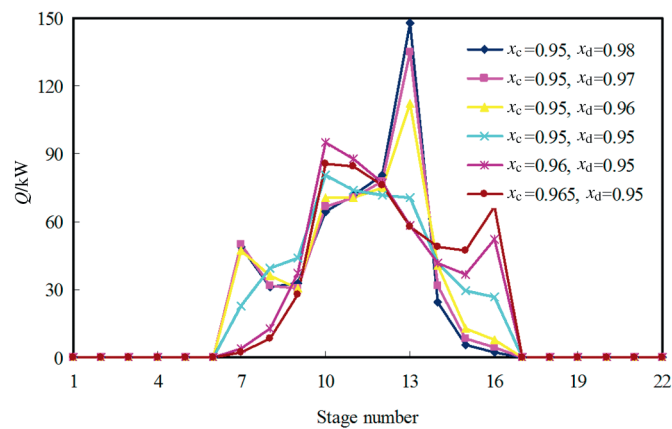
4.3. Dynamic effect of superimposing the RXS onto the SS

In this work, the direct composition control system shown in Fig. 4 is employed. The reflux flow rate is manipulated to control the C composition in the top product; the reboiler heat duty is manipulated to control the D composition in the bottom product and the feed flow rate of A is manipulated to control the concentration of A on the lower feed stage (i.e., the so-called reactant stoichiometry control loop). A flow controller is employed to control the feed flow rate of B. The distillate and bottom product flow rates are manipulated to hold the levels of the reflux-drum and the bottom reboiler, respectively. The dynamics of concentration measurements is assumed to be two first-order lags of 30 s in series. The transmitter span of all composition measurements is taken to be 0.1. The Tyreus–Luyben tuning rule (i.e., $K_C = K_{CU}/3$ and $T_I = 2P_{CU}$) is used to tune the composition controllers for the process design, 5/10/5 [18], and the same controller settings are adopted for those process designs resulted from the reinforcement of IHI between the RO and the SO involved.

The dynamic effect of superimposing the RXS onto the SS is evaluated through intensive comparison of closed-loop responses between the process designs, 5/10/5 and 5/10/7(3). In Fig. 5, their servo responses are depicted when the set-points of the top and bottom control loops have been simultaneously upset by ± 0.01 in magnitudes, respectively. It is noted that the process designs, 5/10/7(3), outperforms 5/10/5 with relatively a short settling time (c.f., Fig. 5a and c) and a small degree of interaction between the top and bottom control loops (c.f., Fig. 5a). At the newly reached steady states, the feed flow rate of reactant A is



(a)



(b)

Fig. 9. Sensitivity analysis of the profile of reaction heat loads: (a) 5/10(0, -4)/5, (b) 5/10(3, -3)/5.

kept closer to the nominal steady state value in the process design, 5/10/7(3), than in 5/10/5 (*c.f.*, Fig. 5f). In Fig. 6, the regulatory responses are depicted when the feed flow rate of reactant B has been upset by $\pm 10\%$, respectively. Again, the process design, 5/10/7(3), outperforms 5/10/5 with relatively a short settling time and small maximum deviations in the top and bottom control loops (*c.f.*, Fig. 6a and c). At the newly reached steady states, however, slightly larger static offsets are observed in the feed flow rate of reactant A in the process design, 5/10/7(3), than in 5/10/5 (*c.f.*, Fig. 6f).

The closed-loop simulation indicates that the process design 5/10/7(3) outperforms 5/10/5 in process dynamics and controllability, thereby corroborating the favorable effect to the operation of the ideal RDC by the superimposition of the RXS onto the SS.

5. Static and Dynamic Effect of Relocating the Lower Feed Stage

5.1. Static effect of ascending the lower feed stage

In accordance with the developed principle of IHI, a process design, 5/10(0, -4)/5, as shown in Fig. 7, is derived by means of the ascent of the lower feed stage. In Table 2, the comparison between the process designs with and without the consideration of IHI through ascending

the lower feed stage is illustrated. The resultant process design, 5/10(0, -4)/5, cuts the reboiler heat duty by 14.38%.

5.2. Steady state operation analysis

Fig. 8a displays the steady state relationship between the reflux flow rate and the composition of top product when the composition of bottom product has been maintained at its nominal steady state value. The resultant process design, 5/10(0, -4)/5, exhibits a smaller slope than 5/10/5, especially when the top product specification becomes much higher than the nominal steady state value, and the degradation in thermodynamic efficiency caused by the variations of operating condition constitutes the main reason. This reality indicates that the reinforcement of IHI through ascending the lower feed stage gives actually certain detrimental effect to process dynamics and controllability.

One inherent phenomenon should be mentioned here for the relocation of the lower feed stage. Although ascending the lower feed location from the bottom of the RXS serves to intensify IHI between the RXS and the SS, the reaction occurs more intensively than before at the top part of the RXS, giving rise to an effect of adverse IHI between the RXS and the RS. Adverse IHI limits the SO in the RS and narrows the feasible operation region (*i.e.*, the so-called pinch effect). For the process

