

Nonlinear model based control of two-product reactive distillation column

Myungwan Han[†] and David E. Clough*

Department of Chemical & Biological Engineering, Chungnam National University,
220, Gung-dong, Yousung, Daejeon 305-764, Korea

*Department of Chemical & Biological Engineering, University of Colorado, Boulder, CO 80309-0424, U.S.A.

(Received 10 January 2006 • accepted 1 March 2007)

Abstract—Nonlinear feedback control scheme for reactive distillation column has been proposed. The proposed control scheme is derived in the framework of Nonlinear Internal Model Control. The product compositions and liquid and vapor flow rates in sections of the reactive distillation column are estimated from selected tray temperature measurements by an observer. The control scheme is applied to an example reactive distillation column in which two products are produced in a single column and the reversible reaction $A+B=C+D$ occurs. The relative volatilities are favorable for reactive distillation so that the reactants are intermediate boilers between the light product C and the heavy product D. Ideal physical properties, kinetics, and vapor-liquid equilibrium are also assumed. It is shown that the proposed control scheme keeps tight product composition control.

Key words: Reactive Distillation, Nonlinear Internal Model Control, Dual Composition Control

INTRODUCTION

Reactive distillation has received increasing attention in recent years because of its potential for reducing capital and energy costs in some systems. When reactions are reversible or when the presence of azeotropes makes conventional separation system complex and expensive, this is particularly advantageous. This interest has led to many studies about reactive distillation regarding simulation algorithm and the design of reaction columns based on steady state.

Only a few researches have been done for the closed-loop control of reactive distillation columns. Bock et al. [1997] proposed a control scheme for a coupled reactive distillation process. Kumar and Daoutidis [1999] studied the control of a reactive distillation. In the column, ethylene oxide and water react to form ethylene glycol and consecutive reactive distillation to form diethylene glycol also occurs. They used an input-output linearizing controller that was effective at moderate purities but was unstable at higher purities. Sneesby et al. [1998] proposed a two-point control scheme for an ethyl tert-butyl ether (ETBE) reactive distillation column in which both bottoms product purity and conversion are controlled. They used conventional PI controllers, and conversion is calculated inferentially from several tray temperatures and flow measurements. Barlett and Wahnschaft [1999] studied the control of a methyl tert-butyl ether (MTBE) reactive distillation column. They explored the selection of control tray temperature. Al-Arfaj and Luyben [2000] compared several control schemes for an ideal two-product reactive distillation column. They found that the interaction between design and control is illustrated by the impact of holdup in the reactive zone. Their six control schemes do not provide a satisfactory performance for dual composition control.

Barolo and Berto [1998] proposed nonlinear internal model control strategy for composition control in batch distillation. Monroy-Loperna and Alvarez-Ramirez [2000] studied output-feedback con-

trol for the regulation of distillate purity via manipulations of reflux ratio in reactive batch distillation. Balasubrahanya and Doyle [1998] applied nonlinear model-based control to a batch reactive distillation column producing ethyl acetate. Lim and Han [2001] also applied nonlinear wave model based control scheme to the same reactive batch distillation column and showed that tight distillate composition control is possible.

The paper is organized as follows. Ideal reactive distillation column of Al-Arfaj and Luyben [2000] is described. A composition and flow estimator for the ideal reactive distillation column is developed. Nonlinear internal model control (NIMC) is introduced for the control of both product compositions of ideal reactive distillation column. Results from the NIMC-based control strategy are compared to those provided by conventional control scheme proposed by Al-Arfaj and Luyben.

PROCESS DESCRIPTION

We consider the reactive distillation column proposed by Al-Arfaj and Luyben [2000] as shown in Fig. 1. The following reversible reaction occurs in the liquid phase of the distillation column.



The volatilities of the products C and D are the lightest and heaviest, respectively, in the system.

$$\alpha_C > \alpha_A > \alpha_B > \alpha_D \quad (2)$$

The reactants A and B are intermediate boilers between the products. The specific reaction rates are given by

