

# DYNAMIC COMPARISON OF LINEAR AND NONLINEAR MODELS FOR GENERIC REACTIVE DISTILLATION SYSTEM

M. J. OLANREWaju and M. A. AL-ARFAJ\*

*Department of Chemical Engineering, King Fahd University of Petroleum & Minerals, Dhahran, Saudi Arabia*

The performance of a linearized dynamic model of a generic reactive distillation system has been compared with that of a rigorous nonlinear model. The effect of steady state sensitivity to different input step changes on the performance of the approximate model has been explored. The accuracy of an approximate linear model is quantified by the use of an average relative error. The approximate linear model nicely averages the process nonlinearities when the magnitude of input change is small and becomes inadequate as the deviation from base steady state increases with increase in the magnitude of input change. The performance and robustness of linear model is improved with the inclusion of internal composition inventory controller in the openloop dynamic model of the system. Simulation studies show that a poor design of an internal composition controller gain could decrease the accuracy of reactive distillation linear process model. Therefore, a well tuned controller gain is a necessity for a good performance of the linear process model.

*Keywords: reactive distillation; linear model; nonlinear model; dynamic simulation.*

## INTRODUCTION

Among several approaches to process intensification is hybrid combination of separation and reaction for chemical synthesis. This combination can lead to high-efficiency process systems with significant economic attributes (e.g., reduce energy requirement, increase conversion, lower solvent use, reduce equipment investments and offer greater selectivity). Reactive distillation technology has been of a special interest in recent times and has shown a significant growth in both patents and journal papers (Al-Arfaj and Luyben, 2000, 2002, 2004; Sundmacher and Kienle, 2003; Malone & Doherty, 2000; Taylor and Krishna, 2000).

An understanding of the dynamic behaviour of a reactive distillation system is important from both process design and control perspectives. Moreover, the primary objective of process control is the design of effective and robust control systems that will keep the process conditions close to its desired steady state value. Even though the reactive distillation system is highly nonlinear, the influence of effective regulatory control is to ensure that the deviations from this steady state will be small, in which case the behaviour will be essentially indistinguishable from that of linear system.

Nonlinear reactive distillation systems are notoriously difficult to analyse and solve, partially because they exist

is such an infinite variety of forms, preventing any cohesive theory for analysis. Thus, it is very important to have approximate linear model that will give good account of the process behaviour near the desired operating conditions if we are to be able to use the powerful linear mathematical techniques in the system analysis and control.

Nonlinearity in reactive distillation model arises because of complex processing configurations, which involves the interaction of the reaction kinetics and distillation concept of vapour–liquid equilibrium. Moreover, the desire for high conversion, selectivity and product purity increases the process nonlinearity. Fuentes and Luyben (1983) pointed out that the response of distillation system becomes highly nonlinear as the purity level increases more than 98%.

Although simplified modelling of distillation columns for design of linear multivariable controllers has a long tradition (Luyben, 1987; Marquardt and Amrhein, 1994; Skogestad and Morari, 1987), there is still no consensus on what constitute an adequate linear model of reactive distillation, on the physical effects to be retained and on a recommended approximation method that will not lead to a false conclusion. These questions can only be addressed by a quantitative comparison of an approximate linear model to that of nonlinear rigorous model.

This paper compares the performance of a linearized dynamic model of a generic reactive distillation system with that of nonlinear model with the sole aim to come up with some conditions and general guidelines under which linear process model could be applied in

\*Correspondence to: Dr M. A. Al-Arfaj, Department of Chemical Engineering, King Fahd University of Petroleum & Minerals, Dhahran, 31261, Saudi Arabia.  
E-mail: maarfaj@kfupm.edu.sa

modelled-based-control applications of reactive distillation. The effect of model stability on the performance of the approximate model is explored. The openloop performance of both linear and nonlinear models in presence of internal composition inventory control is demonstrated. An error index is developed to quantitatively analyse the accuracy of the linear model.

### REACTIVE DISTILLATION DYNAMICS

The process under consideration is a generic reactive distillation column schematically shown in Figure 1. The system consists of  $N$  stages including a partial reboiler and a total condenser. The main column is further divided into three sections which are stripping section ( $N_s$ ), reactive section ( $N_{RX}$ ) and rectifying section ( $N_r$ ). Thus, we are considering a system in which reaction takes place only in the reactive zone. The elementary reversible reaction considered is given in Figure 1. Product C is the lightest and product D is the heaviest in the system. The reactants A and B are intermediate boilers with component A being a lighter reactant than component B. Thus, the fresh feed stream  $F_A$  containing reactant A is fed from the bottom of reactive tray, while the fresh feed  $F_B$  containing reactant B is fed at the top of the reactive zone. The details of an ideal generic reactive distillation are shown in Table 1.

