COORDINATED CONTROL OF REACTIVE DISTILLATION SYSTEMS BY MANIPULATING FEED LOCATIONS

Yu-Cheng Cheng and Cheng-Ching Yu[*]

Dept. of Chem. Eng., National Taiwan University
Taipei 106-17, TAIWAN

Abstract: The effects of feed locations to the control of reactive distillation are explored. In this work, ideal reactive distillation systems are used to illustrate the advantage of feed trays optimisations in control. Because the temperature and composition profiles are critical for an effective utilization of the reactive zone, the optimal feed locations are essential for improved performance. Quantitatively, a systematic procedure is proposed to find the right feed trays. Finally, the idea of optimal feed trays is extended to the operation/control of reactive distillation systems. First, steady-state analysis is carried out to find the optimal feed trays as measurable load variable varies. Then, a control structure is proposed to rearrange the feeds as the disturbance comes into the system. The results indicate that, again, substantial energy can be saved by feed rearrangement via the coordinated control structure. *Copyright © 2005 IFAC*

Keywords: reactive distillation, feed locations, process design and control.

1. INTRODUCTION

The reactive distillation combines both chemical reaction and multicomponent separation in a single unit. It offers significant economic advantages in some systems, particularly when reactions are reversible or when the presence of azeotropes makes conventional separation systems complex and expensive. The applications of reactive distillation in the chemical and petroleum industries have increased rapidly in the past decade (Taylor and Krishna, 2000; Doherty and Malone, 2001). A number of papers and patents have explored in the RD systems. The literature up to 1992 was reviewed by Doherty and Buzad. Most the papers were discussed by steady-state design and optimization problems. Only a few papers studied the dynamic of reactive distillation or the interaction between design and control. Recent books by Doherty and Malone (2001) and Sundmacher and Kienle (2003) present detailed discussions of the technology and its current status.

The literatures state that the most common applications of reactive distillation are etherification and esterification reactions. Most of these papers focus on real chemical systems and each system has its own set of complexities in vapor-liquid equilibrium nonideality (azeotropes), reaction kinetics, physical properties, etc. The discrete nature of chemical species and specific complexities in the VLE seem to cloud the picture in understanding reactive distillation systems. On the other hand, the ideal reactive distillation of Al-Arfaj and Luyben (2000) seems to offer a continue spectrum in studying the process behavior by stripping away all the non-ideal VLE and specific reaction rates. Only a limited number of papers study the ideal reactive distillation systems. Al-Arfaj and Luyben (2000) studied the control of an ideal two products reactive distillation system. Simple ideal physical properties and kinetics are assumed so that the control issue can be explored without being clouded by complexities of a specific chemical system. Sundmacher and Qi (2003) also

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[*] corresponding author; fax: +886-2-2362-3040, e-mail: ccyu@ntu.edu.tw

compare the conceptual design of reactive distillation process configurations for ideal binary mixtures and comparison is made to the conventional process. A recent paper by Kaymak and Luyben (2004) also makes quantitative comparison of simple reactive distillation for different chemical equilibrium constants and relative volatilities.

The reactive distillation differs from the conventional distillation in that a tubular type of reactor, the reactive flash cascades to be specific (Doherty and Malone, 2001), is cascaded with separation units. From this perspective, the composition profile inside the reactive zone becomes important for an effective operation of the reactive flash cascades. Moreover, typical distillation columns follow certain temperature profile. That is the temperature increases as one steps down the column. The *composition* as well as the *temperature* effects should play some role for the performance of a reactive distillation column and the reactant feed location is an obvious design degree of freedom to locate optimal composition and temperatures profiles inside the column. It then becomes obvious that the feed tray location should be included as a design variable. Therefore, the objective of this work is to explore the effects of feed tray location to the performance of reactive distillation systems.