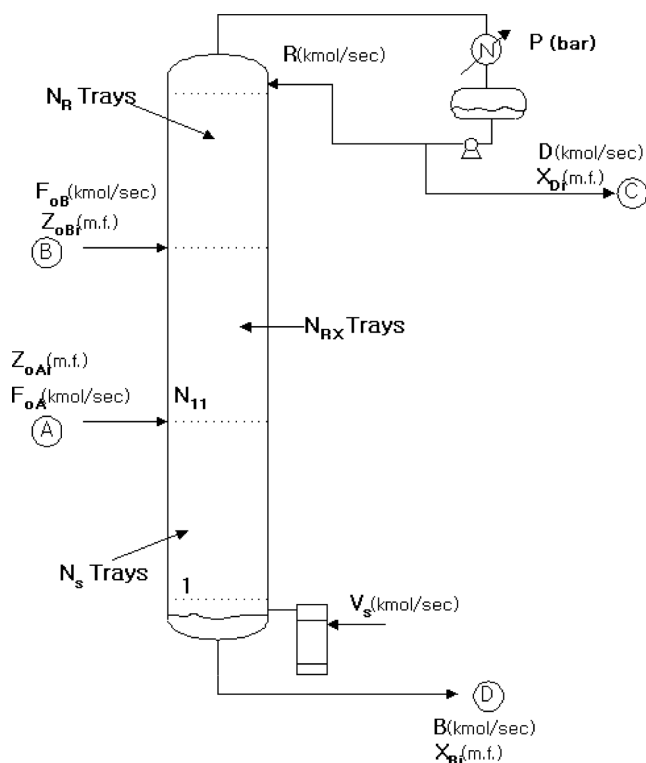
$$k_F = \alpha_F e^{-E_F/RT} \quad (3)$$

$$k_B = \alpha_B e^{-E_B/RT} \quad (4)$$

where α_F and α_B are the preexponential factors, E_F and E_B are the activation energies. Table 1 lists kinetic and physical property data for the system.

[†]To whom correspondence should be addressed.

E-mail: mwhan@cnu.ac.kr


Fig. 1. Schematic diagram of ideal reactive distillation column.
Table 1. Physical properties

Activation energy (cal/mol)	forward	30000			
	backward	40000			
Heat of reaction (cal/mol)		-10000			
Heat of vaporization (cal/mol)		6944			
Relative volatilities	α_c	8			
	α_i	4			
	α_B	2			
Vapor pressure constants			C	A	B
	Avp	13.04	12.34	11.45	10.96
	Bvp	3862	3862	3862	3862
			D		

In the model of the distillation column, vapor holdup and pressure drops are neglected. Liquid hydraulics is considered by including a linearized Francis Weir formula as the following:

$$L_m = L_{m,o} + \frac{M_m - M_o}{\tau} \quad (5)$$

where $L_{m,o}$ is the reference value of the internal liquid flow rate, M_m and M_o are the actual and reference molar holdups and τ is the tray hydraulic time constant. The vapor-liquid equilibrium is assumed to be ideal. The column pressure is fixed at 5.1 bar. Equimolar overflow is also assumed so that the steady state vapor and liquid rates are constant through the stripping and rectifying sections. However, these rates change from tray to tray in the reactive zone because the heat of reaction vaporizes some liquid on each tray.

The reactive distillation column consists of a reactive section in the middle with nonreactive rectifying and stripping sections at the top and bottom. The task of the rectifying section is to recover reac-

tant B from the product stream C. In the stripping section, the reactant A is stripped from the product stream D. In the reactive section the products are separated in situ, driving the equilibrium to the right and preventing undesired reaction between the reactants A (or B) with product C (or D).

FLOW AND COMPOSITION ESTIMATOR

It is well known that an on-line measuring device, such as a gas chromatograph, requires high investment and maintenance costs, and delivers delayed responses. Therefore, it is beneficial to use low cost, fast measurements such as temperatures for estimating tray and product compositions. We use the following equations to calculate the temperature and liquid phase composition on any tray when the vapor-liquid equilibrium of the mixture to be separated exhibits constant relative volatility as in the approach of Quintero-Marmol and Luyben [1992].

$$P_i^o = P \frac{y_i}{x_i}$$

$$\frac{y_i}{x_i} = \frac{\alpha_i}{\sum_{k=1}^{NC} \alpha_k x_{m,k} - B_i} \quad (6)$$

$$\ln P_i^o = \frac{A_i}{T} + B_i$$

where P_i^o is the vapor pressure of component i and α_i is the relative volatility. Therefore, the stage temperature can be determined.

$$T_m = \frac{A_i}{\ln \frac{P_{tot}}{\sum_{k=1}^{NC} \alpha_k x_{m,k} - B_i}} \quad (7)$$

where P_{tot} is the total pressure. In a similar manner, tray composition can be determined from the tray temperature.

$$x_{m,i} = \frac{1}{\alpha_i - \alpha_{i-1}} \left[\alpha_i P_{tot} \left\{ -A_i - \frac{B_i}{T_m} \right\} - \alpha_{i-1} \right] \quad (8)$$

$x_{i-1} = 1 - x_i$ for components $i-1, i$

A temperature is measured in the upper middle section of the column. The composition of the vapor phase of the appropriate binary mixture at that location is calculated from the measured temperature, pressure, and vapor-liquid equilibrium relationship. This calculated binary vapor is fed into a dynamic mathematical model of the column section above this temperature measurement tray.