#### Nonlinear Process Model

A full-order model is employed to simulate the dynamics of the reactive distillation with simple ideal vapour-liquid

Table 1. Column simulation input.

Feed conditions		Column specifications	
Temperature	360 K	Column pressure	9 bar
Feed rates	$F_A = 0.0126 \text{ kmol s}^{-1}$ $F_B = 0.0126 \text{ kmol s}^{-1}$	Rectification stages	7
		Reaction stages	6
		Stripping stages	7
Composition in the two feeds	$Z_a = 100\% \text{ mol A}$ $Z_b = 100\% \text{ mol B}$	Feed stages	$nf1 = 9$ $nf2 = 14$

equilibrium, reaction kinetics and physical properties. The model is based on dynamic mass balance while the energy equations are neglected by assuming constant molar overflow except in the reactive zone where the vapour flowrate increases through the reactive zone due to the cumulative effect of exothermic reaction in all the reactive stages. Constant relative volatilities of all the components are assumed.

We have considered a simplified process model in the present work in order not to cloud the picture with the specific complexities that are typical of many real chemical systems. The simplified process model used in this work is developed to capture the essential dynamics and qualitative properties of a typical reactive distillation column.

The reaction rate of component  $j$  on tray is given by

$$R_{i,j} = \Phi_j M_i (k_{F,i} x_{i,A} x_{i,B} - k_{B,i} x_{i,C} x_{i,D}) \quad (1)$$

where  $\Phi_j$  is the reaction stoichiometric factor of component  $j$  and  $M_i$  is the liquid holdup (kmol) on tray  $i$ . The

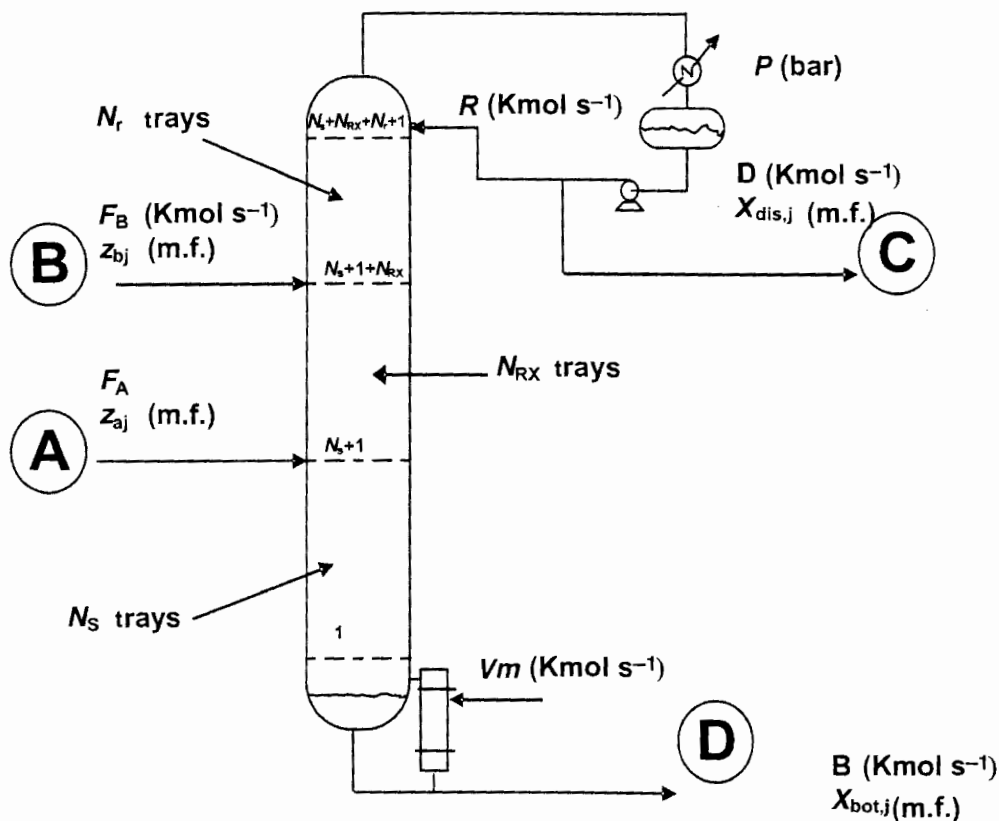


Figure 1. Generic reactive distillation column.