In this work, it is necessary to rearrange the feed tray locations to obtain optimal design (i.e., minimum energy consumption). Therefore, the optimized feed tray location can be extended to the operation aspect of reactive distillation systems. Control structures are devised to maintain optimal composition and temperature profiles as operating condition change

2. PROCESS STUDIES

Consider an ideal reactive distillation (Fig. 1) with a reversible liquid-phase reaction in the reactive section.

$$A + B \Leftrightarrow C + D$$

The forward and backward specific rates following the Arrhenius law on tray j are given by

$$k_{Fj} = a_F e^{-E_F / RT_j} \qquad (1)$$

$$k_{Bj} = a_B e^{-E_B / RT_j} \qquad (2)$$

where a_F and a_B are the pre-exponential factors, E_F and E_B are the activation energies, and T_j is the absolute temperature on tray j. The reaction rate on tray j can be expressed in terms of mole fractions ($x_{j,i}$) and the liquid holdups (M_j).

$$\Re_{j,i} = \nu_i M_j (k_{Fj} x_{j,A} x_{j,B} - k_{Bj} x_{j,C} x_{j,D}) \qquad (3)$$

where $R_{j,i}$ is the reaction rate of component i on the jth tray (kmol/s), ν_i is the stoichiometric coefficient which takes negative value for the reactants, and M_j is the kinetic holdup on tray j (kmol) and it takes a constant value throughout the simulation (This is typically true for catalyst weight based kinetics). In this work, the forward reaction rate is specified as 0.008 kmol/s at 366 K and k_B is set to 0.004 kmol/s at the same temperature. Kinetic and physical property data for the system are given in Table 1 (Al-Arfaj and Luyben, 2000).

As shown in the Figure 1, the column is divided into three sections. The first one is the reactive section containing N_{rxn} trays. The rectifying section above the reactive section has N_R trays and the stripping section, below the reactive zone, has N_S trays. Thus, we are considering a reactive distillation column in which reaction only occurs in the reactive section, which implies a solid-catalysis catalyzed reaction.
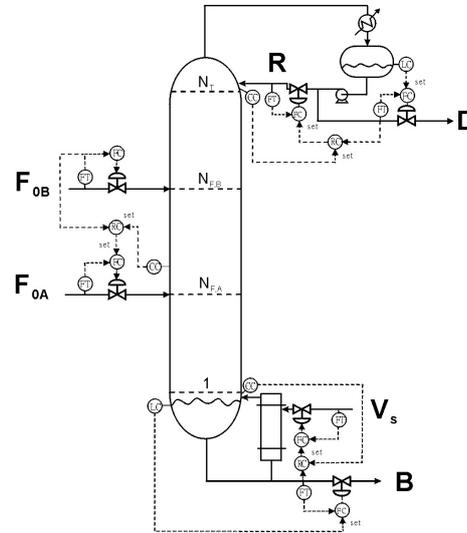

Fig. 1 Control structure of reactive distillation with fixed feed locations.

In this system, ideal vapor-liquid equilibrium is assumed and constant relative volatilities are used. The relative volatilities of the components are in the following order:

$$\alpha_C > \alpha_A > \alpha_B > \alpha_D$$

The products C and D are the lightest and heaviest components, respectively, with the reactants A and B as middle boilers. The thermodynamic behavior indicates that we should remove the product C from the distillate and obtain heavy product D from the bottoms. Figure 1 also shows that the fresh feed stream F_{OA} containing reactant A is fed to the bottom of the reactive zone, and the heavier reactant B is fed to the top of the reactive zone. Because the light reactant A is quite volatile as compared to B and D, it goes up the column and there is little component A

left in the stripping section. Likewise, heavy reactant B goes down the column after being fed on the top tray of the reactive zone, and little component B can be found in the rectifying section. Thus, the primary separation in the stripping section is between B and D and in the rectifying section is between C and A.