Similarly, a tray temperature near the top is measured and the composition of the liquid phase of the appropriate binary mixture at that point is calculated from the measured temperature, pressure, and vapor-liquid equilibrium relationship. This calculated binary liquid is fed into the dynamic model of the column section below this temperature measurement tray. Tray and distillate compositions are estimated by using the dynamic model composed of the mass balance differential equations for rectifying section:

$$M_l \frac{d\hat{x}_l}{dt} = L\hat{x}_{l+1} + \hat{V}y_{l-1} - L\hat{x}_l - \hat{V}\hat{y}_l$$

$$M_m \frac{d\hat{x}_m}{dt} = L\hat{x}_{m+1} + \hat{V}\hat{y}_{m-1} - L\hat{x}_m - \hat{V}\hat{y}_m, \quad m = l+1 \dots N \quad (9)$$

$$M_D \frac{d\hat{x}_D}{dt} = \hat{V}(\hat{y}_N - \hat{x}_D)$$

where l -th and N -th trays are measurement trays and y_{l-1} and x_N are updated every sampling time. However, vapor flow rate in the rectifying section is not a known variable. We allow additional measurement inputs between the two measurements to estimate the vapor flow rate for rectifying section. Holdup of each tray is assumed to be constant. The weighted error between the estimated compositions and the measured compositions (calculated from the temperature measurements) is fed back to the model equation in order to correct the vapor flow rates in rectifying section.

This model provides estimated column composition measurements (\hat{x}_m , $m=l \dots N$) and distillate composition (\hat{x}_D) to nonlinear model based control scheme. Note that the estimated tray composition (\hat{x}_m) is treated as the measured tray composition (x_m) in the proposed control scheme. This model consists of ordinary differential equations so that an initial condition for the state variable is needed. In the same manner as above mentioned, we can estimate the liquid flow rate in the stripping section.

This estimation sometimes gives some offset from the true measurement. We enhance the estimation ability by adding additional feedback to correct the estimated flow rates in the column. The additional feedback is obtained from the mass balance equation around the reflux drum or reboiler drum. The mass equation around the reflux drum is:

$$\dot{M}_D = V_N - R - D \quad (10)$$

The mass equation around the reboiler drum is:

$$\dot{M}_B = L - V_s - B \quad (11)$$

Here, we assume that the distillate and bottom flow rates are known. From these equations, we get the estimated molar holdups for the reflux drum and the reboiler drum. The error between the measured holdup and the estimated one is used to correct the estimated liquid flow rate in stripping section or vapor flow rates in rectifying section. In summary, vapor flow rate in the stripping section is updated by the following equation:

$$\hat{V}_{new} = \hat{V}_{old} + k_1 \sum_{k=1}^m (x_k - \hat{x}_k) + k_2 (M - \hat{M}) \quad (12)$$

The vapor flow rate is adjusted to reduce the error between the measured and the estimated tray compositions as well as the error between the measured holdup and the estimated one of reflux drum.

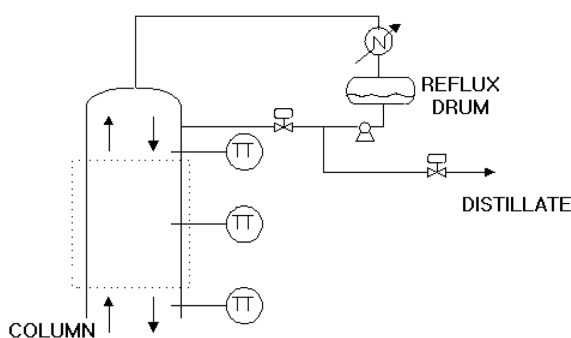


Fig. 2. Schematic diagram of the proposed estimator.

July, 2006

These two feedbacks make more accurate estimation of the flow rate possible, and thereby we can obtain more accurate tray compositions and distillate composition. For a stripping section, we can apply the same methodology to get the estimation of tray compositions and bottom composition. A schematic of the proposed estimation scheme is given in Fig. 2. The temperature measurements are updated every 1 s.