dependence of reaction rate constants  $k_{F,i}$  and  $k_{B,i}$  on temperature  $T_i$  on reactive trays is given by the equations:

$$k_{F,i} = a_F e^{-E_F/RT_i} \quad (2)$$

$$k_{B,i} = a_B e^{-E_B/RT_i} \quad (3)$$

where  $a_F$  and  $a_B$  are the pre-exponential factors,  $E_F$  and  $E_B$  are the activation energies. The absolute temperature  $T_i$  is evaluated using an ideal vapour-liquid equilibrium equation

$$T_i = B_{vp,i} / \left[ A_{vp,i} - \ln \left( \alpha_i P / \sum_{k=1}^{N_c} \alpha_k x_{ik} \right) \right] \quad (4)$$

Mole balances on all the components and the algebraic equation describing the vapour flowrate in the reactive zone give the reactive distillation model:

$$\frac{dx_{ij}}{dt} = [L_{i+1}(x_{i+1,j} - x_{ij}) + V_{i-1}(y_{i-1,j} - x_{ij}) + V_i(x_{i,j} - y_{i,j}) + R_{i,j} + F_i(Z_{ij} - x_{ij})] / M_i \quad (5)$$

$$\frac{dM_i}{dt} = L_{i+1} - L_i + R_{i,j} \lambda / \Delta H_v + F_i \quad (6)$$

$$V_i = Vm + \frac{\lambda}{\Delta H_v} \sum_{k=1}^{i-N_s-1} R_{N_s+1+k,j} \quad (7)$$

where  $Vm$  is the constant vapour boilup ( $\text{kmol s}^{-1}$ ) from the stripping section and  $V_i$  is the vapour flow rate ( $\text{kmol s}^{-1}$ ) on tray  $i$  in the reactive zone.  $F_i$  is the feed flowrate entering the tray  $i$ .  $\lambda$  is the exothermic heat of reaction ( $\text{cal mol}^{-1}$ ) and  $\Delta H_v$  is heat of vaporizations ( $\text{cal mol}^{-1}$ ). Note that the rate of reaction  $R_{i,j}$  is equal to zero in all the nonreactive trays and  $F_i = 0$ , except at  $i = nf1, nf2$  (feed trays). The nonlinear model of reactive distillation can be represented in more compact vector form by decoupling all the state variables in the model. Thus the compacted nonlinear state space equations of the system is given by

$$\frac{dX(t)}{dt} = f(X(t), U(t), d(t); \theta) \quad (8)$$

$$Y = h(X(t)) \quad (9)$$

where the  $5N$ -dimensional vector  $X$  are state variables (liquid mole fractions and holdup in all stages including partial reboiler and total condenser).  $\theta$  represents the model parameters. Reflux flowrate and vapor boil up are the input variables which could be used as manipulated variables in the control strategy, i.e.,  $U = (Vm, R)^T$ . The source of measurable disturbances is the two feed flowrates and compositions, i.e.,  $d = (F_A, F_B, Z_a, Z_b)^T$ .  $Y$  is a vector of measured output variables which could be composition and/or stage temperature.

### Linearized Process Model

To obtain an approximate linear process model, the nonlinear equations (8) and (9) are expanded in Taylor series about the nominal values and the linearized state space

model of reactive distillation is given by

$$\frac{d\hat{x}}{dt} = A\hat{x} + B\hat{u} + E\hat{d} \quad (10)$$

$$\hat{Y} = C\hat{x} \quad (11)$$

where the superscript ' $\wedge$ ' denotes the change in the variable from linearization point. Thus, the elements of matrices  $A$ ,  $B$ ,  $C$  and  $E$  are given by

$$a_{ij} = \left. \frac{\partial f_i}{\partial x_{ij}} \right|_{X_s, U_s, d_s} \quad (12)$$

$$b_{ij} = \left. \frac{\partial f_i}{\partial u_{ij}} \right|_{X_s, U_s, d_s} \quad (13)$$

$$c_{ij} = \left. \frac{\partial h_i}{\partial x_{ij}} \right|_{X_s, U_s, d_s} \quad (14)$$

$$e_{ij} = \left. \frac{\partial f_i}{\partial d_{ij}} \right|_{X_s, U_s, d_s} \quad (15)$$

