Interaction of Separation and Reactive Stages on ETBE Reactive Distillation Columns

Budi H. Bisowarno, Yu-Chu Tian, and Moses O. Tadé
Dept. of Chemical Engineering, Curtin University of Technology, Perth WA 6845, Australia

Reactive distillation is a favorable alternative to conventional series of reactor-distillation processes for ether productions. However, the design of such columns is complicated due to the interaction between vapor-liquid equilibrium and reaction rates. There are conflicting reports on whether adding excessive separation stages degrade the column performance. A comparison is made of several designs of single and double feed reactive distillation columns for ETBE production to investigate the effects of separation and reaction stages on the overall performance. The explanations are presented using simulation results, whose mathematical models are written in the Aspen Plus environment. The results confirm that a conservative approach by adding extra separation and reaction stages can be applied to reactive distillation design. However, output multiplicity may be observed for longer column and should be considered in the early design phase. © 2004 American Institute of Chemical Engineers *AIChE J,* 50: 646-653, 2004

Keywords: reactive distillation, design (process simulation), simulation, process

Introduction

Reactive distillation, which integrates the functionality of distillation and reaction, is a favorable alternative to conventional series of reaction-separation processes. It can reduce capital investments through the reduction of the number of equipment such as pumps, piping, and so on. The operating costs are also reduced via overcoming distillation boundaries such as azeotrope due to the presence of reactions and lifting the reaction limitation such as equilibrium reaction due to the presence of continuous separation of the product (Malone and Doherty, 2000). The conceptual design of reactive distillation estimates the number of separation and reaction stages, feed flows and locations, energy loads, catalyst requirement, and so on. It has been investigated extensively and several methods are proposed. Geometric methods (Okasinski and Doherty, 1998), different-point methods (Lee et al., 2000b), and mathematical programming (Ciric and Gu, 1994; Cardoso et al., 2000) can be used to provide the basis for an economic evaluation of reactive distillation design.

However, reactive distillation design is still an open research area due to its complex interaction between vapor-liquid equilibrium and reaction rates. This interaction results in some counterintuitive behavior as reported in the open literature. Excessive increase of the separation stages was detrimental to the fractionation performance of ethyl *tert*-butyl ether (ETBE) reactive distillation column (Sneesby et al., 1998a). This work employed equilibrium reaction, which can be achieved by assuming high residence time or infinite volume of catalyst. Using the same assumptions, the negative effect of the excessive fractionation was also shown through the McCabe-Thiele diagram for binary reactive distillation (Lee et al., 2000a). The overall reactant conversion deteriorates as the reflux ratio increases for methyl acetate reactive distillation (Sawitoski and Pilavakis, 1988; Lee and Westerberg, 2001). This counterintuitive behaviour is not observed in the ternary reaction system such as ETBE and methyl *tert*-butyl ether (MTBE) productions (Lee and Westerberg, 2001). However, the additional separation stages were also thought not to degrade the column performance if the available degrees of freedom were appropriately chosen (Al-Arfaj and Luyben, 2000). If the reaction conversion and the product purity are specified, additional

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Correspondence concerning this article should be addressed to M. O. Tadé at m.tade@exchange.curtin.edu.au.

Current address of Y.-C. Tian: School of Software Eng. & Data Communications, Queensland University of Technology, GPO Box 2434, Brisbane QLD 4001, Australia.

separation stages do not deteriorate the column performance. This implies that the number of reactive stages with a sufficient amount of catalyst should be capable of maintaining certain conversion regardless of the number of separation stages. This work was demonstrated on ETBE and methyl acetate systems.

The conflicting effect of additional separation stages needs more investigations. If the additional separation stages do not degrade the column performance, the conservative approach of adding a few stages to the calculated theoretical stages can be applied for a reactive distillation column. However, this conservative approach cannot be used if the counterintuitive behavior is present. The reactant conversion, which was not kept constant as conducted in Sneesby et al. (1998a), indicated that the effect of additional separation stages on the fractionation performance cannot be concluded. Beside the additional separation stages, the reactive section and the interaction between the reactive and separation sections on the column performance should be simultaneously evaluated. On the other hand, keeping the constant conversion, as conducted in Al-Arfaj and Luyben (2000) can isolate the effect of additional separation stages on the fractionation performance. However, input multiplicity, which is essentially always present in reactive distillation (Sneesby et al., 1998b), implies that the same constant conversion may result from more than one set of operating conditions. Therefore, specifying the conversion and then comparing the product purity cannot be accepted as the basis of comparison.