2.1 Base case

Equations described the material balances were programmed in FORTRAN code and all simulations were carried out on Pentium PC. It should be emphasized that the convergence of the reactive distillation is far more difficult than conventional distillation. Typically, a steady-state simulation is carried out in a two-step procdure. First, the Wang-Henke method is used to converge the flowsheet (MESH equations) to a certain degree (actually to the point the objective function fluctuates). Then, the temperature and composition profiles are fed to a dynamic program which is integrated until temperatures and compositions converge.

Saturated liquid feeds were assumed and two feed flow rates are 0.0126 kmol/s each with pure A ($N_{F,A}$) or B ($N_{F,B}$) which was introduced to the bottom ($N_{rxn,bot}$) or the top ($N_{rxn,top}$) of the reactive section (see Fig. 1). Note that this is the typical feed arrangement for reactive distillation which is term as the *conventional* feed arrangement hereafter. In this work, the conversion is specified to be 95% and this corresponds to purities of 95% C in the distillation and 95% D in the bottoms. In this work, a holdup (M_j) of 1000 moles is assumed.

2.2 Feed Locations versus Reactants Distribution

It should be emphasized that the reactive section of a reactive distillation column can be viewed as cascade type two-phase reactors with the reactor temperature determined by the bubble point temperature of the tray liquid phase composition. It is clear that the composition and temperature profiles will certainly affect the performance of the reactive zone and the feed tray locations appear to be one of the most effective variables for these profiles redistribution. In this section, we are interested in how the composition profile will be affected by changing the feed tray location and the individual feed tray is changed one at a time.

First we fix the $N_{F,A}$ at the bottom of the reactive zone and change the feed location of B component from top to bottom. As the feed location of component B moves down the column, the mole fraction of heavy reactant B (x_B) increases toward the lower section of reactive zone. That means we have wider and relatively more uniform distribution of component B throughout the reactive zone as the feed tray is lowered. Consequently, the mole fraction of the light reactant A (x_A) becomes smaller in the lower

reactive zone while the profiles of two products (x_C & x_D) remain qualitatively similar. This rearrangement of the reactant composition certainly alters the fraction of conversion as well as the temperature profile in the reactive zone. When we move $N_{F,B}$ down, both the reactant B and conversion increase in the lower reactive section. This implies the lower reactive trays are better utilized, but at the cost of smaller conversion in the upper reactive trays. It seems a balanced usage of the reactive trays is necessary to achieve optimality and this means an optimal feed location exists for component B. The energy consumption (vapor rate V_s to be exact) is a good measure the column performance. The results clearly indicate the energy penalty can be significant if one places the feed at inappropriate location and the conventional design seems to be a pretty good choice.

The same analysis can be carried over to the feed location of the light component A. Now we fix the $N_{F,B}$ at the top reactive zone (i.e., $N_{F,B}=N_{rxn,top}$) by varying $N_{F,A}$. The on-going analysis clearly indicates that the feed tray locations are important design/operation parameter and improved process design can be achieved by simply adjusting the feed locations. One question then arises: how much energy can be saved if we adjust the feed locations simultaneously.

2.3 Optimal Feed Locations

Finding the optimal feed locations can be formulated as an optimization problem where the vapor rate is minimized by varying the feed tray locations.

$$\min_{N_{F,B}, N_{F,A}} V_S \qquad (4)$$

Because the total tray number (N_T) is finite, one can find the optima by exhausting all N_T^2 possibilities. It is reasonable to restrict the search space to the reactive zone and the possible choices are further reduced to N_{rxn}^2. In this work, a brute force approach is taken by fixing $N_{F,A}$ first while varying $N_{F,B}$ until a minimum V_S is found. Next, $N_{F,A}$ is changed and the procedure repeats itself until a global minimum is located. The results indicates that one should move the feed location of the heavy reactant B down to $N_{F,B}$=15 (from 19) and move the feed tray of the light reactant A up to $N_{F,A}$=11 (from 9). This corresponds to 10.9% of energy saving as compared to the conventional feed arrangement. Furthermore, experience from this and many other examples reveals that the feed location of the heavy reactant should *not* be placed *lower* the feed tray of the light reactant. This reduces the search space further down to $(N_{rxn}+1)N_{rxn}/2$.