where  $X_s$ ,  $U_s$  and  $d_s$  are the desired steady state operating conditions. The vector of the of the state variables can be evaluated by

$$X(t) = X_s + \hat{X}(t) \quad (16)$$

### ERROR INDEX

In order to quantitatively assess the performance and accuracy of linear model as compare to rigorous nonlinear model, an error index is defined in term of average relative error (ARE). The numerical values obtained from nonlinear model are considered as the real values for the system, while the values obtained from linear model are taken as the approximate values. In this sense, an ARE is given as

$$ARE = \frac{1}{n} \sum_{k=1}^n abs \left( \frac{\gamma_k^{real} - \gamma_k^{appr}}{\gamma_k^{real}} \right) \times 100 \quad (17)$$

where  $\gamma_k^{real}$  is the real value from the nonlinear model at point  $k$ ,  $\gamma_k^{appr}$  is the approximate value from the linear model at the same point  $k$ .  $n$  is the number of data points.

### STEADY STATE DESIGN DATA

Availability of a suitable and stable steady state values at the desired operating conditions is a fundamental prerequisites to developing a successful linear model. This is normally done by simultaneous solution of a large set of nonlinear algebraic equations of the system. Solving the highly nonlinear coupled algebraic equations of reactive distillation is difficult and often does not converge to the solution. In this work, a dynamic relaxation is used to reach the desired steady state conditions.

The kinetic, physical, and vapour-liquid equilibrium parameter values for reactive distillation column are taken from Luyben (2000). Two steady state designs are used in this study to examine the system sensitivity to input disturbances (see next section). They are termed as low-conversion and high-conversion steady state design. The high conversion is the base design and will be used

Table 2. Base steady state conditions for a high and low-conversion region.

Variables		High-conversion region	Low-conversion region
Flowrates	$V_m$ (kmol s <sup>-1</sup> )	0.0285	0.0281
	R (kmol s <sup>-1</sup> )	0.0331	0.0328
	D (kmol s <sup>-1</sup> )	0.0126	0.0119
	B (kmol s <sup>-1</sup> )	0.0126	0.0133
$X_{dis}$	A	0.0467	0.0345
	B	0.0033	0.0008
	C	0.9501	0.9647
	D	0.0000	0.0000
$X_{bot}$	A	0.0009	0.0519
	B	0.0445	0.0822
	C	0.0000	0.0000
	D	0.9545	0.8658

throughout this study. The low-conversion design is considered to justify the consistency of the linearized model as long as the deviation in process variables due to a disturbance is within the region of the base steady states around which the model is linearized. Olanrewaju and Al-Arfaj (submitted) modified Luyben's (2000) optimum design to ensure openloop stability. In this work, we consider the modified optimum as the base design for high-conversion region. The flowrates of the fresh feed  $F_A$  and  $F_B$  are fixed at 0.0126 kmol s<sup>-1</sup> of pure reactants and the initial holdup in all the trays is 1 kmol. Table 2 shows summary of the of steady state conditions for openloop system.

### STEADY STATE SENSITIVITY

The performance of a linear model is based on the sensitivity of the steady state values to disturbances. The deviation from the steady states when disturbance is introduced into the system must not only be small enough but also be within the region of the steady states used in developing the approximate model. Thus, linearization may lead to inaccurate or false conclusion if the original model exhibits a drastic deviation from the base steady state region. This would also be true if the nonlinear model is unstable under certain disturbances.

Figure 2(a) shows the composition profile of the column comparing linear and nonlinear models when component B fresh feed flowrate ( $F_B$ ) is increased by 2%. The linear and nonlinear models show a consistent deviation within the vicinity of the base steady state composition profiles. As more B is fed into the column, the two models predict shifting of reactant A profile in the middle of the column downward and of reactant B profile upward. The shifting down of product D profile in the stripping section showed by the two models indicates an increase in impurity in the bottoms as a result of excess of reactant B.

Figure 2(b) shows steady state temperature profile of linear and nonlinear models with 2% disturbance in  $F_B$ . There is consistent deviation from base steady state temperature profile, which indicates that a linear model predicts the original nonlinear model well within the desired steady state region. Note that both the models show that the temperature in the stripping section is reduced as a result of more B in the bottoms, and temperature in the rectifying section is increased due to some of B which is lost to the overhead.