CONTROL SYSTEM DESIGN

Al-Arfaj and Luyben [2000] explored a wide variety of control structures for the reactive distillation column. The following control loops are used:

1. Pressure is controlled by condenser heat removal.
2. The base level is controlled by manipulating the bottoms flow rate.
3. The concentration of reactant A on tray 11 at the bottom of the reactive section is measured and controlled by manipulating the fresh feed rate of component A.

The control loop requires a composition analyzer so that the inventory of component A in the system can be measured, and feedback is used to maintain a certain amount of one of the reactants in the system. In this system nearly all reactants are converted into products so that the stoichiometric ratio of the reactants should be maintained. Otherwise, one of the reactants should build up or be depleted gradually. Therefore, feedback of information about reactant inventory inside the system is required for an effective control system. It is our next research topic to get the information using tray temperature measurements. We follow Al-Arfaj and Luyben to select tray 11

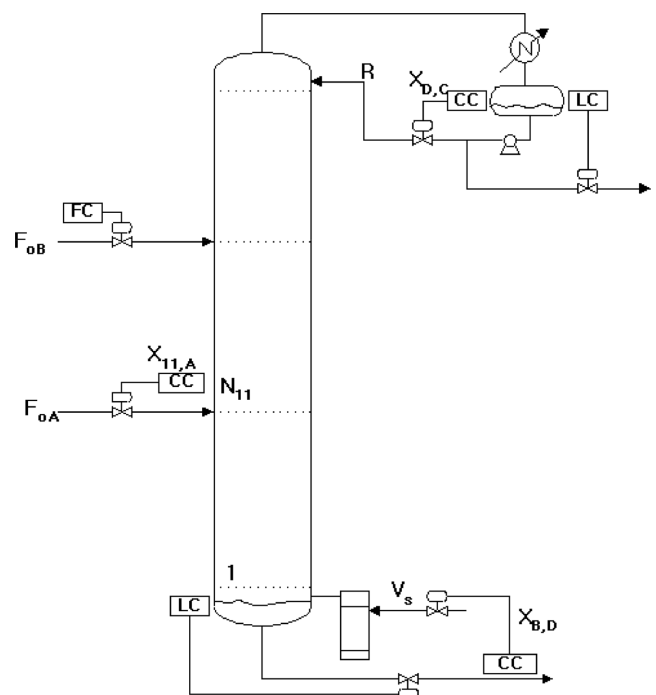


Fig. 3. Control structure CS1 (Al-Arfaj and Luyben); control purities.

as a control tray for maintaining the stoichiometric ratio of reactants.

We employ nonlinear model based control strategy for the control of product purities in the reactive distillation column. The proposed controller for distillate composition is the same as that considered by Barolo and Berto [1998]. It relies on the NIMC structure proposed by Henson and Seborg. Our control objective is to keep top composition, x_D , constant. This is achieved if the vapor mole fraction of component C in the overhead vapor is constant and equal to the distillate composition setpoint. This is because as-

sumption of total condensation of the overhead vapor makes the reflux affect the distillate composition only through a change in the overhead composition. Therefore, we can use the estimated overhead vapor composition as the controlled variable. The mass balance differential equations for distillate and top tray compositions can be written as the following:

$$\begin{aligned} \dot{x}_D &= \frac{V}{M_D}(y_N - x_D) \\ \dot{x}_N &= \frac{V}{M}(y_{N-1} - y_N) + \frac{R}{M}(x_D - x_N) \end{aligned} \quad (13)$$

Time differential of the vapor composition of top tray is

$$\dot{y}_N = \frac{\partial y_N}{\partial x_N} \dot{x}_N = \frac{V}{M}(\dot{y}_{N-1} - \dot{y}_N) \frac{\partial y_N}{\partial x_N} + \frac{R}{M}(x_D - x_N) \frac{\partial y_N}{\partial x_N} \quad (14)$$