This article discusses reactive distillation design with emphasis on the effect of the number of separation and reaction stages on the column performance. Both reboiler duty and reflux ratio are varied to obtain optimum ETBE purity in the bottom product, which is the observable parameter of the column performance. Interaction between separation and reaction, which causes input or/and output multiplicity, is also presented. It has been known that both multiplicity phenomena create difficulties in the design of a control system. ETBE reactive distillation columns consisting of reactive stages packed with internal catalyst, rectifying stages, and stripping stages are explored as the case study.

ETBE Reactive Distillation System

ETBE is synthesized from isobutylene and ethanol by using an acidic ion-exchange resin such as Amberlyst-15. This exothermic reaction is equilibrium limited in the industrial temperature range (40–70°C). The principal side reactions are dimerization of isobutylene and hydration of isobutylene. The former can be minimized by using excess ethanol, and the latter is neglected in the process analysis because it can only occur with the presence of water. The column performance is evaluated in both the presence and in the absence of the dimerization reaction. Although it is possible to perform all of the reactions inside the column, a pre-reactor is normally preferable because it can prevent catalyst deactivation. Besides, it is not easy to load and unload the internal catalyst. In this work, the existence of the pre-reactor is included by adding about 30% of ETBE in the feedstream, which is common in the commercial etherification processes. However, the absence of the pre-reactor is also simulated for comparison.

Although most etherification reactive distillation column employ a single feedstream, both single and double feed columns are simulated in this work. The single feed column contains catalyst on packed section above the feed point. For the double feed column, the reactive stages are located between the feed points. Ethanol stream is fed at the top of the reactive section, while the mixture of hydrocarbon containing isobutylene is fed at the bottom of the reactive section. Separation stages are required to be above and below the reactive section to produce necessary separation and to recycle the unreacted feed into the reactive section. The reactive distillation column for ETBE production is shown in Figures 1 and 2, respectively.

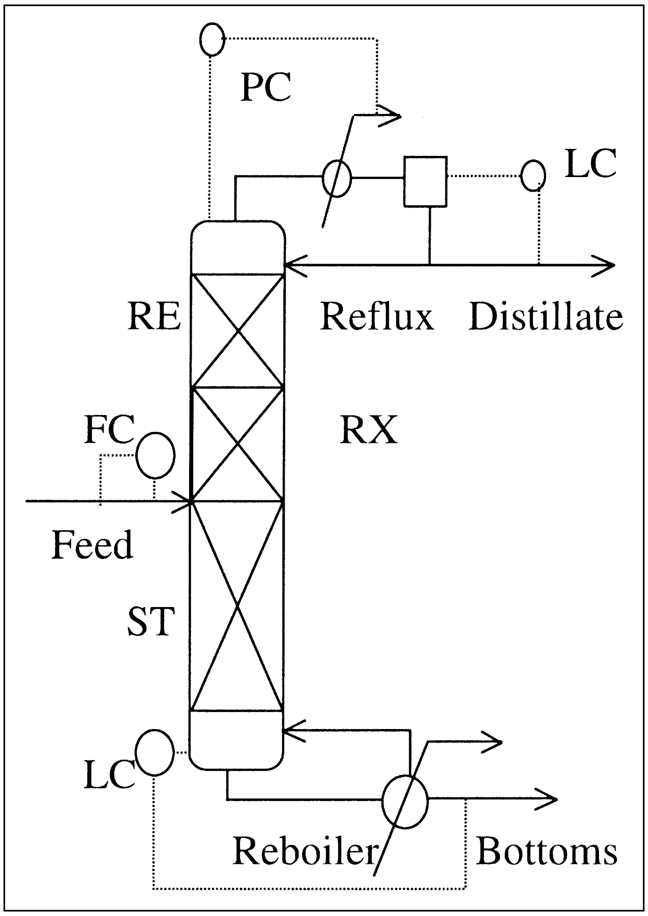

Figure 1. Single feed reactive distillation column for ETBE production.