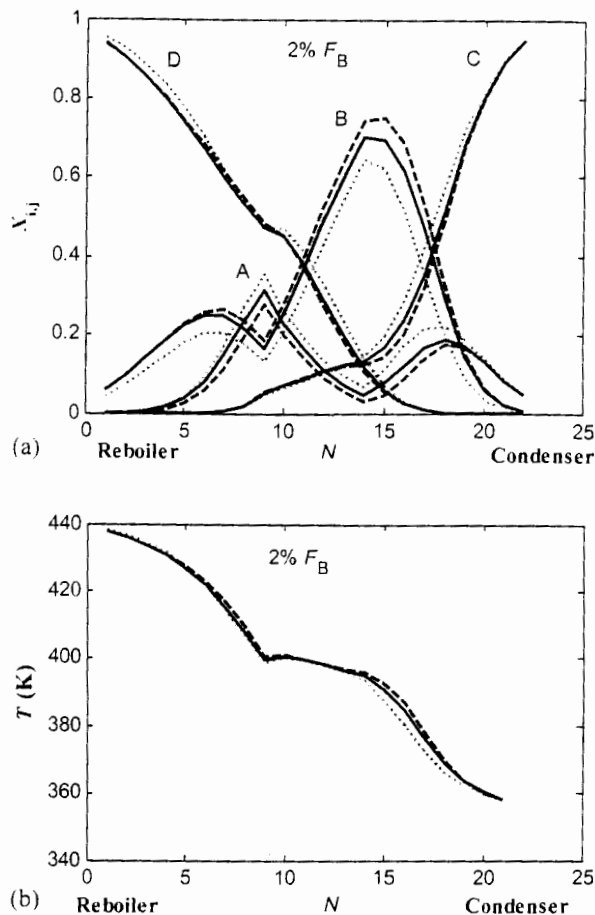


Figure 2. (a) Composition profiles with 2% increase in  $F_B$ . (b) Temperature profiles with 2% increase in  $F_B$ . (---) Base steady state profile with no disturbance; (—) linear model; (—) nonlinear model.

Figure 3(a) compares the composition profile for both linear and nonlinear models when fresh feed flowrate of reactant A ( $F_A$ ) is increased by 2%. Linear model predicts the composition profiles in the vicinity of the base composition profile as it was developed around that profile. In contrast, the nonlinear model shows a significant deviation in composition profile from the base steady state values. When there is an excess of reactant A, the light reactant, there must be an increase in the heat duty to strip out the unreacted A from product D. Since the heat duty is kept constant in this steady state analysis, this resulted in reactant A flooding the stripping section as predicted by nonlinear model.

Figure 3(b) shows the temperature profiles of both models under the same conditions, i.e., +2% in  $F_A$ . The linear model shows a slight deviation around the base steady state, whereas nonlinear model shows a significant change in the temperature profile along the column. The sharp drop in temperatures predicted by nonlinear model especially in the stripping section is the result of excess of unreacted reactant A in the zone.

When  $F_A$  is increased by 2%, the system drifts to another low conversion state. It is expected that the linear model will not predict the drift since this is a nonlinear characteristics of the system. However, to verify the linear model applicability