In addition to the percentage of energy saving, comparison is also made in terms of profiles of temperature, composition, fraction of conversion, and reaction rate constants. The result shows that the case of optimal feed arrangement has a much *sharper* temperature profile in the reactive zone than the case of conventional feed locations. Furthermore, the profiles of tray conversion and rate constant also take qualitatively similar shape as that of the temperature.

In summary, for the system with relative volatilities of $\alpha_C/\alpha_A/\alpha_B/\alpha_D=8/4/2/1$, one should move the feed locations of the heavy reactant downward and light reactant upward. In terms of the search space for the optimal feed trays, we have the following heuristics:

Heuristic H1: Never place the heavy reactant feed below the feed tray of the light reactant (similarly, do not place the light reactant feed above the feed tray of the heavy reactant).

3. OPERATION AND CONTROL

On-going analyses clearly show that improved design can be achieved by treating the feed tray location as an optimization variable and results indicate that significant energy saving can be obtained by simply rearranging the feeds. However, these analyses are limited to the design aspects with different thermodynamics parameters (i.e., relative volatilities) as well as kinetics parameters (i.e., activation energy and pre-exponential factor). In this section, we are more interested in how this finding will affect the operation and control of reactive distillation.

Despite clear economic incentives of reactive distillation, only a few papers studying the dynamics and control of reactive distillation have been published. Al-Arfaj and Luyben (2000) give a review on the closed-loop control of reactive distillation. They presented a comparison of several control structures for an ideal two-product reactive distillation system as well as several real chemical systems (Al-Arfaj and Luyben, 2002 & 2004). One important principle in the control of reactive distillation is that we need to control one intermediate composition (or tray temperature) in order to maintain stoichiometric balance (Al-Arfaj and Luyben, 2000).

3.1 Optimal feed location under production rate variation

In general, production rate variation is one of the most important load disturbance in plantwide control and operation (Luyben et al. 1999) and, more importantly, it can be measured. In this work, we are interested in whether significant energy saving can be obtained by adjusting the feed tray locations as the production rate changes. If appreciable amount of operating cost can be reduced, the feed tray location is not only dominant design variable but useful manipulated variable for control.

Let us take the base case as an example, the optimal feed trays are: $N_{F,A}=11$ and $N_{F,B}=15$ (4 trays apart). The control objective is to maintain the product compositions (C & D) at 95%. Both positive and negative production rate variations are explored. First, consider the case with +40% feed flow rate increase. The optimization is performed to find the optimal feed locations by minimizing the vapor rate. One obtains $N_{F,A}=10$ and $N_{F,B}=16$ (6 trays apart) and this corresponds to a 28% of energy saving! This is not totally unexpected, because an increasing the production can be viewed, in a sense, as a decrease in the rate constant. Therefore, we should move the feed trays away from each other. But the percentage of energy saving is larger than our expectation. Next, the optimization is carried out for a -40% change in the feed flow rate. The optimal feed trays become $N_{F,A}=12$ and $N_{F,B}=15$ (3 trays apart) and a 9% saving in the vapor rate is observed. As pointed earlier, this has the same effect as that from reaction rate increases and one should moves the feed trays closer to each other. The results clearly indicate that one should change the feed tray locations as the production rate changes, because 9 or 28% energy can be saved by simply moving the feed trays. The next question then becomes how can we implement such a control strategy? The coordinated control of Doukas and Luyben (1976) offers some light in this direction (Chang et al., 1998).