The column aims to obtain high ETBE purity to eliminate high volatile components and high isobutylene conversion to maximize the value added by the process. Due to operational and control considerations, the reboiler duty is the main manipulated variable to optimize the ETBE purity while the reflux ratio or reflux rate is set constant. The overhead pressure is maintained constant by adjusting the condenser duty. The condenser and reboiler sump level are controlled by using the distillate and bottom flow rates, respectively. These control schemes [LV or (L/D)V] outperform other control schemes for setpoint tracking and disturbance rejection of single feed ETBE reactive distillation columns (Bisowarno and Tadé, 2002).

The steady-state model of the ETBE reactive distillation is implemented using Aspen Plus and employing RadFrac, which is based on an equilibrium-stage approach. This equilibrium approach, which was found to be satisfactory for MTBE reac-

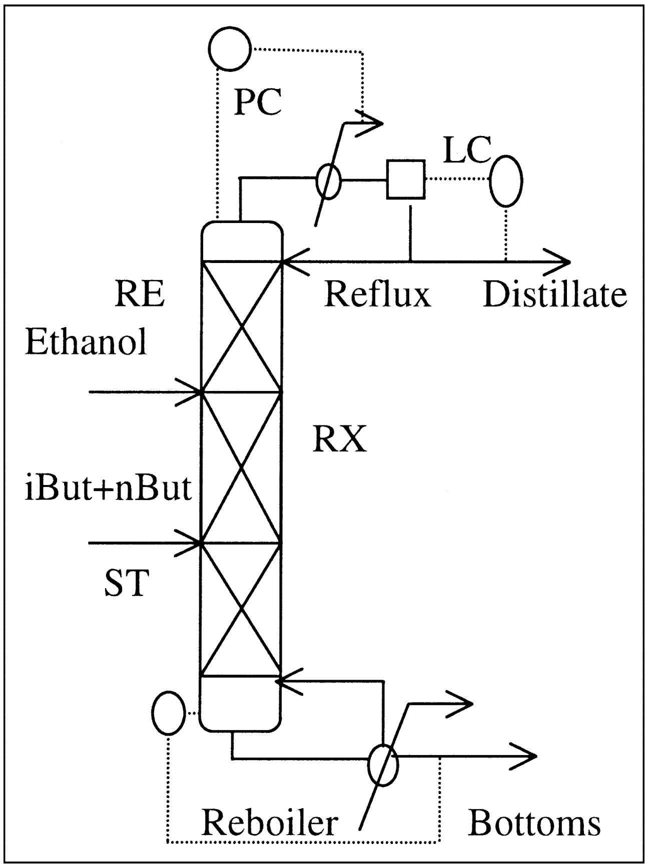

Figure 2. Double feed reactive distillation column for ETBE production.

tive distillation columns (Sundmacher et al., 1999), can be expected to produce similar results for the ETBE reactive distillation column. Both the ETBE synthesis and the dimerization of the isobutylene are assumed to achieve equilibrium instantly on each reactive stage. Although these assumptions may not represent the actual process, it can be used as the basis of evaluation and comparison of the overall performance. The details of the verified mathematical model for the ETBE reactive distillation can be found in a publication from our group (Sneesby et al., 1997). The UNIFAC model was used for liquid-phase activities even though the UNIQUAC model and Wilson equation can also be used to predict liquid phase activities in ether systems. The Soave-Redlich-Kwong (SRK) was used for fugacity coefficients, enthalpy, and other properties even though alternatives methods (such as Peng-Robinson) would also be acceptable for the pressure and temperature conditions. The equilibrium reaction expressions were adopted from Jensen and Datta (1995).

Single Feed Columns

The specifications and operating conditions of the single feed ETBE reactive distillation columns considered are shown in Table 1. The basic column (BS), which consists of 28 theoretical stages, including seven rectifying, seven reactive, and 14 stripping stages, respectively, has been previously simulated (Al-Arfaj and Luyben, 2000). The first column (SC-1) considers the conservative approach by adding more separation stages to the separation sections of the basic column. Additional stages may be required to compensate for such uncertainties as the vapor-liquid equilibrium model, efficiency estimation, and product quality requirement. The second column (SC-2) reduces the separation stages of the basic column, while keeping the reactive stages constant. For studying the effect of reactive stages, extra reactive stages are added to the basic column to obtain the third column (SC-3). Beside the additional amount of the catalyst, reactive stages could be added to the theoretical reactive stages to compensate for the uncertainties in the reaction kinetic models. All columns employ a total condenser and a partial reboiler.

