

## NONLINEAR MODEL-BASED CONTROL OF A BATCH REACTIVE DISTILLATION COLUMN

Lalitha S. Balasubramhanya \* Francis J. Doyle III<sup>\*,1</sup>

\* School of Chemical Engineering, Purdue University

\*\* Dept. of Chem. Eng., Univ. of Delaware, Newark, DE 19716

**Abstract:** The inherent trade off between model accuracy and computational tractability for model-based control applications is addressed in this article by the development of reduced order nonlinear models. Traveling wave phenomena is used to develop low order models for multicomponent reactive distillation columns. A motivational example of batch esterification column is used to demonstrate the synthesis procedure. Tight control of the column is obtained with the use of reduced model in a model predictive control algorithm. normalsize Copyright © 1998 IFAC

**Keywords:** batch reactive distillation, nonlinear MPC, traveling waves

### 1. INTRODUCTION

Reactive distillation is an attractive alternative to a series of reactors and distillation columns for separating mixtures of reactants and products. The use of reactive distillation is particularly advantageous when the conversion is limited by equilibrium or when azeotropic mixtures are formed. Examples of commercial success in implementing the principles of reactive distillation include Nylon 6,6 process, Methyl Acetate process, and Methyl *tert*-Butyl Ether process (Doherty and Buzad 1992).

However, in spite of the numerous studies conducted on understanding the fundamental thermodynamics and kinetics in the operation of the columns, very little work has been reported on the control of reactive distillation columns. For batch reactive distillation columns, Reuter *et al.* (1989) and Sørensen and Skogestad (1994) have analyzed various linear control strategies. Kumar and Daoutidis (1995) presented a differential-algebraic-equation framework to control continuous reactive distillation columns. A Dynamic Matrix Control algorithm for startup and continuous operation of a reactive distillation column has also

been proposed by Baldon *et al.* (1997). Though great strides have been made in understanding the behavior of reactive distillation columns, control of these columns is still an open research area.

Models developed from first principles are a good source of information regarding any process, however, they are invariably complex, coupled, nonlinear partial differential equations. Linear models, though simple, do not capture the nonlinear physical behavior. Data driven models based only on the input-output information, do not always capture the physically significant parameters. Understanding the physical significance of parameters and variables is often essential in developing an effective control strategy. This has fueled the need for low order nonlinear models that can capture the essential nonlinearity of the system while retaining mathematical simplicity as well as physical significance.

The approach taken in this paper is to develop low order nonlinear models to capture the essential nonlinear dynamic transport behavior of processes. Conservation of mass, energy and momentum often results in traveling waves which can be represented by wave fronts, wave pulses and wave trains (Marquardt 1990). In chemical engineering, the propagation of waves has been studied extensively in connection with reaction engineering,

<sup>1</sup> To whom correspondence should be addressed: (doyle@che.udel.edu).

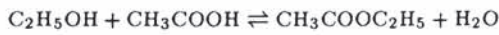


combustion technology and separation processes. Examples include temperature profiles in fixed bed reactors (Puszyński and Hlavacek 1984) and compositions profiles in adsorption columns (Rhee *et al.* 1986). Low order models can then be used in model based control strategies (Balasubramhanya and Doyle III 1997, Doyle III *et al.* 1996, Marquardt and Gilles 1990) to achieve tight control of the processes.

In this paper, an esterification process is used to motivate the development of low order models for reactive distillation columns.

## 2. BATCH REACTIVE DISTILLATION COLUMN CASE STUDY

The case study under consideration is an esterification process where *Ethanol* reacts with *Acetic Acid* to produce *Ethylacetate* and *Water* according to:



The reaction kinetics are given by  $R = k_1 \tilde{x}_2 \tilde{x}_0 - k_2 \tilde{x}_1 \tilde{x}_3$  where  $\tilde{x}$  are the liquid mole fractions and  $k_1$  and  $k_2$  are the reaction rate constants. The components are numbered as follows: Acetic Acid (0) < Water (1) < Ethanol (2) < Ethylacetate (3). Constant relative volatility is assumed to represent the VLE. Using *Acetic Acid* as the reference, the relative volatilities of the components can be calculated:

$$\alpha_0 = 1 < \alpha_1 = 1.72 < \alpha_2 = 3.9281 < \alpha_3 = 4.0286 \quad (1)$$