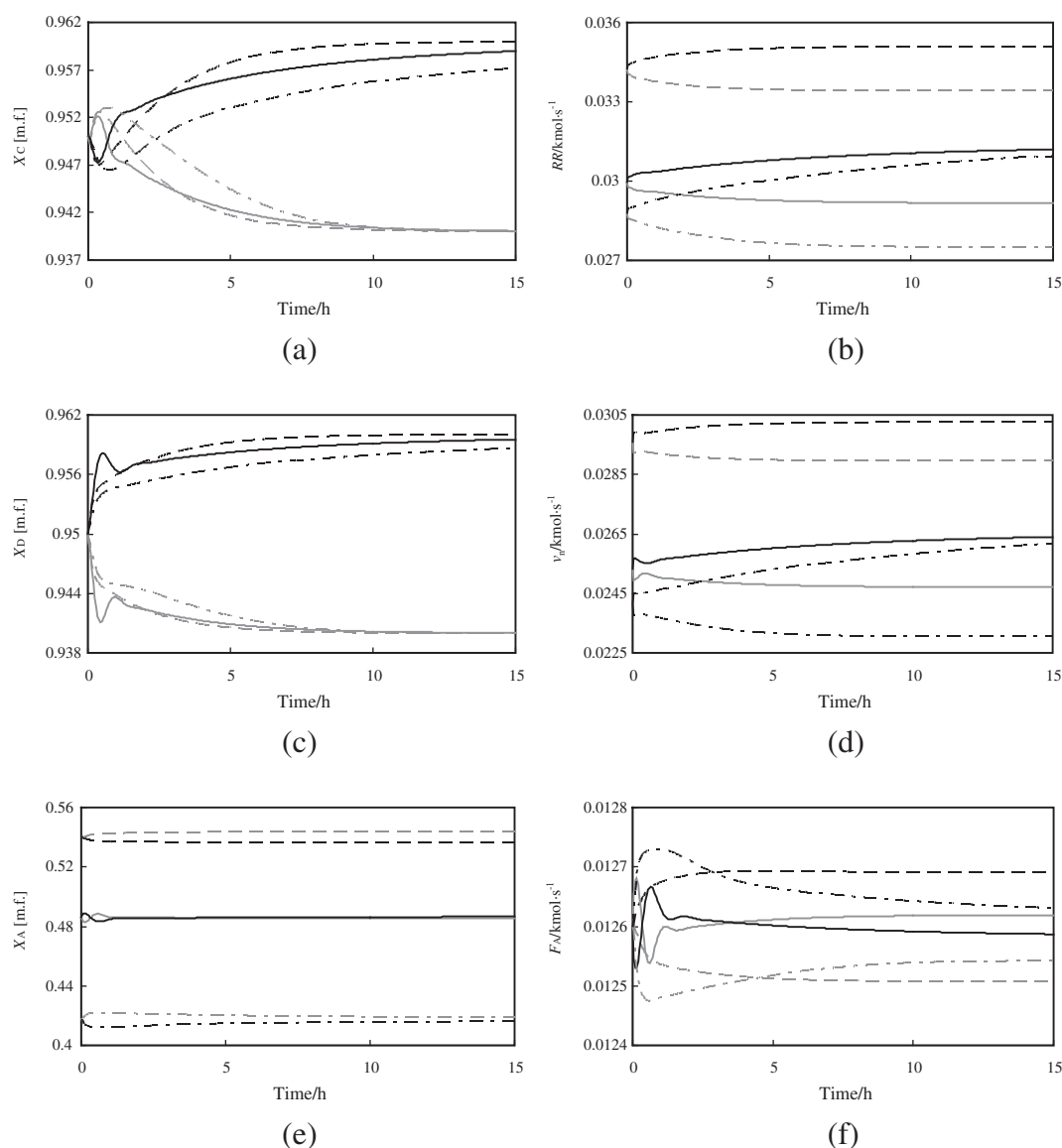


Fig. 10. Servo responses of the ideal reactive distillation columns (solid lines: 5/10(0, -4)/5; dashed and dotted lines: 5/10(3, -3)/5; dashed lines: 5/10/5).

design, 5/10(0, −4)/5, the top product can only reach a purity of about 97 mol%.

Fig. 8b displays the steady state relationship between the reboiler heat duty and the composition of bottom product when the composition of top product has been held at its nominal steady state value. Again, the resultant process design, 5/10(0, −4)/5, displays a smaller slope than 5/10/5 due to the degradation in thermodynamic efficiency. In particular, the phenomenon of output multiplicity occurs, which is caused by the intensified interaction between the RO and the SO involved.

The sensitivity of the profile of reaction heat loads to the changes in product specifications is shown in Fig. 9a. Great variations take place in the shape of the profile of reaction heat loads and they should be responsible for the degradation in the thermodynamic efficiency. The high sensitivity stems again from the intensified interaction between the RO and the SO caused by relocating the lower feed stage.

5.3. Dynamic effect of relocating the lower feed stage

In Fig. 10, the servo responses are depicted when the set-points of the top and bottom control loops have been simultaneously upset by ± 0.01 in magnitudes, respectively. At the onset, the resultant process design, 5/10(0, −4)/5, responds more quickly than 5/10/5. However,

it displays a drastic change in dynamic behavior around the time of 0.5 h and then moves sluggishly to the new set-points, leaving a clear turning point there (c.f., Fig. 10a and c). This phenomenon is certainly caused by the sensitivity of thermodynamic efficiency to the changes in operating condition.

In Fig. 11, the regulatory responses are depicted when the feed flow rate of reactant B has been upset by $\pm 10\%$, respectively. The resultant process design, 5/10(0, −4)/5, outperforms 5/10/5 with relatively a short settling time and small maximum deviations in the top and bottom control loops (c.f., Fig. 11a and c). In the case of negative perturbation in the production throughput, a serious oscillation occurs in the bottom control loop around the time of 0.35 h. It is considered to be aroused by the output multiplicity in the resultant process design, 5/10(0, −4)/5.