In the previous work (Sneesby et al., 1998a) from our group, the composition of C4 components in the bottom product was specified and the optimized ETBE purity was obtained by varying the reboiler duty at constant reflux ratio. This work did not keep the conversion constant, thus the effects of additional separation stages on the column performance could not be concluded (Al-Arfaj and Luyben, 2000). In the present work, both reboiler duty and reflux ratio were varied to obtain the optimum ETBE purity. An overhead pressure of seven atm was selected, although wider operating pressures have been completed, which produced similar results. Although the results were obtained from constant feed rate, the same result can be expected for fixed production rate because almost all of the ETBE was withdrawn in the bottom product.

Multiplicity

Multiplicity in reactive distillation processes has been an active area of research in recent years. Three types of multiplicity phenomena (such as input, output, and pseudo), which occur in reactive distillation for ether productions, have been identified (Sneesby et al., 1998b). Their causes and effects on the operation and control were discussed. However, the presence and effects of the multiplicity phenomena at wider operating conditions in the design phase are still an interesting research area. Recently, the Damköhler number, which is a dimensionless parameter formulated as the ratio of a charac-

Table 1. Single Feed Reactive Distillation Column Characteristics and Inputs

Feed Conditions		Column Specifications	BS	SC-1	SC-2	SC-3
Temperature	30°C	Number of rectification stages	7	10	4	7
Rate	150 kmol/h	Number of reaction stages	7	7	7	10
Composition (mol)	9.1% EtOH	Number of stripping stages	14	17	11	14
	7.3% iBut	Total number of stages	28	34	22	31
	29.1% ETBE	Feed stage	16	19	13	19
	54.5% nBut	Overhead pressure (atm)	7			
Excess EtOH	5.0 mol. %	Reflux ratio	4, 5, 6, 7, 8			

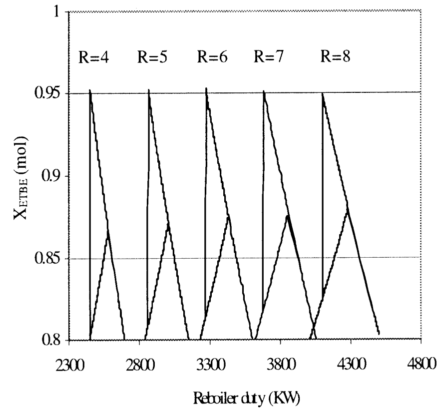

Figure 3. Multiplicity in the single feed basic column.

teristic liquid residence time to a characteristic reaction time, was used to investigate the cause of multiplicity (Chen et al., 2000, 2002; Mohl et al., 2001). Using MTBE reactive distillation system as the case study, an increase in the Damköhler number via decreasing the feed rate or increasing the number of catalytically active sites would shift the column from the kinetic regime into closer to chemical equilibrium and avoids the presence of output multiplicities (Mohl et al., 2001). The multiple (output) steady states of a MTBE reactive distillation column have also been shown to disappear under conditions of the column at the kinetic regime, that is, lower Damköhler number (Chen et al., 2000, 2002). The conflicting reports imply that the degree of reaction in the reactive section characterized by the Damköhler number may not the only deciding factor in the multiplicity. However, the input multiplicity, which is also affected by the changes in the Damköhler number, can still be observed at both kinetic and chemical equilibrium regimes.

For the basic column, the relationships between the ETBE purity (an output variable) and reboiler duty (an input variable) at different reflux ratio are shown in Figure 3. The figure shows the existence of input and output multiplicity. If the reboiler duty increases at constant reflux ratio, the ETBE purity increases to its optimum value before decreasing at higher reboiler duty. Decreasing the reboiler duty may result in different ETBE purity from that of increasing the reboiler duty so that the output multiplicity exists. The figure indicates that the operating conditions (such as reboiler duty at constant reflux rate/ratio) should be varied to optimize the ETBE purity. Table 2 shows the corresponding outputs of the basic column at different reflux ratios corresponding to the optimum reboiler duty. The values of the ETBE purity indicate that the reflux ratio and, consequently, the reboiler duty should be optimized to obtain a 'global' optimum ETBE purity. Table 3 shows the outputs at the global optimum of the reflux ratio and reboiler duty for the reactive distillation columns under consideration.