The closed-loop transfer function (CLTF) for setpoint change is

$$\frac{Z(s)}{Z_{sp}(s)} = \frac{1}{(\varepsilon s + 1)} \quad (15)$$

where ε is the closed-loop time constant. In time domain, the equation becomes

$$\frac{1}{\varepsilon}(x_D^{sp} - \hat{y}_N) - \dot{\hat{y}}_N = 0 \quad (16)$$

Therefore, the control equation is

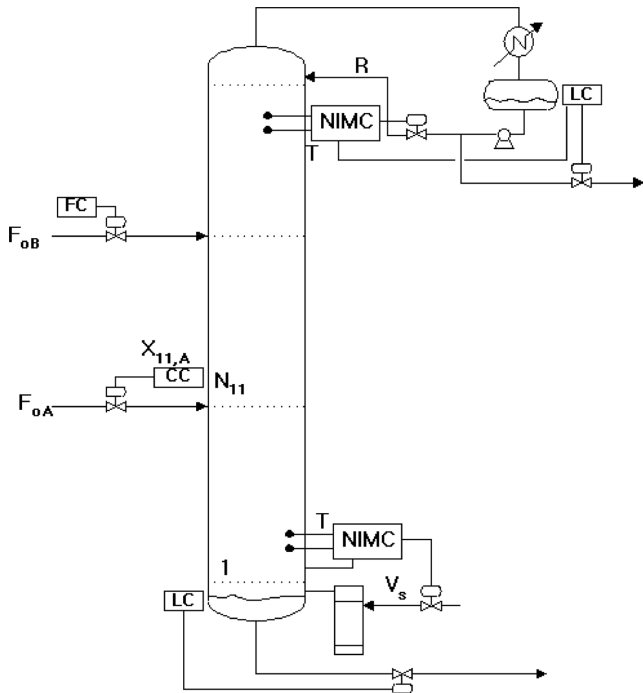


Fig. 4. The proposed control structure.

Table 2. Base-case conditions; Ideal System

Flow rate (kmol/s)	
Fresh feed F_{oA}	0.01260
Fresh feed F_{oB}	0.01260
Reflux	0.03712
Vapor boilup	0.03248
Top tray vapor	0.04971
Distillate	0.01260
Bottoms	0.01260
Pressure (bar)	
	5.1
Tray holdup (kmol)	
	1.00
Tray number	
Stripping	10
Reactive	10
Rectifying	10
Temperature (K)	
Base	411.8
Top stripping	379.8
Top reactive	369.9
Top rectifying	340.0

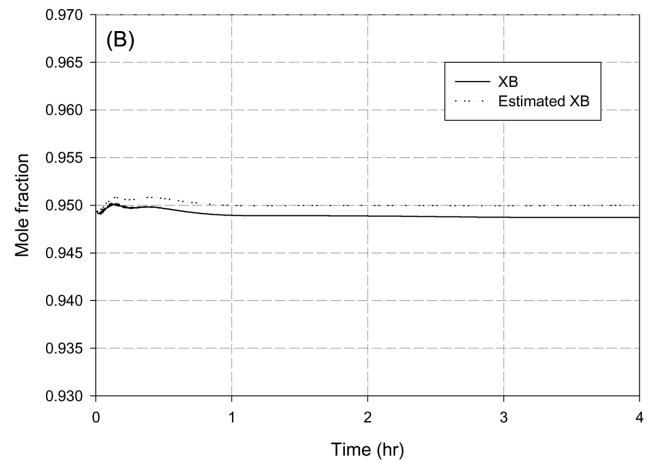
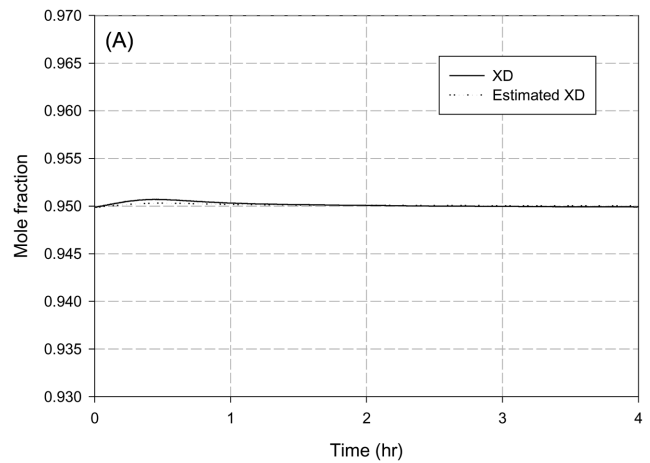


Fig. 5. The proposed control scheme; +20% F_{oB} : (A) x_D ; (B) x_B .

$$R = \frac{\frac{1}{\varepsilon}(x_D^{sp} - \hat{y}_N) - \frac{\hat{V}}{M}(\hat{y}_{N-1} - \hat{y}_N) \frac{\partial \hat{y}_N}{\partial \hat{x}_N}}{\frac{1}{M}(\hat{x}_D - \hat{x}_N) \frac{\partial \hat{y}_N}{\partial \hat{x}_N}} \quad (17)$$

Here, \hat{V} , \hat{x}_D , \hat{x}_N , \hat{y}_N , \hat{y}_{N-1} can be estimated by the proposed observer.