6. Static and Dynamic Effect of Relocating the Upper and Lower Feed Stages Simultaneously

6.1. Static effect of relocating the upper and lower feed stages simultaneously

By means of relocating the upper and lower feed stages sequentially, a process design, 5/10(3, −3)/5, is derived as shown in Fig. 12. In

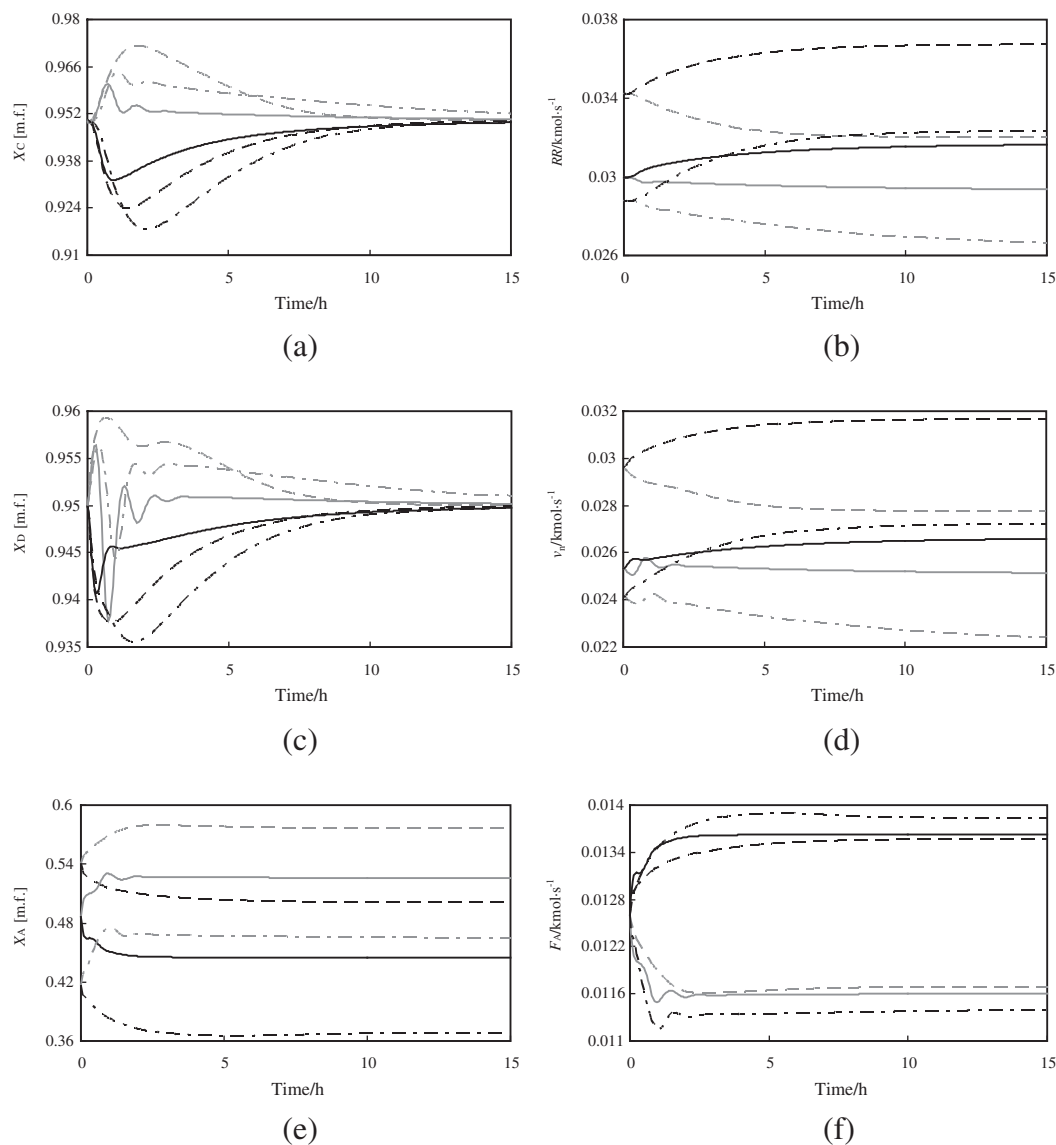


Fig. 11. Regulatory responses of the ideal reactive distillation columns (solid lines: 5/10(0, −4)/5; dashed and dotted lines: 5/10(3, −3)/5; dashed lines: 5/10/5).

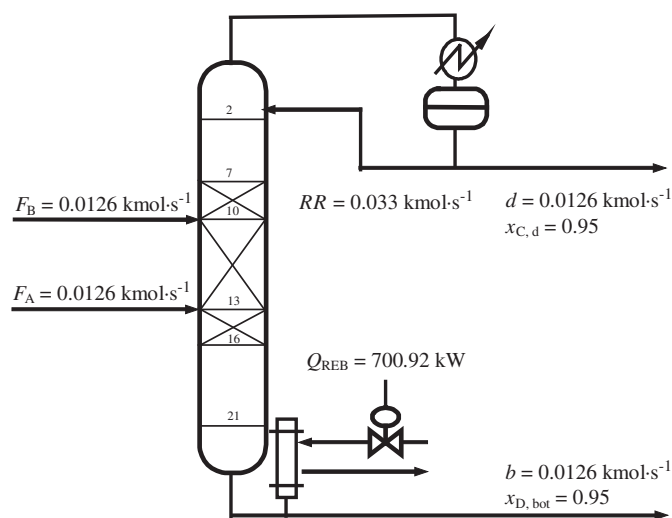


Fig. 12. Schematic of the hypothetical ideal reactive distillation column, 5/10(3, −3)/5.

Table 2, the comparison between the process designs with and without the consideration of IHI through relocating the upper and lower feed stages is illustrated. The resultant process design, 5/10(3, −3)/5, cuts the reboiler heat duty by 18.42%.

6.2. Steady state operation analysis

In Fig. 8a, the steady state relationship between the reflux flow rate and the composition of top product is also given for the resultant process design, 5/10(3, −3)/5, when the composition of bottom product has been held at its nominal steady state value. It is noted that it even has a smaller slope than 5/10(0, −4)/5, implying that descending the upper feed stage presents a negative effect to process dynamics and operability. In spite of the fact that descending the upper feed location from the top of RXS helps to reinforce the IHI between the RXS and the SS, adverse IHI is introduced between the RXS and the RS. One of the side-effects of adverse IHI is the severe confinement of the separation ability in the RS despite the fact that three additional stages have been included (i.e., the so-called pinch effect). In this case, the top product of 5/10(3, −3)/5 can only reach about 96 mol%, lower than those of the process designs, 5/10/5 and 5/10(0, −4)/5.