Compared with the previous results (Sneesby et al., 1998a; Al-Arfaj and Luyben, 2000), the ETBE purity of 83.60 mol. % can be obtained by using the operating conditions at the reflux ratio of 1.2 and the reboiler duty of 1424.26 kW (Sneesby et al., 1998a) or at the reflux ratio of 4.74 and the reboiler duty of 2692.47 kW (Al-Arfaj and Luyben, 2000). Figure 4 shows three reboiler duties, which can produce the specified ETBE purity at each constant reflux ratio. The global optimum ETBE purity for this basic column was obtained at the reflux ratio of 5.5 and the reboiler duty of 3,067 kW, which resulted in the ETBE purity of 95.316 mol. %, as shown in Table 3. Figure 4 also shows that different operating conditions can produce the same isobutylene conversion. Therefore, specifying the isobutylene conversion and then comparing the ETBE purity resulting from the reboiler duty, which is optimized at a constant reflux ratio, is not acceptable as the basis of comparison.

For a particular reactive distillation column, a specified ETBE purity may be obtained at different sets of reflux ratio and reboiler duty. This creates problems for process identification and control. This input multiplicity is essentially always present in reactive distillation columns, because the need to optimize the inputs results in conflicting effects on separation and reaction. Therefore, multiplicity in reactive distillation is more easily found due to the presence of chemical reactions than in the distillation column (Sneesby et al., 1998b). For all reactive distillation columns as shown in Table 3, the output multiplicity was also observed in the operating conditions considered. The window of the multiplicity phenomena is becoming larger as the internal rates (such as reflux ratio and reboiler duty) increase. In this work, the Damköhler number cannot be presented because of chemical equilibrium regime assumption. However, the lower internal rates, which are less likely to occur at the equilibrium regime (that is, lower Damköhler number), result in narrower window of multiplicity and, therefore, support the work of Mohl et al. (2001). The range of operating conditions, which produces output multiplicity and how to achieve and keep the tight range for the optimum ETBE purity, need to be investigated in the early design phase.

Table 2. Outputs of the Basic Reactive Distillation Column with Single Feed

Nre/Nrx/Nst		7/7/14				
Reflux ratio		4	5	6	7	8
Distillate (mol. %)	ETBE	0	0	0	0	0
	EtOH	0.010	0.019	0.024	0.009	0.005
	iBut	0.044	0.063	0.008	0.132	0.267
	nBut	99.946	99.918	99.968	99.859	99.728
Bottoms (mol. %)	ETBE	95.240	95.224	95.311	95.111	94.912
	EtOH	4.760	4.776	4.689	4.889	5.088
	iBut	0	0	0	0	0
	nBut	0	0	0	0	0
Reboiler duty (kW)		2452	2863	3272	3685	4100
IBut Conversion (mol. %)		99.93	99.91	99.99	99.80	99.60
Bottoms rate (kmol/h)		57.29	57.28	57.28	57.28	57.30
Output Multiplicity?		Yes	Yes	Yes	Yes	Yes

Table 3. Effect of Different Number of Separation and Reactive Stages (Optimum Output)

		Basic[*]		Basic[†]	SC-1	SC-2	SC-3
Nre/Nrx/Nst		7/7/14		7/7/14	10/7/17	4/7/11	7/10/14
Reflux ratio		5.5		10	7.8	5.3	5.9
Distillate (mol. %)	ETBE	0	0	0	0	0	0
	EtOH	0.026	0.03	0.001	0.006	0.064	0.009
	iBut	0.005	0.01	0.25	0.012	0.005	0.002
	nBut	99.969	99.96	99.75	99.982	99.931	99.989
	DIB	—	0	—			
Bottoms (mol. %)	ETBE	95.316	95.22	94.92	95.280	95.369	95.298
	EtOH	4.684	4.75	5.08	4.720	4.631	4.702
	iBut	0	0	0	0	0	0
	nBut	0	0	0	0	0	0
	DIB	—	0.03	—			
Reboiler duty (kW)		3067	3067	4515	4009	2987	3230
IBut Conv. (mol. %)		99.99	99.92	99.63	99.98	99.99	100
Bottoms (kmol/h)		57.28	57.29	57.32	57.29	57.25	57.29
Output Multiplicity?		Yes	Yes	Yes	Yes	Yes	Yes
Top reactive temp. (°C)		55.87	55.88	55.79	55.86	55.86	55.81
Bottom Reactive temp. (°C)		72.11	81.87	77.77	83.41	69.66	75.99
R_{ETBE} bottom react. section, mol/s		0.0008	−0.0014	0.007	−0.0003	0.00124	−0.0001
$K_{\mathrm{ETBE\text{-}EtOH}}$ bottom react. section		2.07	1.63	2.51	1.48	2.28	1.81

[*] Comparison with and without the existence of isobutylene dimerization.