The product of interest is *Ethylacetate* and the objective is to push the reaction in the forward direction and increase the yield of *Ethylacetate*. The operation under consideration is a batch reactive distillation column consisting of eight trays. For simulation purposes, the process is modeled using a tray-by-tray description resulting in thirty-one differential equations. The trays and the condenser are assumed to have constant molar overflow. The partial vapor pressure and the vapor composition are calculated using Antoine's constants. The values of the parameters used in simulating the detailed column are presented in Table 1. For a known pressure drop in the column, the compositions and the temperature are iteratively calculated to satisfy the mole balances. This essentially assumes that the thermal equilibrium is attained and the equilibrium temperature depends only on the pressure and composition. Further detail can be introduced in the model (hydrodynamics, energy balances, etc.) resulting in a complex model of higher order.

For the purposes of analysis, a spatial distribution of the components is considered. Based on the constant relative volatility description of the vapor-liquid-equilibrium (VLE), the vapor compositions can be represented as ( $j = 0, 1, 2, 3$ ):

$$\tilde{y}_j = \frac{\alpha_j \tilde{x}_j}{(\alpha_1 - 1)\tilde{x}_1 + (\alpha_2 - 1)\tilde{x}_2 + (\alpha_3 - 1)\tilde{x}_3 + 1}$$

The reboiler generates a vapor phase which acts as the feed to the distillation column. For the following theoretical development, we consider a continuous column versus a set of discrete trays. The distribution of the various species in the distillation column can then be represented by the following partial differential equations (mass balances):

$$\begin{aligned} \frac{\partial \tilde{x}_1}{\partial t} - \frac{\partial \tilde{x}_1}{\partial z} + \nu \frac{\partial \tilde{y}_1}{\partial t} + \mu \frac{\partial \tilde{y}_1}{\partial t} &= R \\ \frac{\partial \tilde{x}_2}{\partial t} - \frac{\partial \tilde{x}_2}{\partial z} + \nu \frac{\partial \tilde{y}_2}{\partial t} + \mu \frac{\partial \tilde{y}_2}{\partial t} &= -R \\ \frac{\partial \tilde{x}_3}{\partial t} - \frac{\partial \tilde{x}_3}{\partial z} + \nu \frac{\partial \tilde{y}_3}{\partial t} + \mu \frac{\partial \tilde{y}_3}{\partial t} &= R \end{aligned} \quad (2)$$

where

$$\begin{aligned} R &= k_1 \tilde{x}_2 (1 - \tilde{x}_1 - \tilde{x}_2 - \tilde{x}_3) - k_2 \tilde{x}_1 \tilde{x}_3 \\ \tilde{x}_0 &= 1 - \tilde{x}_1 - \tilde{x}_2 - \tilde{x}_3 \end{aligned} \quad (3)$$

$R$  describes the rate of generation of the components,  $\nu (= 0.01)$  is the ratio of the vapor to liquid holdup, and  $\mu = \frac{V}{L}$  is the ratio of the vapor to the liquid flow rates. Since the mole fractions sum up to 1,  $\tilde{x}_0 = 1 - \sum_{j=1}^3 \tilde{x}_j$ . Equation 2 is a continuous representation of the discrete set of equations for the column without the reboiler. These equations can be analyzed to demonstrate the formation of shock waves (Balasubramhanya 1997).

## 3. LOW ORDER MODEL DEVELOPMENT

In the derivation of the reduced model the following simplifications are introduced: (1) components with similar relative volatilities are lumped together; (2) reaction equilibrium is assumed; (3) a constant structure for the composition profile of  $\tilde{x}_3$  is assumed; and (4) a constant structure for the temperature profile is assumed.