3.2 Control structure

Before getting into the feed rearrangement control structure, let us first construct the fundamental control configuration for the reactive distillation with two feeds. Recall that, unlike the control of conventional distillation, one needs to control an internal composition (or temperature) to maintain stoichiometric amounts of the two fresh feeds (Al-Arafaj and Luyben, 2000). For the purpose of illustration, in this work, we choose to control composition of the reactant A on tray 13 where a significant break in the composition A is observed (Fig. 6B). Thus, we have three compositions to be controlled, top composition of C, bottoms composition of D and composition A on tray 13. For the manipulated variables, the ratio scheme is used and these three ratios are: reflux ratio, boilup ratio, and feed ratio. Figure 1 shows the control configuration for the reactive distillation without feed rearrangement.

(1) The fresh feed F_{OB} is the throughput manipulator which is flow control.

(2) F_{OA} is ratioed to F_{OB} and the ratio is set by the tray 13 composition (x_A).

(3) The top composition of C is maintained by changing the reflux ratio.

(4) The bottoms composition of D is controlled by changing the boilup ratio.

(5) The base level is controlled by manipulating bottoms flow rate.

(6) The reflux drum level is maintained by adjusting the distillate flow rate.

This structure consists of 3 composition loops and 2 level loops. In this paper, decentralized control structure with PI controllers is employed for the composition loops and perfect level control is assumed for the level loops. In the identification phase, the relay feedback method (Yu, 1999) is used to obtain the ultimate gain and ultimate period and the controllers are tuned using the Tyreus–Luyben turning method (Tyreus and Luyben, 1992). Note that five minutes of analyzer dead time was assumed for the composition measurement.

Because both fresh feed flows are measured, one can coordinate the feed location as the production rate changes. Let us take the upper feed flow as an example to illustrate the feed rearrangement. Nominally, the feed tray for the heavy reactant is tray 15 and, as the flow rate (F_{OB}) increases by a factor of 40%, the feed location should be switched to tray 16. Following the approach of Doukas and Luyben (1976), a linear combination is used to provide a gradual transition from tray 15 to tray 16 as can be seen in Fig. 2. That means 50% of the heavy reactant B comes into the column via tray 15 and the other half comes in from tray 16 for a 20% feed flow increase. The feed coordination is a bit complicated (switching among trays 10, 11, & 12), but it can be implemented in a straightforward manner as shown in Fig. 2.
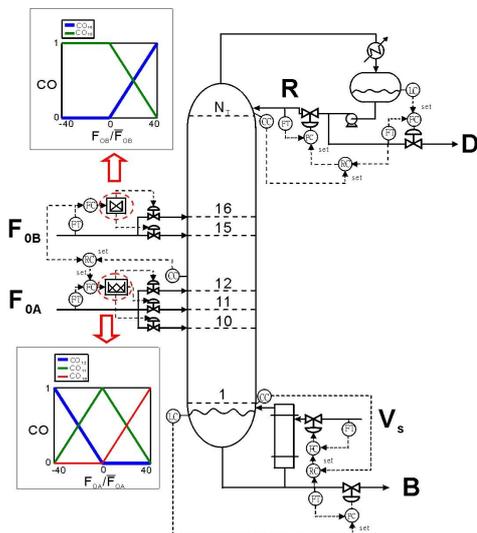

Fig. 2 Control structure of reactive distillation with coordinated feed locations as the production rate changes.