[†] The pre-reactor was not used, no ETBE in the feedstream.

Effects of the Number of Stages

For distillation column, the separation performance can normally be increased through increasing theoretical stages or increasing the internal vapor and liquid flows, except in some azeotropic columns (Knapp and Doherty, 1994). The performance can be observed from the increasing purity of at least one of the distillation products. However, this conservative approach may not be applied in reactive distillation design due to the counter-intuitive behavior (Al-Arfaj and Luyben, 2000).

The effect of a different number of reactive and separation stages are presented in Table 3. The presented outputs resulted from using the optimum reflux ratio and reboiler duty. The optimum reboiler duty was approached from the larger values due to the presence of the output multiplicity. For all columns, the ETBE purities are above 95 mol. %, mixed mainly with the unreacted ethanol. The distillate is almost pure *n*-butane so that the isobutylene is nearly converted completely to ETBE.

Comparing the first column (SC-1) to the basic one, the additional separation stages can maintain relatively constant optimum ETBE purity by using larger internal rates (such as higher reflux ratio and reboiler duty). Although conventional approach by adding a few number of separation stages can be applied, it requires higher energy consumption to maintain a similar performance. The high reboiler duty produced high reactive temperature, which reversed the ETBE product into the reactants in the bottom reactive section. However, the additional stripping stages could maintain quite similar ETBE purity to that of the basic column. The second column (SC-2), which employs less separation stages, shows that shorter column may be applied to produce relatively similar ETBE purity by using both lower reflux ratio and reboiler duty. The overall isobutylene conversion is also nearly completed. The third column (SC-3) employs an equal number of separation stages to the basic column with longer reactive section. Although increasing the number of the reactive section may promote further reaction, the limited stripping stages could reduce the ETBE purity.

Table 3 also shows the effect of isobutylene dimerization on the basic column performance. It increased the reactive section temperature and decomposed the existing ETBE in the bottom

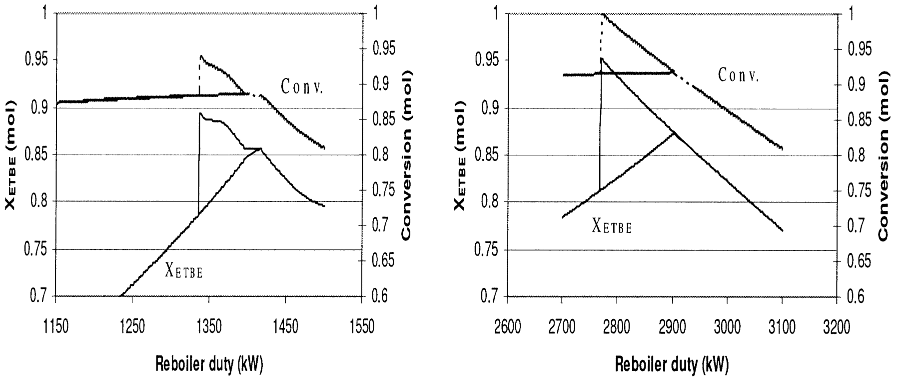

Figure 4. Comparing the multiplicity at R = 1.2 (left) and R = 4.74.

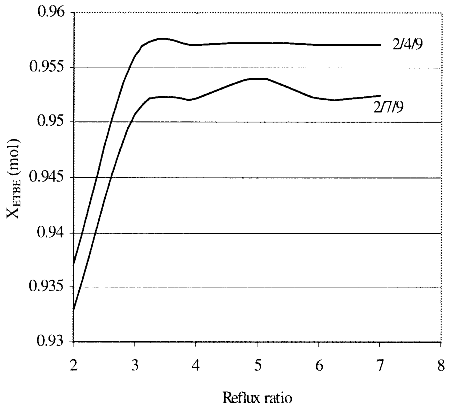

Figure 5. Effect of the optimum operating condition on shorter columns.

reactive section. Therefore, the ETBE purity degraded in the bottom product. Table 3 also shows that this single feed column is capable of producing ETBE without the introduction of the pre-reactor. However, higher internal rates were needed to produce comparable results. This result indicates that the associated problems of the internal catalysts including its construction are critical issues to be addressed.