In Fig. 8b, the steady state relationship between the reboiler heat duty and the composition of bottom product is also delineated for the resultant process design, 5/10(3, −3)/5, when the composition of top product has been held at its nominal steady state value. Again, the resultant process design, 5/10(3, −3)/5, has a smaller slope than 5/10(0, −4)/5. As for the occurrence of output multiplicity, an even wider region is observed.

The sensitivity of the profile of reaction heat loads to the changes in product specifications is shown in Fig. 9b. Again, a great degree of sensitivity is noticed, which stems again from the intensified interaction between the RO and the SO caused by relocating the lower and upper feed stages.

6.3. Dynamic effect of relocating the upper and lower feed stages simultaneously

In Fig. 10, the servo responses are also given for the process design, 5/10(3, −3)/5, when the set-points of the top and bottom control loops have been simultaneously upset by ± 0.01 , respectively. It is readily seen that its dynamic responses feature a much long settling time and are worse than those of 5/10(0, −4)/5. In Fig. 11, the regulatory responses are also given for the process design, 5/10(3, −3)/5, when the feed flow rate of reactant B has been upset by $\pm 10\%$, respectively. Again,

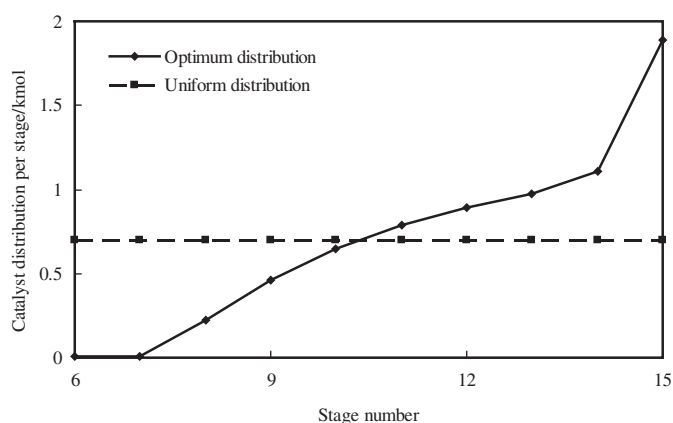


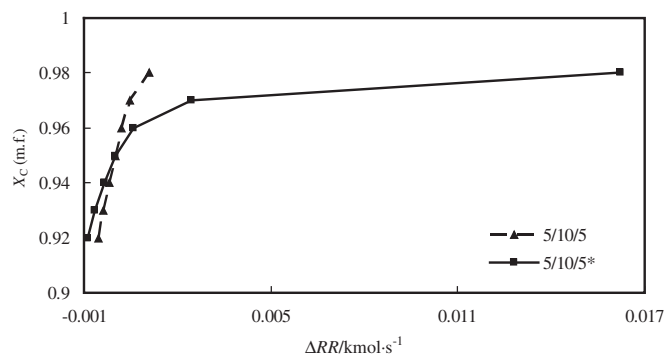
Fig. 13. Distribution of catalyst in the reactive section.

its dynamic responses are not competitive to those of 5/10(0, −4)/5 and 5/10/5.

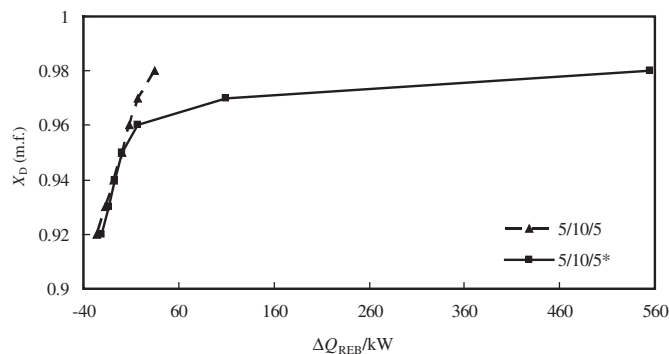
7. Static and Dynamic Effect of Redistributing Catalyst in the RXS

7.1. Static effect of redistributing catalyst within the RXS

With the same total amount of catalyst as the process design 5/10/5, the distribution of catalyst in the RXS is optimized with the minimization of reboiler heat duty as the objective function. The optimization problem is tackled with a modified steepest gradient method, and Fig. 13 depicts the resultant solution. Note that the catalyst is arranged much more intensively at the bottom of the RXS with almost no distribution on the top two reactive stages, thereby facilitating IHI between the RXS and the SS and suppressing at the same time the effect of



(a)



(b)

Fig. 14. Static operation analysis of the ideal reactive distillation column, 5/10/5*.

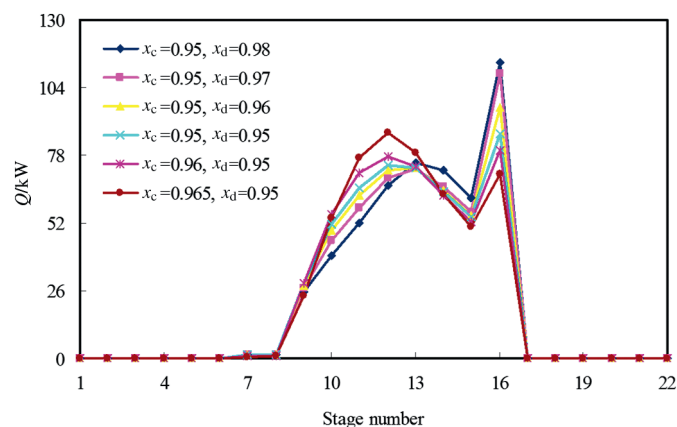


Fig. 15. Sensitivity analysis of the profile of reaction heat loads.

adverse IHI between the RXS and the RS. In Table 2, the improvement of thermodynamic efficiency through redistributing catalyst within the process design, 5/10/5*, is displayed, and a reduction of 4.59% in the reboiler heat duty has been secured.