In the same manner as the previous approach, we can obtain the following equations for the control of bottom composition.

$$\varepsilon \hat{x}_B + x_B = x_{B,sp} \quad (18)$$

$$\hat{x}_B = (L_S x_1 - B \hat{x}_B - V_S y_B) / M_B \quad (19)$$

Combining these two equations gives:

$$\frac{M_B}{\varepsilon}(x_{B,sp} - x_B) = L_S x_1 - B x_B - V_S y_B \quad (20)$$

Therefore, one can obtain the vapor flow rate as a manipulated variable:

$$V_S = \frac{L_S x_1 - B x_B - \frac{M_B}{\varepsilon}(x_{B,sp} - x_B)}{y_B} \quad (21)$$

Fig. 5 shows dynamic response of top and bottom compositions under the proposed control scheme. The disturbance is a 20% increase in the F_{oB} fresh feed flow rate. Very good control performance is obtained and the feed disturbance does not affect the product compositions. The bottom composition shows a very slight offset

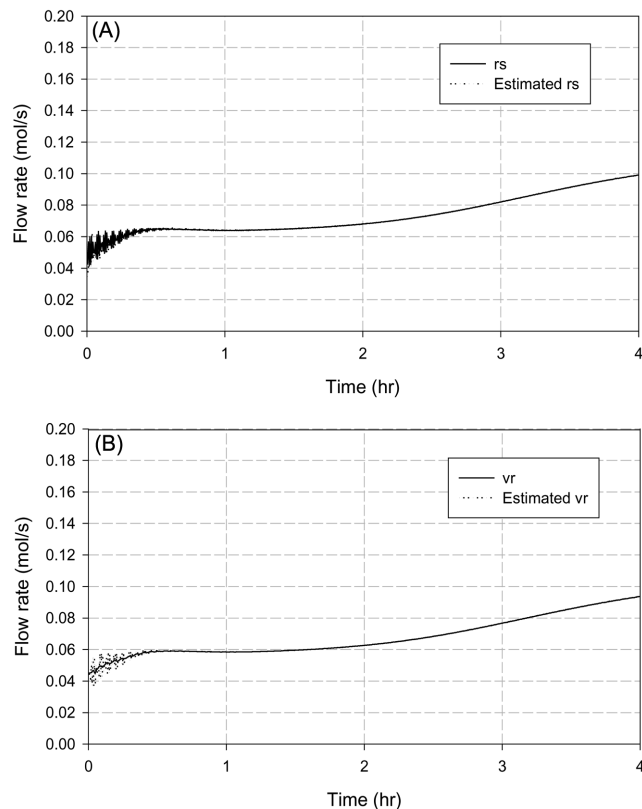


Fig. 6. Estimation of flow rates for the feed disturbance of +20% F_{oB} : (A) liquid flow rate in the stripping section; (B) vapor flow rate in the rectifying section.

from its setpoint, but the estimated bottom composition gives no offset. It indicates that the offset comes from the estimation error. Fig. 6 gives the estimated liquid flow rate in the stripping section and vapor flow rate in the rectifying section. The estimations show somewhat oscillatory responses during the first half hour but soon become stabilized. The reflux flow rate and the vapor flow rate are shown to be oscillatory at first and then to give smooth changes. The oscillation comes from the oscillatory estimation of the flow rates in both rectifying and stripping sections. The manipulated variables are passed through a rate of change filter because of the occurrence of excessive control action.

CS1 is a control scheme proposed by Al-Arfaj and Luyben in which purity of the products is measured and controlled. In the distillate product, concentration of component C is controlled by manipulating the reflux flow rate. In the bottom product, composition of component D is controlled by manipulating the vapor boilup. Proportional-only level controllers are used. Composition controllers are PI with measurement lags (30 s lag for each controller). The dynamic response of the reactive distillation column under CS1 control structure is shown in Fig. 7. This indicates that feed composition disturbance is not effectively prevented to affect the product purities. As Al-Arfaj and Luyben indicated, trying to hold a constant composition on tray 11 with PI controller drives to instability due to the increase of boilup to its limit. Figs. 8 and 9 show the responses of the closed-loop system for setpoint change in purity of bottom product. Comparing these two results illustrates that the proposed control scheme gives much better performance than CS1 control structure does.