When considering the outputs of the second column (SC-2), further reductions of the number of the reactive and separation stages were employed. Figure 5 shows the relationship between the ETBE purity and the reflux ratio for the other two columns. The first column reduces the number of the separation stages while keeping the number of reactive stages constant and the second one reduces both the separation and the reactive stages. The figure shows that increasing the reflux ratio and, consequently, the reboiler duty does not degrade the column performance. The graphical method has also been used to show that increasing the reflux rate does not deteriorate the column performance for ternary systems such as MTBE and ETBE production (Lee and Westerberg, 2001).

Overall, the conservative approach of adding a few extra stages can be applied to the reactive distillation design. However, increasing the number of separation stages imposes more difficulties on the operation and control considerations due to the wider window of multiplicity. These simulation results also show that the optimum operating conditions (reflux ratio and reboiler duty) of the shorter column can produce comparable performance to that of the longer columns. For shorter columns, increasing the reflux ratio and, consequently, the reboiler duty does not reduce the ETBE purity. It also consumes less energy due to the lower reflux ratio and the reboiler duty.

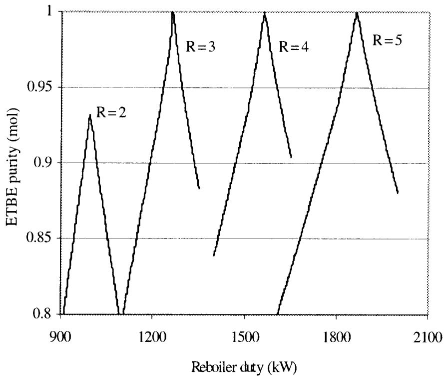

Figure 6. Multiplicity in the double feed basic column.

Double Feed Columns

The specifications of the double feed ETBE reactive distillation columns are shown in Table 4. The basic column consists of 30 theoretical stages including 10 rectifying, 10 reactive, and 10 stripping stages, respectively. The next two columns considered, which were designed to study the effect of separation stages, have been previously simulated (Al-Arfaj and Luyben, 2000). The third column is used to study the effect of additional reactive stages on the column performance. All columns employ a total condenser and a partial reboiler.

In this simulation, both the reflux ratio and the reboiler duty were varied to obtain the optimum ETBE purity. Although wider operating pressures were studied, the overhead pressure of 7 atm was again selected for illustrating the results. The other pressures produced similar results.

Multiplicity

For the basic column (DB), the existence of multiplicity is shown in Figure 6. Varying the reboiler duty at constant reflux ratio/rate always revealed the input multiplicity. However, the output multiplicity was not observed in the operating conditions (up to reflux ratio of 8) considered. The figure indicates that the reboiler duty should be varied to optimize the ETBE purity at a constant reflux ratio. Table 5 presents the outputs of the basic column at different reflux ratio obtained at its optimum reboiler duty. Increasing the reflux ratio and, consequently, its corresponding reboiler duty does not degrade the ETBE purity. Table 6 presents the outputs of the columns considered here at the "minimum" reflux ratio, whose larger value does not significantly increase the ETBE purity obtained at its optimum reboiler duty.

Similar to the input multiplicity of ETBE purity and isobu-

Table 4. Double Feed Reactive Distillation Column Characteristics and Inputs

Feed Conditions		Column Specifications	BD	DC-1	DC-2	DC-3
Temperature	30°C	Number of rectification stages	10	15	5	10
Rate:		Number of reaction stages	10	10	10	13
EtOH	40 kmol/h	Number of stripping stages	10	15	5	10
HC	100 kmol/h	Total number of stages	30	40	20	33
	40% iBut	Feed stage: EtOH	11	16	6	11
	60% nBut	iC4 + nC4	22	27	17	25
		Overhead pressure (atm)	7			
		Reflux ratio	2, 3, 4, 5			

Table 5. Optimal Outputs of the ETBE Reactive Distillation Columns with Double Feed