7.2. Steady state operation analysis

Fig. 14a displays the steady state relationship between the reflux flow rate and the top product composition when the bottom product composition has been maintained at its nominal steady state value. With reference to the process design, 5/10/5, one can readily find that the thermodynamic efficiency of the process design, 5/10/5* drops to a certain extent especially when the top product composition is enhanced far from the nominal steady state value. Fig. 14b displays the steady state relationship between the reboiler heat duty and the composition of bottom product when the composition of top product has been maintained at its nominal steady state value. Again, a certain degree of degradation in thermodynamic efficiency happens when the bottom product specification is disturbed from its nominal steady state value. Extremely similar to the cases of relocating feed positions, although the redistribution of catalyst enhances process thermodynamic efficiency, it deepens the sensitivity to operating condition changes due to the intensified interaction between the RO and the SO involved.

The tendency can also be confirmed from the sensitivity of the profile of reaction heat loads to the changes in product specifications as shown in Fig. 15. Since the optimum distribution of catalyst shown in Fig. 13

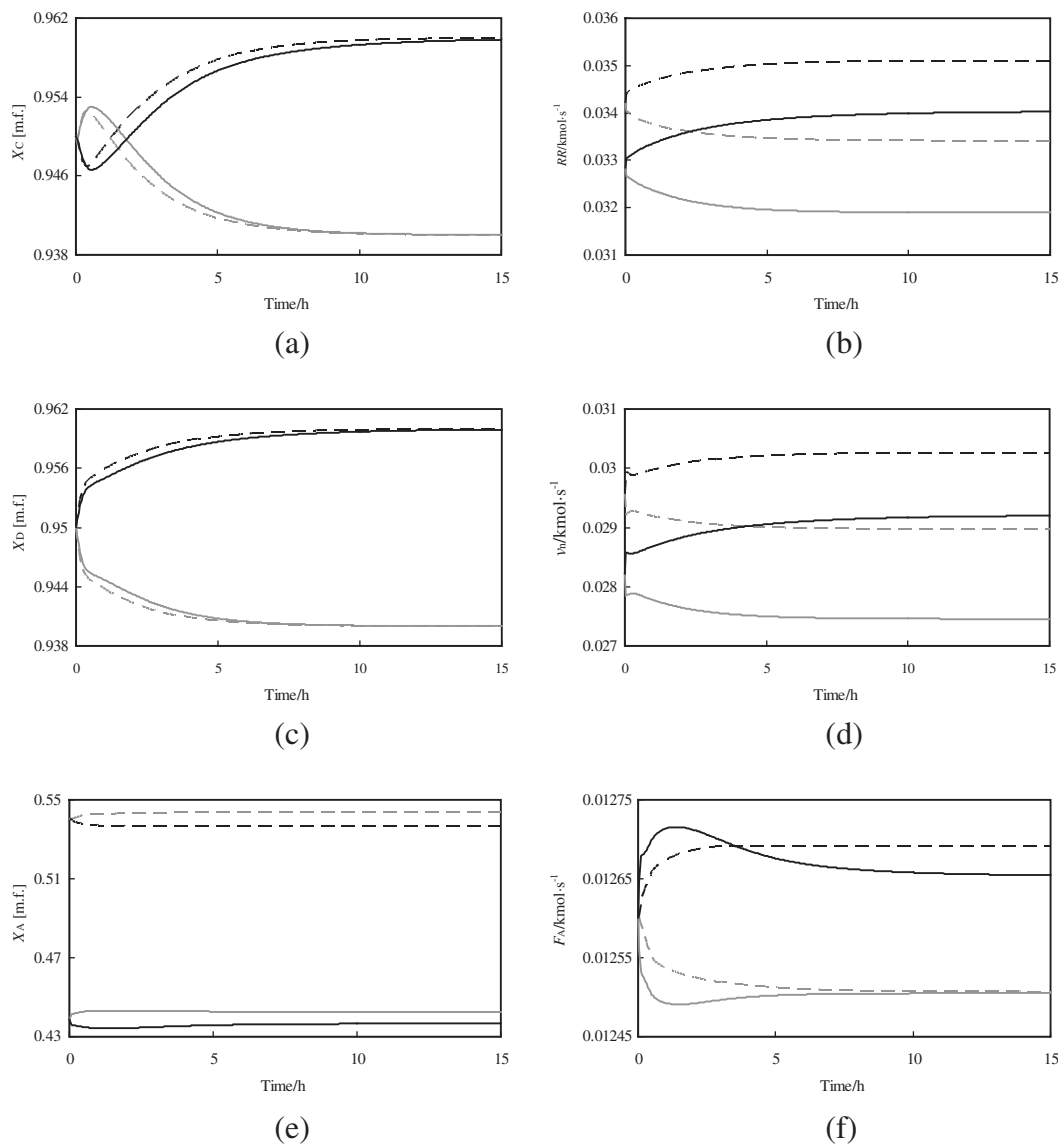


Fig. 16. Servo responses of the ideal reactive distillation columns (solid lines: 5/10/5*; dashed lines: 5/10/5).

corresponds to the given nominal steady state, the variations in product specifications leads certainly to a somehow degradation in thermodynamic efficiency, inevitably worsening process dynamics and operability.