CONCLUSIONS

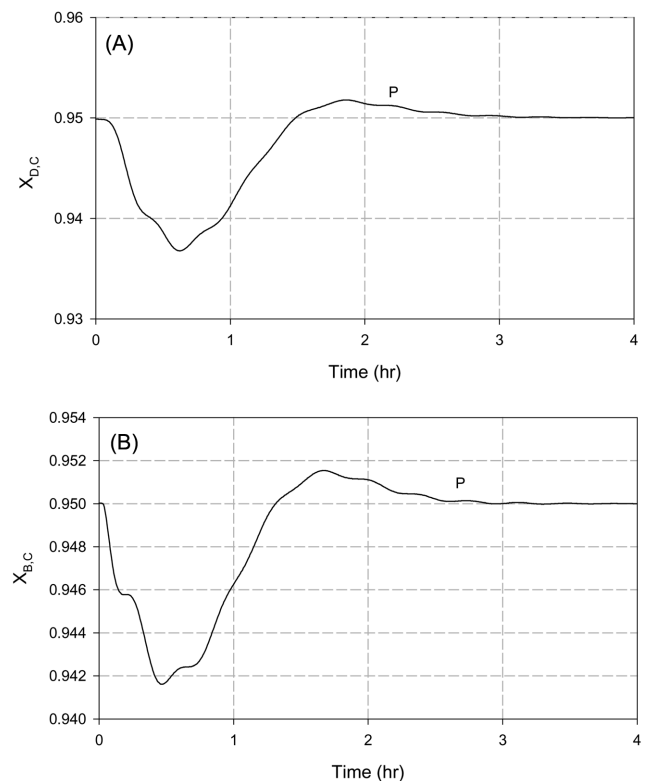


Fig. 7. CS1; +20% F_{oB} .

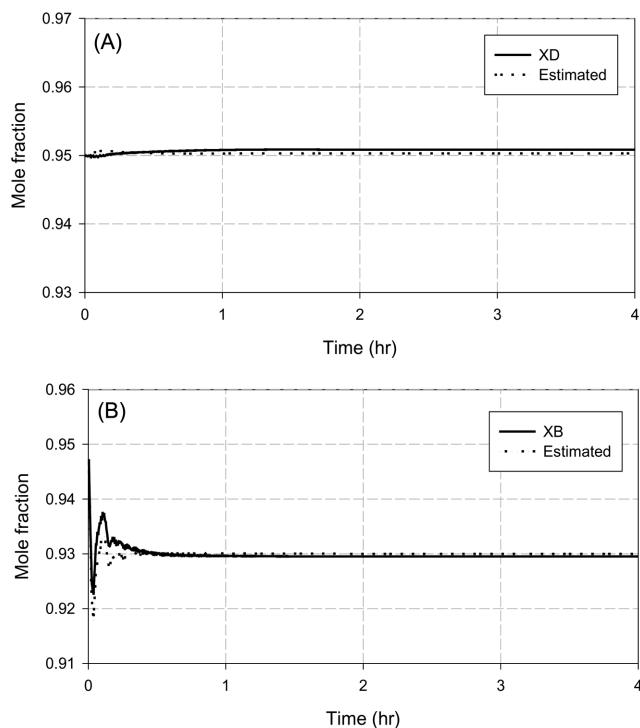


Fig. 8. The proposed control scheme; $x_{B,D}$ changed to 0.93: (A) x_D ; (B) x_B .