The relative volatilities of *Ethanol* ( $\alpha_2 = 3.9281$ ) and *Ethylacetate* ( $\alpha_3 = 4.0286$ ) are very close to each other indicating similar vapor-liquid-equilibrium. Thus, the mole fractions of the two components can be lumped together. Similarly, the relative volatilities of *Water* ( $\alpha_1 = 1.72$ ) and *Acetic Acid* ( $\alpha_0 = 1$ ) are relatively close and their mole fractions can be lumped together as well. We define the new variables:

$$\tilde{x}_{l_1} = \tilde{x}_3 + \tilde{x}_2 \quad (4)$$

$$\tilde{x}_{l_2} = \tilde{x}_1 + \tilde{x}_0 = 1 - \tilde{x}_{l_1} \quad (5)$$

Let the relative volatility of  $\tilde{x}_{l_1}$  with respect to  $\tilde{x}_{l_2}$  be given by  $\alpha_l$ , then the vapor composition is given by



$$\tilde{y}_{l_1} = \frac{\alpha_l \tilde{x}_{l_1}}{(\alpha_l - 1)\tilde{x}_{l_1} + 1} \quad (6)$$

$$\tilde{y}_{l_2} = \frac{\tilde{x}_{l_2}}{(\alpha_l - 1)\tilde{x}_{l_1} + 1} = 1 - \tilde{y}_{l_1} \quad (7)$$

$\alpha_l$  is the average relative volatility and for this case it is computed to be 2.9253. Using the lumping simplification, Equation 2 is reduced to

$$\frac{\partial \tilde{x}_{l_1}}{\partial t} - \frac{\partial \tilde{x}_{l_1}}{\partial z} + \nu \frac{\partial \tilde{y}_{l_1}}{\partial t} + \frac{V}{L} \frac{\partial \tilde{y}_{l_1}}{\partial t} = 0 \quad (8)$$

The equilibrium relation is given by Equation 6,  $\tilde{x}_{l_2} = 1 - \tilde{x}_{l_1}$ , and  $\tilde{y}_{l_2} = 1 - \tilde{y}_{l_1}$ . Lumping of the components leads to a pseudo-binary distillation column which can be analyzed for the formation of shock waves. The VLE relation is used to show that

$$\frac{d^2 \tilde{y}_{l_1}}{d\tilde{x}_{l_1}^2} = -\frac{\alpha_l(\alpha_l - 1)}{((\alpha_l - 1)\tilde{x}_{l_1} + 1)^3} < 0 \quad (9)$$

Since  $\frac{d^2 \tilde{y}_{l_1}}{d\tilde{x}_{l_1}^2} < 0$ , the characteristics for the Equation 8 overlap which results in the formation of shock waves. The system travels with a shock velocity given by

$$\lambda_{sh} = \frac{-1 + \mu \left[ \frac{\tilde{y}_{l_1}}{\tilde{x}_{l_1}} \right]}{1 + \nu \left[ \frac{\tilde{y}_{l_1}}{\tilde{x}_{l_1}} \right]} \quad (10)$$

where  $[\cdot]$  represents a jump in the quantity across the shock.

Using a second order approximation for the VLE, Marquardt and Gilles (1990) analytically derived a function to describe the shape of the composition profile for an ordinary binary distillation column. A similar expression is used to approximate the concentration profile of the lumped system:

$$\tilde{x}_{l_1}(z) = \tilde{x}_{l_{min}} + \frac{\tilde{x}_{l_{max}} - \tilde{x}_{l_{min}}}{1 + \exp(-\gamma_l(\xi - \xi_s))} \quad (11)$$

where  $\tilde{x}_{l_{min}}$  and  $\tilde{x}_{l_{max}}$  are the constants determining the boundary of the concentration profiles and  $\gamma_l$  reflects the steepness of the profile.  $\xi = z - \lambda_l t$ , and  $\xi_s$  represents the stagnation or the inflection point of the profile. The time derivative of this stagnation point is given by the shock velocity in Equation 10. Thus, from Equation 10 and Equation 11 the distribution of  $\tilde{x}_{l_1}$  along the column, and its dynamic behavior, can be obtained.