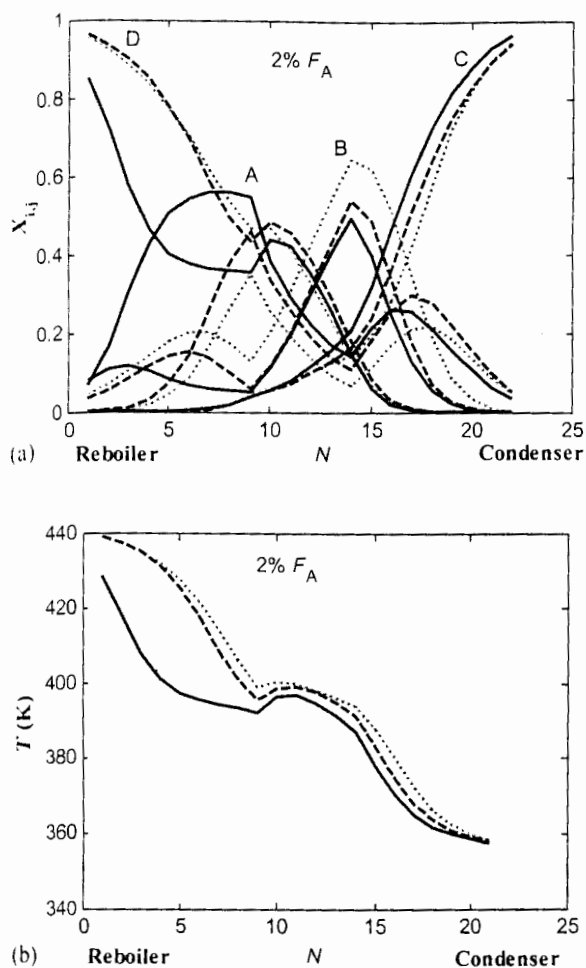


Figure 3. (a) Composition profiles with 2% increase in  $F_A$ . (b) Temperature profiles with 2% increase in  $F_A$ . (---) Base steady state profile; (—) linear model; (—) nonlinear model.

when the system is openloop stable under a given disturbance, the low conversion state is taken as a new base (see Table 1) and the model is linearized around that design, then similar disturbance is introduced. Figure 4 shows the composition profiles at low conversion steady state region. Since the system of that state is stable under the same disturbance (+2%  $F_A$ ), both linear and nonlinear models show a good matching and consistent deviations from the low conversion base steady state profiles.

This indicates that a linear model will give good predictions of a nonlinear model when the base steady state design is openloop stable under a given disturbance. In contrast, it may lead to false predictions whenever the disturbance from the base design is either significant or results in a drift to another steady state region, i.e., openloop unstable.

### ROBUSTNESS OF LINEAR MODEL

Assessing the robustness of a linear model under various magnitudes of disturbances is very important before its applicability can be considered. In this section, the performance of openloop dynamic linear model is compared

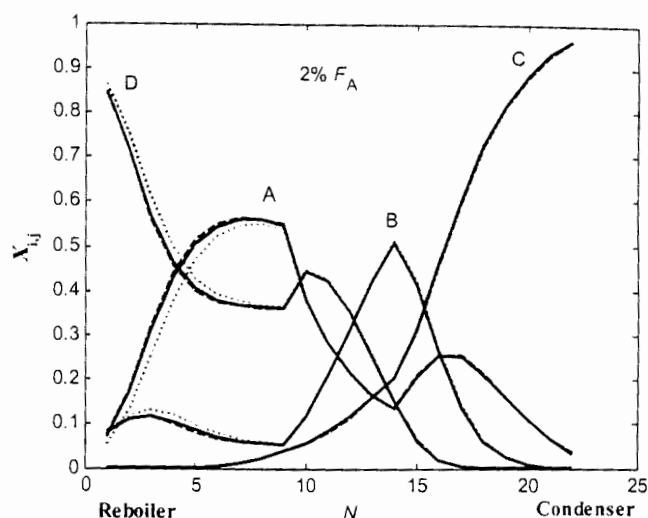


Figure 4. Composition profiles with 2% increase in  $F_A$ . (---) Base steady state profile; (—) linear model; (—) nonlinear model.

to that of a rigorous dynamic nonlinear model. Two dynamic scenarios are investigated:

- (1) Openloop (OL): where only the pressure as well as the base and reflux drum level inventories are controlled while  $F_A$ ,  $F_B$ ,  $Z_a$ ,  $Z_b$ ,  $V_m$  and  $R$  are fixed.
- (2) Openloop with internal composition control (OL + IC): in addition to level control loops, reactant A inventory is controlled through an the internal composition controller by manipulating the feed flowrate  $F_A$ .

The process variables considered as sources of disturbances are:

- (1) Feed flowrate of reactant B ( $F_B$ )
- (2) Feed composition of reactant A ( $Z_a$ )
- (3) Vapour boilup ( $V_m$ )

The two models are excited by step change of magnitudes 1%, 2%, 5% and 10%. These changes in  $Z_a$  are the percentage amount of reactant B in reactant A fresh feed. Due to space limitations, the responses of the two models when  $F_B$  is changed are presented. The average relative error of all the disturbances studied under various magnitudes is summarized in Tables 3 and 4.

### Openloop Model (OL)

In this scenario, the inventory loops incorporated into the system are: the pressure (controlled by the heat removal from the condenser), the reflux drum level (controlled by the distillate flowrate) and the base level (controlled by the bottoms flowrate). Figure 5 shows the steady state composition and temperature profiles of linear and nonlinear models with 5% change in  $F_B$ . Comparing this result with that shown in Figure 2 but with 2% change in  $F_B$  shows that the prediction of linear model becomes poor as the magnitude of the disturbance is increased. Note that both temperature and composition profiles along the length of the column show that the difference between the linear and nonlinear models is most significant at feed trays. This gives an indication of higher nonlinearity effect in feed trays than any other parts of the column. There are many reasons that could be

Table 3. The average relative error for openloop model without internal controller (OL).