3.3 Closed loop performance

Next, the closed-loop performance of both control structures (with and without feed rearrangement) is evaluated (Figs. 1 and 2). First, consider the case of a 40% production rate increase. The control structure with coordinated control (Fig. 2) gives fast dynamics in the product composition as can be seen in Fig. 3 where top and bottoms composition return to set point in less than 10 hours (solid line in Fig. 3. On the other hand, the conventional control structure (Fig. 1) shows a little slower dynamic responses and the product compositions do not return to the set points after 10 hours. More importantly, the coordinated control structure results in a 21% energy as compared to the conventional control structure which can be seen from the smaller vapor rate in Fig. 3. Note that 21% energy saving is smaller than 28% from steady-state analysis and the reason is that we fix the tray 13 composition of A to the nominal value. Nonetheless, the amount of energy saved is still quite significant. Figure 4 shows the responses for -40% step changes in the production rate. Again, faster dynamics for top and bottoms products are observed for the coordinated control structure (Fig. 2). Moreover, 7.5% energy saving can be achieved with this improved dynamics.
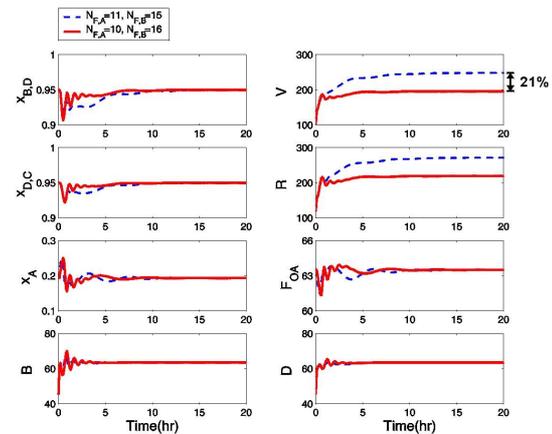

Fig. 3 Closed-loop responses for a +40% production rate increase with fixed feed locations (dashed) and coordinated feed trays (solid).

The results presented in this section clearly show that the concept of optimal feed tray location can be carried over to process operation and control. With a simple modification in the control structure (Fig. 2), improved closed loop performance can be achieved with substantial energy saving

4. CONCLUSION

In this paper, the effects of feed locations to the control of reactive distillation are explored and ideal
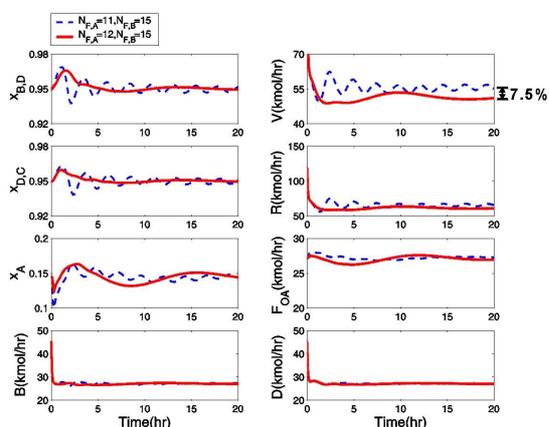
Fig. 4 Closed-loop responses for a 40% production rate decrease with fixed feed locations (dashed) and coordinated feed trays (solid).

reactive distillation systems are used to illustrate the advantage of feed trays optimisations. Because the temperature and composition profiles play a vital role for the effective utilization of the reactive section, the optimal feed locations are essential to obtain improved performance. Quantitatively, a systematic procedure is proposed to find the right feed trays. Finally, the idea of optimal feed trays can be carried over to the operation/control of reactive distillation system. First, steady-state analysis is performed to find the optimal feed trays as the measurable load variable changes. Then, a coordinated control structure is proposed to rearrange the feeds as the disturbance comes into the system. The results indicate that, again, substantial energy can be saved during process operation by feed rearrangement while showing improved closed-loop dynamics.

Table 1. Physical properties for the high activation energies case

activation energy (cal/mol)	Forward (E_F)	30000			
	Backward (E_B)	40000			
specific reaction rate at 366 K ($kmol\ s^{-1}\ kmol^{-1}$)	Forward (k_F)	0.008			
	Backward (k_B)	0.004			
heat of reaction (cal/mol)		-10000			
heat of vaporization (cal/mol)		6944			
relative volatilities($\alpha_C/\alpha_A/\alpha_B/\alpha_D$)		8/4/2/1			
		C	A	B	D
Vapor pressure constants[a]	A_{VP}	13.04	12.34	11.45	10.96
	B_{VP}	3862	3862	3862	3862

[a] $\ln P^S_i = A_{VP,i} - B_{VP,i}/T$ where T is in degrees Kelvin and P^S_i is the vapor pressure of pure component i in bar.

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