The second assumption employed in this development was that reaction equilibrium is attained. This results in the following relationship between the components  $K_r = \frac{k_1}{k_2} = \frac{\tilde{x}_3 \tilde{x}_1}{\tilde{x}_2 \tilde{x}_0}$ . Using the reaction equilibrium assumption the composition of component 1 can be represented as

$$\begin{aligned} \tilde{x}_1 &= \frac{k_1 \tilde{x}_2 - k_1 \tilde{x}_2^2 - k_1 \tilde{x}_3}{k_1 \tilde{x}_2 + k_2 \tilde{x}_3} \\ &= \frac{k_1(\tilde{x}_{l_1} - \tilde{x}_3) - k_1 \tilde{x}_{l_1}^2 + k_1 \tilde{x}_3 \tilde{x}_{l_1}}{k_1 \tilde{x}_{l_1} + (k_2 - k_1) \tilde{x}_3} \end{aligned} \quad (12)$$

Also  $\tilde{x}_2 = \tilde{x}_{l_1} - \tilde{x}_3$  and  $\tilde{x}_0 = 1 - \tilde{x}_1 - \tilde{x}_{l_1}$ . Thus, the mole fractions of all the components can be represented as functions of  $\tilde{x}_3$  and  $\tilde{x}_{l_1}$ . Since the distribution of  $\tilde{x}_{l_1}$  is given by Equation 11, the only component of interest is  $\tilde{x}_3$ .

The distribution of  $\tilde{x}_3$  can be obtained from Equation 2

$$\frac{\partial \tilde{x}_3}{\partial t} + \nu \frac{\partial \tilde{y}_3}{\partial t} - \frac{\partial \tilde{x}_3}{\partial z} + \mu \frac{\partial \tilde{y}_3}{\partial z} = 0 \quad (13)$$

The right hand side of Equation 2 is identically zero as reaction equilibrium is assumed. Assuming VLE is attained, we get

$$\begin{aligned} (1 + \nu \left[ \frac{\partial \tilde{y}_3}{\partial \tilde{x}_3} \right]_{\tilde{x}_{l_1}}) \frac{\partial \tilde{x}_3}{\partial t} + (-1 + \mu \left[ \frac{\partial \tilde{y}_3}{\partial \tilde{x}_3} \right]_{\tilde{x}_{l_1}}) \frac{\partial \tilde{x}_3}{\partial z} = \\ (\nu \lambda_l - \mu) \left[ \frac{\partial \tilde{y}_3}{\partial \tilde{x}_3} \right]_{\tilde{x}_3} \frac{d\tilde{x}_{l_1}}{d\xi} \end{aligned} \quad (14)$$

The characteristics for the above partial differential equation indicate the formation of shock waves. Details of the analysis can be obtained from (Balasubramhanya 1997). Thus, the reduced model is able to capture the formation of shocks.

Since the partial differential equation for  $\tilde{x}_3$  admits shock, the composition profile travels with a shock velocity. Also, as the concentrations of the species are interlinked via the VLE, all the composition profiles travel with the same wave velocity. This shock velocity can be represented as:

$$\lambda_{sh} = \frac{-1 + \mu \left[ \frac{\tilde{y}_j}{\tilde{x}_j} \right]}{1 + \nu \left[ \frac{\tilde{y}_j}{\tilde{x}_j} \right]} \quad j = 1, 2, 3 \quad (15)$$

It is assumed that the structure of the composition profile of  $\tilde{x}_3$  remains constant and can be represented as:

$$\tilde{x}_3 = \tilde{x}_{3_{min}} + \frac{\tilde{x}_{3_{max}} - \tilde{x}_{3_{min}}}{1 + \exp(-\gamma_3(\xi - \xi_s))} \quad (16)$$

This structure is similar to the structure derived theoretically by Marquardt and Gilles (1990) (Equation 11). It is assumed that the structure obtained from a second order approximation to VLE can be used to model the profile of component  $\tilde{x}_3$ .  $\tilde{x}_{3_{min}}$  and  $\tilde{x}_{3_{max}}$  fix the end points of the profile while  $\gamma_3$  gives the steepness of the profile.