System: Open model (OL)												
Variable/Magnitude	Variable											
	$F_B$				%mol of B in feed A				$V_m$			
	1%	2%	5%	10%	1%	2%	5%	10%	1%	2%	5%	10%
Bulk steady state												
$X_{i,A}$	4.09	8.51	23.82	67.64	15.55	29.73	44.69	72.10	14.88	23.99	36.34	71.55
$X_{i,B}$	2.46	6.62	15.16	45.21	9.64	12.93	16.14	39.63	11.48	15.41	23.83	38.82
$X_{i,C}$	2.24	6.21	14.71	20.09	8.62	18.33	23.33	33.29	17.63	31.48	50.86	62.13
$X_{i,D}$	1.42	4.77	10.97	18.38	6.51	12.17	26.72	58.81	11.13	14.74	21.97	23.78
$T$	0.05	0.21	0.54	0.98	0.29	0.72	0.84	1.34	0.55	0.88	1.01	1.98
Individual variables												
Steady state												
D	0.02	0.06	0.67	2.60	0.02	0.26	4.24	11.87	0.06	0.67	3.01	6.85
B	0.02	0.06	0.68	2.79	0.02	0.26	3.88	9.78	0.06	0.68	2.96	7.35
$X_{bot,D}$	0.03	0.07	1.02	4.28	0.10	0.65	5.36	14.64	0.12	0.43	1.32	1.87
$X_{dis,C}$	0.01	0.02	0.27	1.13	0.20	0.44	1.35	3.55	0.02	0.21	3.18	11.67
$X_{nfl,A}$	3.37	10.55	18.74	23.89	14.21	23.24	32.09	36.30	17.21	23.12	33.12	59.34
$X_{nfl2,B}$	1.82	5.31	6.07	10.33	6.13	11.94	9.82	22.82	8.43	9.78	13.87	25.24
Dynamic												
D	0.03	0.09	0.62	2.58	0.02	0.24	3.90	11.38	0.07	0.67	2.96	6.58
B	0.03	0.09	0.61	2.42	0.02	0.24	3.79	9.38	0.07	0.69	2.65	8.27
$X_{bot,D}$	0.05	0.14	0.91	3.85	0.16	0.65	4.82	13.77	0.11	0.45	1.29	1.44
$X_{dis,C}$	0.01	0.02	0.21	1.01	0.22	0.45	1.30	3.356	0.02	0.26	3.24	10.44
$X_{nfl,A}$	2.31	7.30	16.41	17.76	10.29	27.23	30.84	35.75	15.80	21.08	32.23	58.68
$X_{nfl2,B}$	1.20	3.66	8.98	15.98	4.306	9.66	10.81	22.31	7.11	9.25	14.15	24.95

responsible for this behaviour. First, higher concentration of reactants in these trays indicates places with higher reaction rates than any other parts of reactive zone. Secondly, these trays serve as possible entrance of disturbances into the column. Thirdly, these trays are the locations in the

column with high interactive effect of reaction kinetics and separation.

Figure 6 shows the dynamic response of the bottoms flowrate (B) for both linear and nonlinear models to an increase with different magnitudes in  $F_B$ . This is shown

Table 4. The average relative error for openloop model with internal controller (OL + IC).