Nre/Nrx/Nst		10/10/10			
Reflux ratio		2	3	4	5
Distillate (mol. %)	ETBE	0	0	0	0
	EtOH	0.34	0.17	0.14	0.01
	iBut	4.44	0.18	1.84	0.15
	nBut	95.23	99.65	98.02	99.74
Bottoms (mol. %)	ETBE	93.24	99.97	97.39	99.96
	EtOH	6.54	0.02	2.61	0.04
	iBut	0.07	0.005	0	0
	nBut	0.14	0.005	0	0
Reboiler duty (kW)		993	1263	1582	1864
IBut Conversion (mol. %)		92.95	99.73	97.19	99.78
Bottoms rate (kmol/h)		39.88	39.90	39.92	39.93
Output Multiplicity?		No	No	No	No

tylene conversion shown in Figure 4 for single-feed reactive distillation column, Figure 6 also implies that certain isobutylene conversion may result from several sets of reflux ratio and reboiler duty. Therefore, specifying the isobutylene conversion and then comparing the ETBE purity resulting from the reboiler duty, which is optimized at a constant reflux ratio, is not acceptable for the double feed reactive distillation column.

Table 6 also shows that additional separation stages may produce output multiplicity, which is not observed in the shorter columns. In the first column (DC-1), the window of multiplicity is larger if the internal rates (such as higher reflux ratio and reboiler duty) are increased. The second (DC-2) and third columns (DC-3) do not reveal the output multiplicity. As in the single feed columns, the input multiplicity is always found in the double feed columns.

Effects of the number of stages

Comparing the first column (DC-1) to the basic one (DB), the introduction of additional separation stages does not degrade the column performance. The ETBE purity can be maintained at its high value by using higher internal rates. Longer separation sections can purify further the *n*-butane and ETBE, respectively, from the unreacted feed compared to that of the basic column. However, this first column (DC-1) produced output multiplicity and the optimum reboiler duty should be approached from its larger value. The second column (DC-2) confirms that the shorter column may be employed to obtain similar ETBE purity by using lower reflux ratio and reboiler duty. The isobutylene was also almost completely converted to ETBE. Output multiplicity is not observed in this second column (DC-2). The third column (DC-3), where three reactive stages were added to the basic column while keeping the separation stages constant, shows the effect of reactive stages on the column performance. The unreacted isobutylene, which was withdrawn from the bottom product in the basic design, reacted further to form ETBE.

In general, the ETBE purity and isobutylene conversion increase as the reflux ratio and, consequently, the reboiler duty increase. Although adding separation stages does not degrade the overall performance, a shorter column with appropriately chosen operating condition is preferable. Longer columns especially at high internal rates can result in output multiplicity, which eventually create difficulties on operation and control considerations.

Conclusions

This article confirms that the conservative approach of adding a few stages to the calculated theoretical stages can be applied in reactive distillation columns, because they do not degrade the column performance if the operating conditions are chosen appropriately. However, the longer column, which consequently demands larger reflux ratio/rate and reboiler duty, may produce output multiplicity. For the single feed column, longer column requires both reflux ratio and reboiler duty to be adjusted to optimize the ETBE purity, while the increasing reflux ratio does not reduce the ETBE purity for the shorter columns. The shorter column can produce comparable performance to that of the longer column by using less energy due to

Table 6. Effect of Different Number of Separation and Reactive Stages (Optimum Output)

		Basic	DC-1	DC-2	DC-3
Nre/Nrx/Nst		10/10/10	15/10/15	5/10/5	10/13/10
Reflux ratio		6.0	7.0	5.0	4.0
Distillate (mol. %)	ETBE	0	0	0	0
	EtOH	0.10	0.01	0.61	0.14
	iBut	0.09	0.01	0.55	0.18
	nBut	99.81	99.98	98.84	99.68
Bottoms (mol. %)	ETBE	99.95	99.98	99.74	99.96
	EtOH	0	0	0.01	0.04
	iBut	0.02	0.01	0.10	0
	nBut	0.03	0.01	0.15	0
Reboiler duty (kW)		2163	2458	1888	1564
IBut Conversion (mol. %)		99.85	99.98	99.06	99.73
Bottoms rate (kmol/h)		39.96	40.00	39.73	39.95
Output Multiplicity?		No	Yes	No	No

the lower reflux ratio and the reboiler duty. For the double feed column, increasing reflux ratio does not degrade the column performance as well. This should be considered in the early design phase. Both single and double feed columns can be employed to produce ETBE without a pre-reactor. However, the related problems of the catalyst should be addressed.

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