The energy balances are intrinsically linked to the mass balances in a column and hence traveling composition profiles imply traveling temperature profiles. As was explained earlier, for a given



pressure drop, fixing the temperature fixes the composition and vice versa. The relationship is based on the vapor liquid equilibrium (relative volatility or the Antoine's equations). Since the temperature is closely related to the composition profiles, a constant structure for the temperature profile is assumed:

$$T_m = T_{m_{min}} + \frac{T_{m_{max}} - T_{m_{min}}}{1 + \exp(-\gamma_{T_m}(\xi - \xi_s))} \quad (17)$$

where, as before,  $T_{m_{max}}$  and  $T_{m_{min}}$  fix the end points of the profile while  $\gamma_{T_m}$  gives the "steepness" of the profile. This is equivalent to assuming that energy balances reach steady state faster than the mass balances. The temperature profile also travels with the shock velocity given by Equation 15.

To summarize, the reduced model consists of the following components:

- mass balance equations (four differential equations) to analyze the dynamics of the reboiler
- distribution of  $\tilde{x}_{l_1}$  is given by Equation 11
- distribution of  $\tilde{x}_3$  is given by Equation 16
- $\tilde{x}_2 = \tilde{x}_{l_1} - \tilde{x}_3$ ,  $\tilde{x}_1 = \frac{k_1 \tilde{x}_2 - k_1 \tilde{x}_2^2 - k_1 \tilde{x}_3}{k_1 \tilde{x}_2 + k_2 \tilde{x}_3}$ , and  $\tilde{x}_0 = 1 - \tilde{x}_1 - \tilde{x}_{l_1}$
- the temperature profile for the system is given by Equation 17
- the velocity with which the waves travel is given by  $\lambda_{sh} = \frac{-1 + \mu \frac{[\tilde{y}_2]}{[\tilde{x}_3]}}{1 + \nu \frac{[\tilde{y}_3]}{[\tilde{x}_3]}}$

Thus, the total number of 5 differential equations and 6 algebraic equations are needed to represent the reduced model, compared with 31 differential equations and 10 algebraic equations in the original (full) model.

#### 4. OPEN-LOOP MODEL VALIDATION

In this section, the dynamic behavior of the detailed column is compared to the nonlinear reduced model and a linear model. In industry, it is difficult to obtain the composition measurements at frequent time intervals to perform feedback control. Hence, temperature measurements are used as the secondary output for control purposes. In a typical distillation column the regions of intense mass transfer are located in the middle of the column while the ends of column are used for purification. These regions are more sensitive to disturbances and inputs compared to the ends of columns. Similar behavior is observed in the reactive distillation column under discussion. Hence, the temperature on Tray 2, which is one of the most sensitive to the changes in the column, is used as the secondary output in this study. The inputs to the system are the liquid flow rate,  $L$ , and the vapor flow rate,  $V$ .

The detailed column can be represented as a 31st order nonlinear state-space model. The column can be linearized around the initial operating point to obtain a linear model. The third model under consideration is the nonlinear wave model discussed in the previous section, which can be represented as a 5th order nonlinear state-space model. In this case, the differential equations consist of the mass balances for the reboiler and the equation defining the shock dynamics of the traveling wave (Equation 15). The parameters defining the reduced model are presented in Table 2.

The traveling waves in the detailed column are compared to the traveling waves in the reduced model in Figure 1. The reduced model is qualitatively able to capture the traveling wave phenomenon exhibited by the detailed column. The normalized distance of Tray 2 from the reboiler is equal to 0.22.