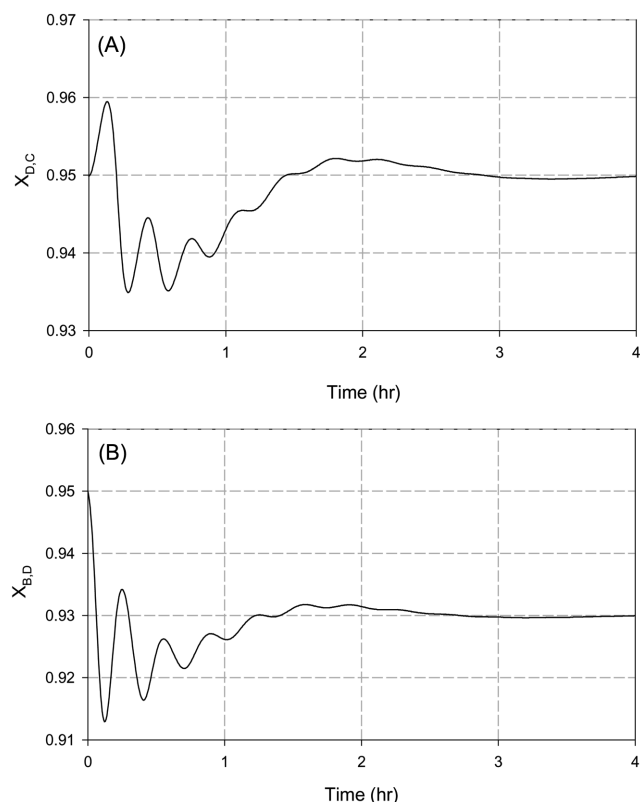


Fig. 9. CS1: $x_{B,D}$ changed to 0.93.

In this paper we propose a nonlinear model based control for an ideal two-product reactive distillation column. The strategy relies on

an NIMC structure. We propose a new state estimator for product purity and flow rate in sections of the distillation column based on several tray measurements and dynamics of reflux and reboiler drums. The estimation is necessary for practical implementation of the proposed control scheme. We examine control performance of the proposed control scheme and Al-Arfaj and Luyben's CS1 structure. The control schemes are applied to an ideal reactive distillation column and the proposed control scheme shows a superior performance compared to CS1 structure.

NOMENCLATURE

- A : reactant component
- B : reactant component; bottoms flow rate [kmol/s]
- C : product component
- D : product component; distillate flow rate [kmol/s]
- E_B : activation energy of the reverse reaction [cal/mol]
- E_F : activation energy of forward reaction [cal/mol]
- E_{O_A} : fresh feed flow rate of reactant A [kmol/s]
- F_{O_B} : fresh feed flow rate of reactant B [kmol/s]
- k_F : specific reaction rate of the forward reaction [$\text{kmol}^{-1} \cdot \text{kmol}^{-1}$]
- k_B : specific reaction rate of the reverse reaction [$\text{kmol}^{-1} \cdot \text{kmol}^{-1}$]
- L_n : liquid flow rate from tray n [kmol/s]
- M_n : liquid holdup on tray n [kmol]
- r_s : liquid flow rate in the stripping section
- V_s : vapor flow rate in the stripping section
- r_r : liquid flow rate in the rectifying section
- V_r : vapor flow rate in the rectifying section

REFERENCES

- Al-Arfaj, M. and Luyben, W. L., "Comparison of alternative control structures for an ideal two-product reactive distillation column," *Ind. Eng. Chem. Res.*, **39**, 3298 (2000).
- Balasubramhanya, L. S. and Doyle, F. J., *Nonlinear model-based control of a batch reactive distillation column*, DYCOPS-5, Corfu, Greece, June 8-10 (1998).
- Barlett, D. A. and Wahnschafft, O. M., *Dynamic simulation and control strategy evaluation for MTBE reactive distillation*, Paper presented at FOCAP (1999).
- Barolo, M. and Berto, F., "Composition control in batch distillation: binary and multicomponent mixtures," *Ind. Eng. Chem. Res.*, **37**, 4689 (1998).
- Bock, H., Wozny, G and Gutche, B., "Design and control of a reaction distillation column including the recovery system," *Chem. Eng. Progress.*, **36**, 101 (1997).
- Guttinger, T. E. and Morari, M., "Predicting multiple steady states in equilibrium reactive distillation. 1. Analysis of nonhybrid systems," *Ind. Eng. Chem. Res.*, **38**, 1633 (1999).
- Kumar, A. and Daoutidis, P., "Dynamics and control of an ethyl acetate reactive distillation column," *AIChE J.*, **45**, 51 (1999).
- Lim, C. and Han, M., "Profile position control of reactive batch distillation column," *Journal of Control, Automation and Systems Engineering*, **7**(3), 263 (2001).
- Monroy-Loperena, R. and Alvarez-Ramirez, J., "Output-feedback con-

- trol of reactive batch distillation columns," *Ind. Eng. Chem. Res.*, **39**, 378 (2000).
- Quintero-Marmol, E. and Luyben, W., "Inferential model-based control of multicomponent batch distillation," *Chem. Eng. Sci.*, **47**, 4, 887 (1992).
- Sneesby, M. G., Tade, M. O., Data, R. and Smith, T. N., "Detrimental influence of excessive fractionation on reactive distillation," *AIChE J.*, **44**, 388 (1998).