7.3. Dynamic effect of redistributing catalyst within the RXS

In Fig. 16 the servo responses are depicted when the set-points of the top and bottom control loops have been simultaneously upset by ± 0.01 , respectively. The resultant process design, $5/10/5^*$, fails to compete with $5/10/5$, presenting relatively a long settling time and a great degree of interaction between the top and bottom control loops (c.f., Fig. 16a and c). In Fig. 17 the regulatory responses are given when the feed flow rate of reactant B has been upset by $\pm 10\%$, respectively. The resultant process design, $5/10/5^*$, cannot compete with, $5/10/5$, either (c.f., Fig. 17a and c).

8. Discussion

Although not shown in the current work, the changes in production throughput appear not to affect the profile of reaction heat loads

considerably [4,19]. The sharp degradation in the regulatory responses (e.g., in the process design, $5/10(0, -4)/5$ and $5/10(3, -3)/5$) is therefore mainly due to the adverse IHI between the RXS and the RS. Remember the fact that the thermodynamic efficiency of a RDC with the reinforcement of IHI is quite sensitive to the perturbation magnitudes in operating condition (c.f., Figs. 9 and 15), one should take this factor into consideration when evaluating the impact of IHI on process dynamics and controllability. In process synthesis and design, enough redundancy must be guaranteed to counteract the uncertainties in both static and dynamic behaviors.

According to the above static and dynamic analysis, it has been clarified that the three strategies for IHI present different impacts to process dynamics and controllability and judicious discrimination between them should therefore be made. For superimposing the RXS onto the SS, it should be considered in process development since favorable effect has been observed on process dynamics and operability. For ascending the lower feed location, it can still be employed in process development despite its detrimental effect. For descending the upper feed location, it should be avoided in process development because of the introduced adverse IHI between the RXS and the RS. For

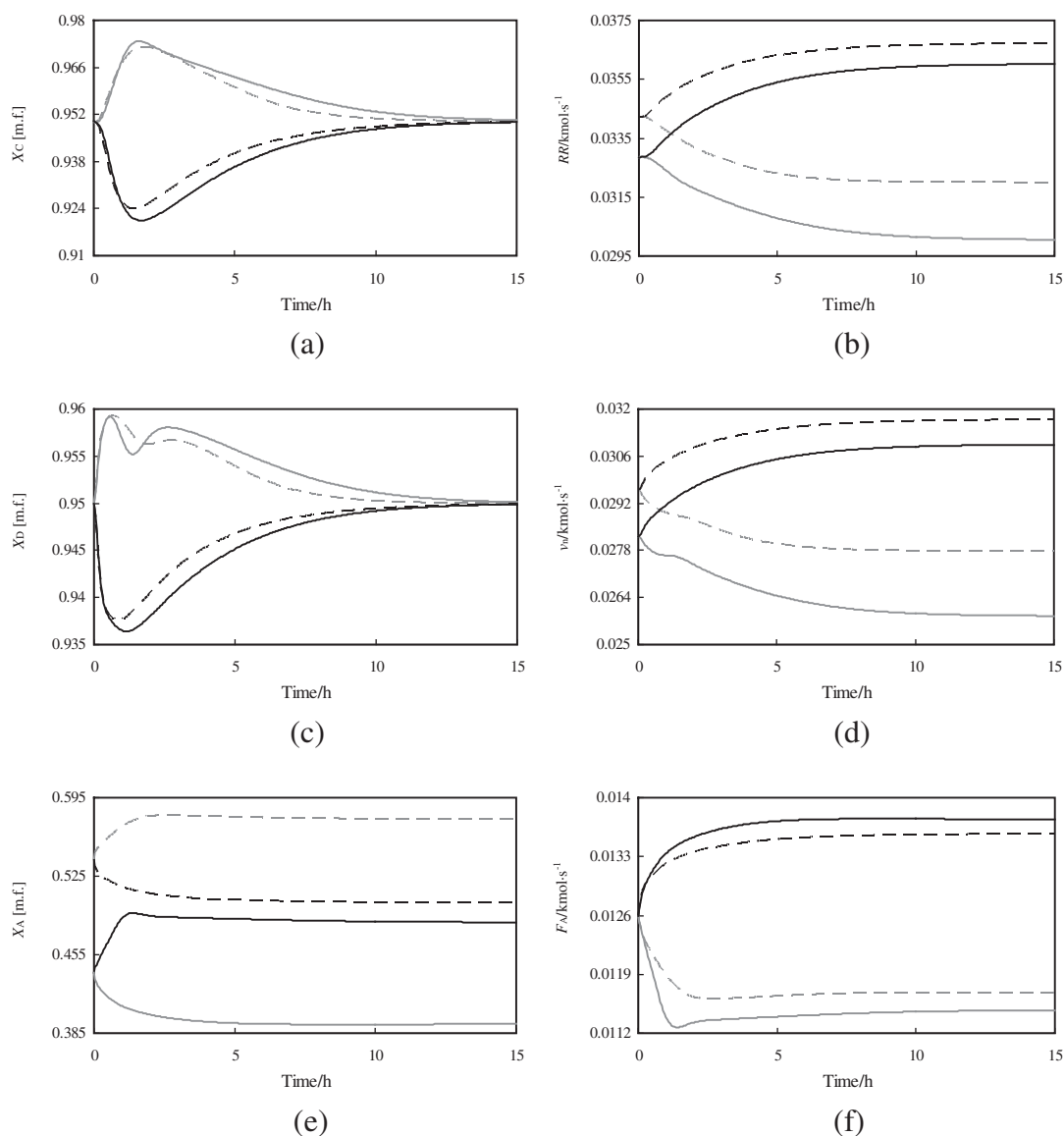


Fig. 17. Regulatory responses of the ideal reactive distillation columns (solid lines: $5/10/5^*$; dashed lines: $5/10/5$).

redistributing catalyst within the RXS, it should be taken into account in process development because its negative influence is usually small. With a careful use of these strategies, one can still get favorable impact on process dynamics and controllability besides their economical benefit. Figs. 18 and 19 compare, respectively, the servo and regulatory responses of the process designs, 5/10/5 and 5/10(0, −3)/7(3)*. The latter is derived with the combinatorial applications of the three strategies, and its static performance is also listed in Table 2. One can readily find that it is superior to 5/10/5 in not only thermodynamic efficiency but also process dynamics and controllability.