#### 5. CONTROLLER DESIGN

The control methodology for the batch column is based on model predictive control (MPC). Most MPC algorithms are based on linear models such as step or impulse response models, however, in this work a nonlinear model is employed leading to a nonlinear optimization program which must be solved at each time step. The reduced order model developed earlier based on the traveling wave phenomenon is used as the simplified model to represent the column. The formulation of the control problem is a nonlinear modification of the formulation suggested by Ricker (1990). In our formulation, the reduced wave model is used to predict outputs into the future and a nonlinear optimization routine (obtained from MATLAB) is used to calculate the input moves as well. Hence, the approach used is the Nonlinear Quadratic Dynamic Matrix Control with State Estimation (NLQDMC/SE)

The primary objective is to obtain as pure a sample of the product over the entire batch as possible. As discussed in the previous section, the temperature on tray 2 will be used as the controlled variable. The objective function for the minimization problem to be solved at each instance of time is given by:

$$\begin{aligned} \min_{\mathbf{u}} \quad & \sum_{i=k}^{k+p} (r_{dis}(i) - y_{dis}(i))^2 \\ 0.0 \leq \mathbf{u}(i) \leq \mathbf{u}_{max} \quad & i = k, \dots, k+m \\ T_{tray2}(i) \leq T_{max} \quad & i = k, \dots, k+p \\ \Delta \mathbf{u}(i) \leq \Delta \mathbf{u}_{max} \quad & i = k, \dots, k+m \\ y_{dis}(i) = y_{dis}(i-1) + \\ (V(i-1) - L(i-1))T_s \quad & i = k, \dots, k+p \end{aligned} \quad (18)$$

$r_{dis}(i)$  is the reference trajectory for the amount of distillate to be produced at time instant  $k+i$ ,



$y_{dis}(i)$  is the predicted amount of distillate produced,  $u_{max}$  is the maximum input,  $\Delta u_{max}$  is the maximum input change at every time period, and  $T_{max}$  is the maximum temperature allowed on tray 2.  $u$  is a vector of magnitude  $2m \times 1$  corresponding to the 2 inputs and the  $m$  moves that the controller calculates at every time step. The temperature constraint at  $T_{max}$  places a lower bound on the purity of the distillate.  $T_{tray2}(i)$  is the output of the reduced model used to predict the temperature  $i$  steps into the future. A linear filter is used to update the states and the outputs of the reduced model based on the measurements of temperature obtained from the real column. A schematic of the controller design is presented in Figure 2. The solid lines trace the control loop associated with the amount of distillate collected while the dashed lines trace the temperature feedback to the controller. The dotted line represents the MPC controller that minimizes the difference between the reference trajectory and the output prediction within the constraints imposed.

For a reference trajectory consisting of a series of steps, the output of the condenser and the input moves are shown in Figure 3. The model used to predict the temperature is the nonlinear wave model developed earlier. As shown, the output tracks the reference trajectory very closely. The closed-loop simulation time using the detailed column was 225.39 cpu seconds while the time required using the reduced model was 34.34 cpu seconds. Thus, the computational time can be reduced by a factor of about 6.5 by using the reduced model. All the simulations were carried out on a Sun3000 workstation. For both the simulations  $p = 2$ ,  $m = 1$ ,  $T_{max} = 370K$ ,  $u_{max} = [3.8 \ 4.00]^T$  kmol/hr, and  $|\Delta u_{max}| = [0.6 \ 0.6]^T$  kmol/hr.

The responses of the wave model (dashed line), detailed column (solid line), and the linear model (dashed dot) to the input moves from Figure 3 are analyzed and shown in Figure 4. As can be seen, the wave model does a much better job than the linear model in capturing the dynamics of the detailed model. Next, the mole fraction of the product of interest, *Ethyl Acetate*, in the vapor stream exiting the column and the distillate collected is plotted in Figure 5. Thus, by maintaining the temperature below a maximum temperature, the mole fraction of the product can be maintained above a certain value.

### 6. CONCLUSIONS

In this study, a reduced model for reactive distillation columns was developed based on the traveling wave phenomena. A motivating example of esterification was used to demonstrate the approach.

The reduced model was obtained by introducing the following simplifications in the partial differential representation of the detailed model: 1) lumping of the components with similar VLE; 2) reaction equilibrium; 3) constant structure of the composition profiles; 4) constant structure for the temperature profile. The reduced model consists of 5 differential equations- four to represent the dynamics of the reboiler and one to represent the dynamics of the column and 6 algebraic equations. The method outlined for obtaining the reduced model can be extended for systems with more than four components by similar lumping approaches.

Nonlinear optimization problems have presented a considerable challenge for reliable application of nonlinear MPC. One method to address feasibility and robustness of such approaches is to minimize the computational complexity by using a reduced order fundamental model of the type described in this paper.

### Acknowledgments

The authors would like to thank Dr. Ramkrishna and R. M. Wajge at Purdue University for helpful discussions.

Table 1. Detailed Column Parameters

Holdup (kmols)			
$M_{0,init}$			4.7980
$M_j \ (j=1, \dots, 8)$			0.0125
$M_9$			0.1
Reaction Constants (litres/gmol min)			
$k_1$			$2900 \exp(-7150/T(K))$
$k_2$			$7380 \exp(-7150/T(K))$
Vapor Pressure (Torr); $\log_{10}(P) = A_p - \frac{B_p}{T(^\circ C) - C_p}$			
Components	Antoine's Constants		
	$A_p$	$B_p$	$C_p$
Acetic Acid	8.02100	1936.010	258.451
Ethanol	8.11220	1592.864	226.184
Ethylacetate	7.10179	1244.951	217.881
Water	8.07131	1730.630	233.426
Density (g/cc) $\rho_l = A_d B_d^{-(1-T_r)^{2/7}}$ ; $T_r = T(K)/T_c(K)$			
Components	$A_d$	$B_d$	$T_c \ (K)$
Acetic Acid	0.3512	0.2	594.4
Ethanol	0.2903	0.2760	516.2
Ethylacetate	0.3084	0.252	523.2
Water	0.3471	0.274	647.3

### 7. REFERENCES

Balasubramhanya, L. S. (1997). Low Order Models for Nonlinear Process Control. PhD thesis. Purdue University.

Balasubramhanya, L. S. and F. J. Doyle III (1997). Nonlinear control of a high-purity distillation column using a traveling wave model. *AIChE J.* **43**(3), 703-714.

Baldon, J. L., J. J. Strifezza, M. S. Basualdo and C. A. Ruiz (1997). Control policy for the startup, semi-continuous and continuous operation of a reactive distillation column. In: *IFAC Symposium on Advanced Control of Chemical Processes*. pp. 125-130.

Doherty, M. F. and G. Buzad (1992). Reactive Distillation by Design. *Chem. Eng. Res. Des.* **70**(A), 448-458.



Doyle III, F. J., H.M. Budman and M. Morari (1996). "Linearizing" Controller Design for a Packed-Bed Reactor Using a Low-Order Wave Propagation Model. *Ind. Eng. Chem. Res.* **35**, 3567–3580.

Kumar, A. and P. Daoutidis (1997). Nonlinear Control of a High-Purity Ethylene Glycol Reactive Distillation Column. In: *IFAC Symposium on Advanced Control of Chemical Processes*. pp. 371–376.

Marquardt, W. (1990). Traveling waves in chemical processes. *Int. Chem. Eng.* **30**(4), 585–606.

Marquardt, W. and E. D. Gilles (1990). Nonlinear Wave Phenomena as Fundamentals for Model Based Control System Design in Distillation. *AIChE Annual Meeting*, Chicago, IL.

Puszyński, J. and V. Hlavacek (1984). Experimental Study of Ignition and Extinction Waves and Oscillatory Behavior of a Tubular Nonadiabatic Fixed Bed Reactor for the Oxidation of Carbon Monoxide. *Chem. Eng. Sci.* **39**(4), 681–692.

Reuter, E., G. Wozny and L. Jeromin (1989). Modeling of Multicomponent Batch Distillation Processes With Chemical Reaction and Their Control Systems. *Comput. Chem. Engng.* **13**(4-5), 449–510.

Rhee, H., R. Aris and N. R. Amundson (1986). *First-Order Partial Differential Equations: Theory and Application of Hyperbolic Systems of Quasilinear Equations*. Vol. 2. Prentice-Hall, Inc.

Ricker, N. L. (1990). Model Predictive Control with State Estimation. *Ind. Eng. Chem. Res.* **29**(3), 374–382.