Although the above findings were derived based on the hypothetical ideal reactive distillation column studied, they are considered to be of general significance to the design and operation of other reactive distillation columns. This is because the dynamic effects of IHI by the three strategies are governed primarily by the interaction between the RO and the SO involved. Despite the fact that other design parameters may also affect process dynamics and controllability, their impacts are usually rather small as compared with the process structural variations caused by the application of these three strategies. Our research outcomes obtained so far are all in excellent accordance with the interpretation. It should be mentioned here that the reduction in the sizes of processes brought about by IHI has not been taken into account in the

current study. Since it can give positive influences to process dynamics and operability, it appears reasonable to deduce that all the findings obtained are on a more conservative stance.

9. Conclusions

In this article, the impact of the three strategies for the reinforcement of IHI, i.e., superimposing the RXS onto the SS, relocating feed stages, and redistributing catalyst, upon process dynamics and controllability are explored respectively based on the hypothetical ideal RDC involving an exothermic reaction ($A + B \leftrightarrow C + D$) with highly thermal effect. For superimposing the RXS onto the SS, favorable effect has been observed on process dynamics and operability and it should be considered in process development. For ascending the lower feed location, it can still be employed in process development despite its detrimental effect. For descending the upper feed location, it should be avoided in process development because of the introduced adverse IHI between the RXS and the RS. For the redistribution of catalyst in the RXS, it should be taken into account in process development because its negative influence is usually small. If carefully adopting these strategies for the reinforcement of IHI in a systematic manner, net favorable effect can still be expected on process dynamics and

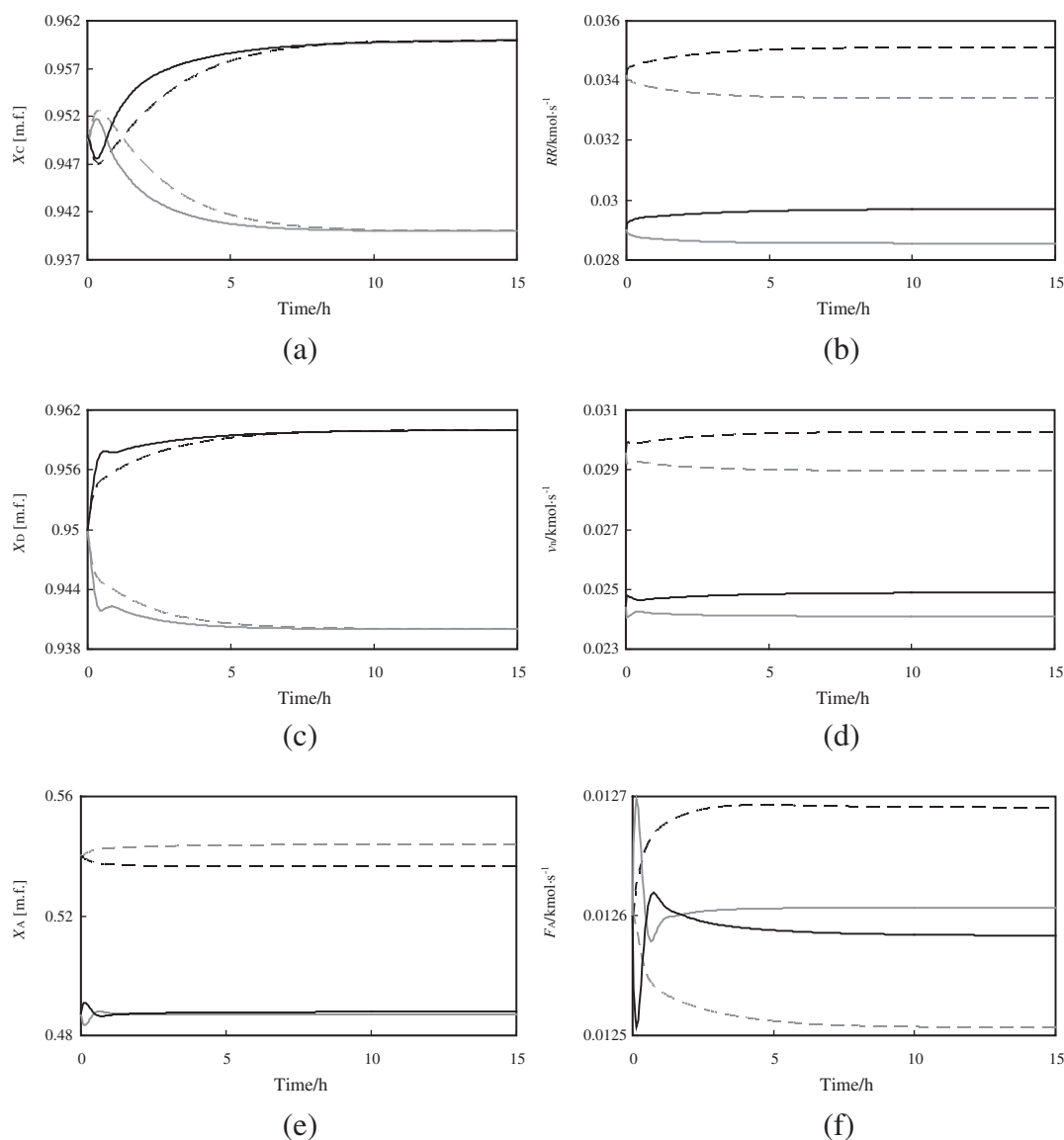


Fig. 18. Servo responses of the ideal reactive distillation columns (solid lines: 5/10(0, −3)/7(3)*; dashed lines: 5/10/5).