Sørensen, E. and S. Skogestad (1994). Control strategies for reactive batch distillation. *J. Proc. Cont.* **4**(4), 205–217.

Table 2. Wave Model Parameters

Parameters	Numerical Values
$\nu$	0.01
$\alpha_3$	4.0286
$\alpha_2$	3.9281
$\alpha_1$	1.72
$\alpha_0$	1.0
$\tilde{x}_{lmin}$	0.15
$\tilde{x}_{lmax}$	1.0
$\gamma_l$	8.8
$\tilde{x}_{3min}$	0.025
$\tilde{x}_{3max}$	0.65
$\gamma_3$	10.0
$T_{min}$	372.0
$T_{max}$	350.5
$\gamma T_m$	10.5

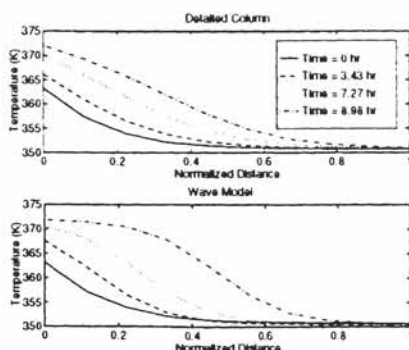


Fig. 1. Open-loop response of traveling temperature profiles in the detailed model and the nonlinear wave model(  $u = [2.15 \ 2.3]^T$  kmol/hr)

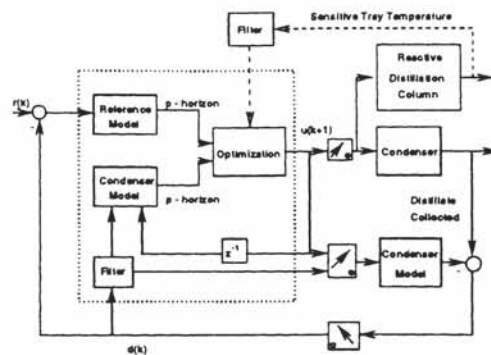


Fig. 2. Schematic diagram of MPC flowsheet with temperature feedback.

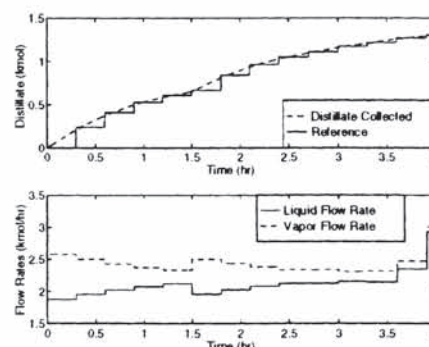


Fig. 3. Closed-loop response to filtered sequence of step changes in distillate reference (controller model = wave model)

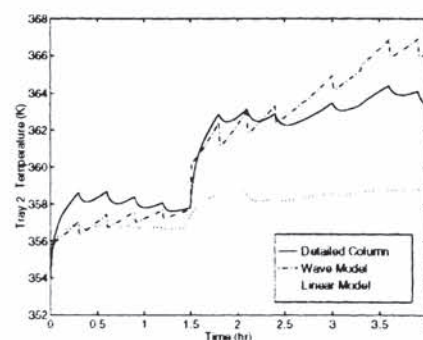


Fig. 4. A comparison of the open-loop responses of the detailed column, wave model, and the linear model to the manipulated input moves presented in Figure 3.

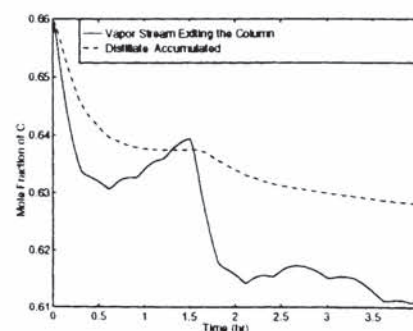


Fig. 5. The instantaneous mole fraction of Ethyl Acetate in the vapor stream and the distillate collected in response to the input moves in Figure 3.