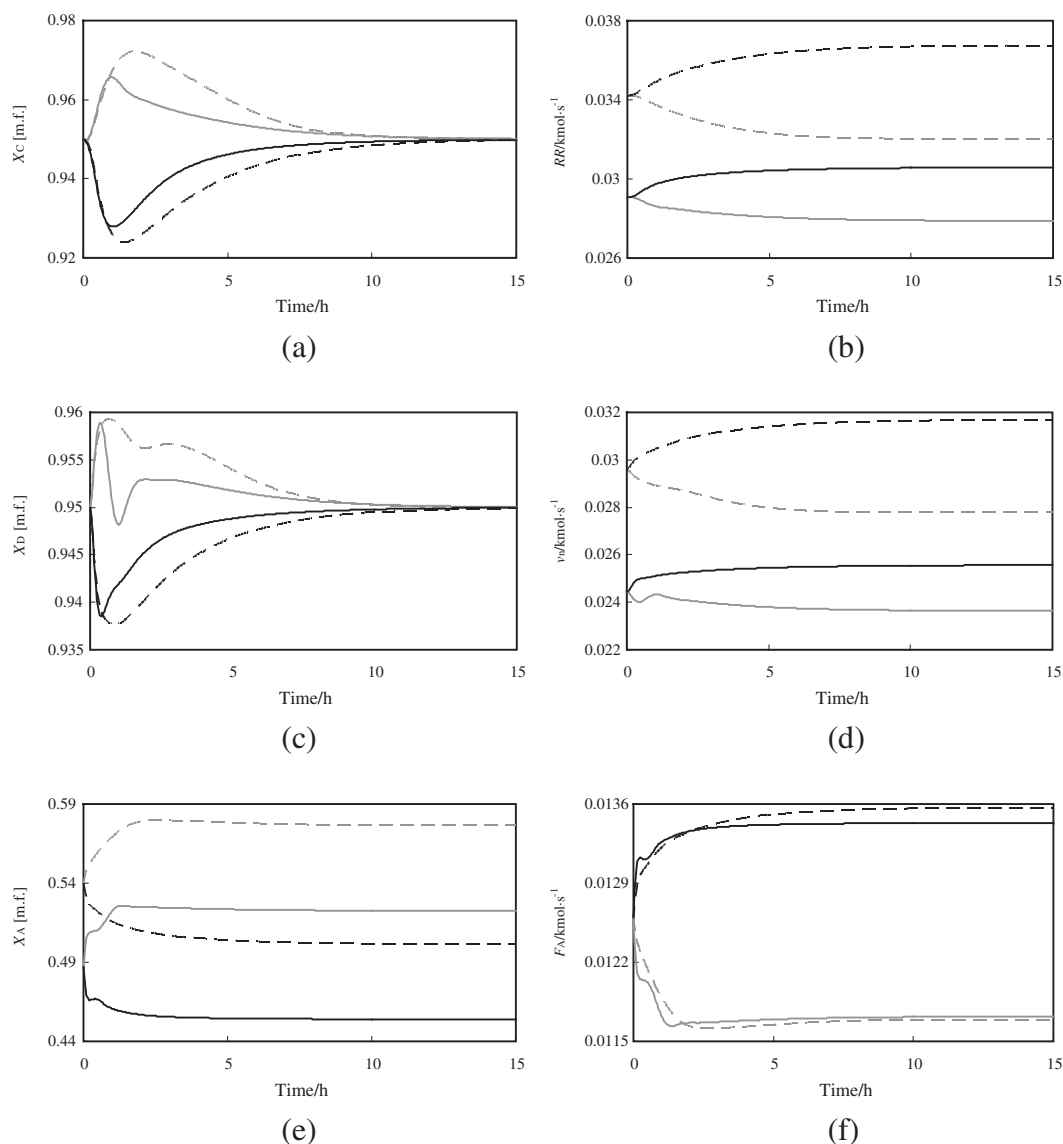


Fig. 19. Regulatory responses of the ideal reactive distillation columns (solid lines: 5/10(0, -3)/7(3)*; dashed lines: 5/10/5).

controllability. Although the conclusions are derived based on the hypothetical ideal reactive distillation column studied, they are considered to be of general significance to the design and operation of other reactive distillation columns.

Nomenclature

A_{vp}	vapor pressure constant, Pa
B_{vp}	vapor pressure constant, Pa·K
E	activation energy of a reaction, $\text{kJ} \cdot \text{kmol}^{-1}$
F	feed flow rate of reactants, $\text{kmol} \cdot \text{s}^{-1}$
H	stage holdup, kmol
ΔH_R	thermal heat of a reaction, $\text{kJ} \cdot \text{kmol}^{-1}$
ΔH_V	heat of vaporization, $\text{kJ} \cdot \text{kmol}^{-1}$
K	specific reaction rate, s^{-1}
K_C	proportional gain
K_{CU}	ultimate gain
L	liquid flow rate, $\text{kmol} \cdot \text{s}^{-1}$
n	number of stages
P	pressure, Pa
P_{CU}	ultimate period, s
Q	reaction heat load, kW
Q_{REB}	reboiler heat duty, kW

R	ideal gas law constant, $\text{kJ} \cdot \text{kmol}^{-1} \cdot \text{K}^{-1}$
r	net reaction rate, $\text{kmol} \cdot \text{s}^{-1}$
T	temperature, K
T_i	integral time, s
V	vapor flow rate, $\text{kmol} \cdot \text{s}^{-1}$
x	liquid composition
y	vapor composition
z	feed composition
α	pre-exponential factor
ν	stoichiometric coefficients of a reaction

Superscripts

s	saturation
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Subscripts

b	backward reaction
bot	bottom effluent
d	distillate
f	forward reaction
i	component index
j	stage index
m	feed stage index

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