System: Open model with internal controller (OL + IC)												
Variable/Magnitude	Variable											
	$F_B$				%mol B in feed A				$V_m$			
	1%	2%	5%	10%	1%	2%	5%	10%	1%	2%	5%	10%
Bulk steady state												
$X_{i,A}$	0.22	0.62	3.23	7.33	0.12	0.44	2.43	5.29	2.69	8.99	34.75	55.65
$X_{i,B}$	0.18	0.64	3.00	8.35	0.12	0.44	2.17	6.02	2.38	6.12	20.83	37.56
$X_{i,C}$	0.20	0.75	3.78	11.46	0.17	0.61	3.07	8.88	3.20	10.83	43.06	59.55
$X_{i,D}$	0.19	0.54	2.91	10.60	0.10	0.32	1.42	6.67	2.35	10.27	17.82	21.69
$T$	0.04	0.05	0.07	0.26	0.05	0.06	0.13	0.41	0.08	0.28	0.71	1.21
Individual variables												
Steady state												
$F_A$	0.03	0.12	0.66	2.23	0.004	0.01	0.06	0.27	0.26	0.92	1.30	0.37
D	0.03	0.11	0.61	2.05	0.01	0.05	0.32	1.20	0.18	0.59	1.48	4.10
B	0.002	0.01	0.04	0.15	0.02	0.06	0.38	1.43	0.07	0.34	0.25	5.62
$X_{bot,D}$	0.01	0.06	0.30	0.95	0.02	0.08	0.50	1.92	0.06	0.12	1.17	1.21
$X_{dis,C}$	0.01	0.03	0.19	0.63	0.01	0.02	0.11	0.42	0.09	0.34	2.21	10.44
$X_{nfl,A}$	0.01	0.07	0.39	1.34	0.001	0.01	0.04	0.20	0.15	0.55	0.80	0.93
$X_{nfl2,B}$	0.09	0.33	1.82	5.96	0.02	0.07	0.45	1.61	0.92	3.14	3.76	1.64
Dynamic												
$F_A$	0.02	0.09	0.48	1.67	0.01	0.02	0.13	0.51	0.18	0.64	1.32	1.35
D	0.02	0.08	0.46	1.61	0.01	0.04	0.24	0.92	0.15	0.48	1.48	3.93
B	0.002	0.006	0.04	0.13	0.02	0.06	0.36	1.40	0.04	0.20	0.25	5.22
$X_{bot,D}$	0.01	0.04	0.25	0.87	0.02	0.07	0.42	1.68	0.06	0.16	1.04	1.18
$X_{dis,C}$	0.005	0.02	0.10	0.37	0.002	0.01	0.07	0.26	0.05	0.20	1.76	9.19
$X_{nfl,A}$	0.01	0.05	0.28	1.01	0.003	0.01	0.08	0.37	0.11	0.40	0.84	0.92
$X_{nfl2,B}$	0.06	0.25	1.36	4.54	0.01	0.05	0.28	1.03	0.64	2.22	3.91	3.98

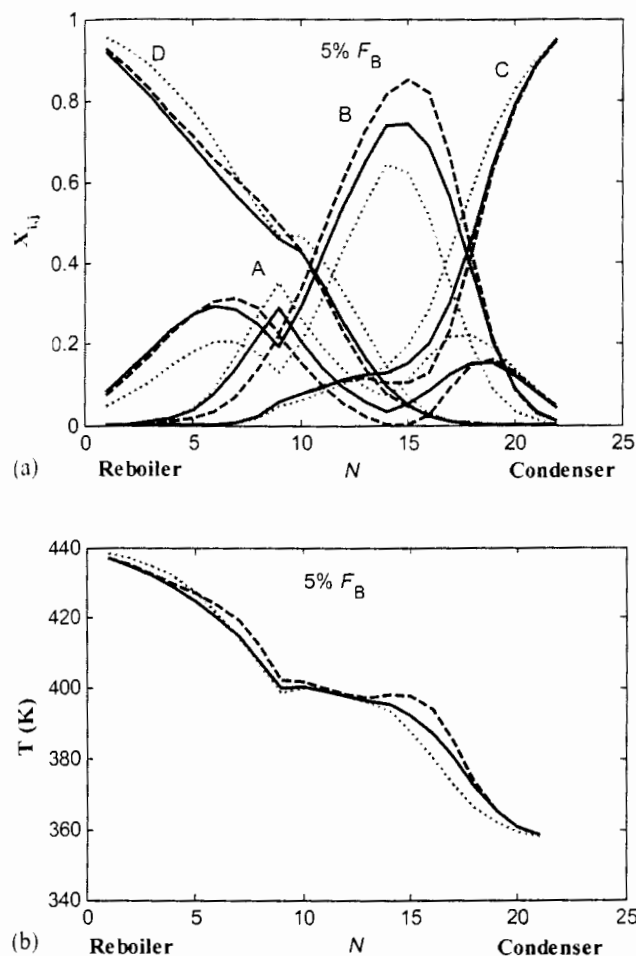


Figure 5. (a) Composition profiles with 5% increase in  $F_B$ . (b) Temperature profiles with 5% increase in  $F_B$ . (---) Base steady state profile; (---) linear model; (—) nonlinear model.

as an illustration of the output performance of the linear model as compared to that of rigorous model. The two models show an increase in the bottoms flowrate (B) with increase in reactant B due to the excess of unreacted B that goes down to the bottoms of the column. Figure 7

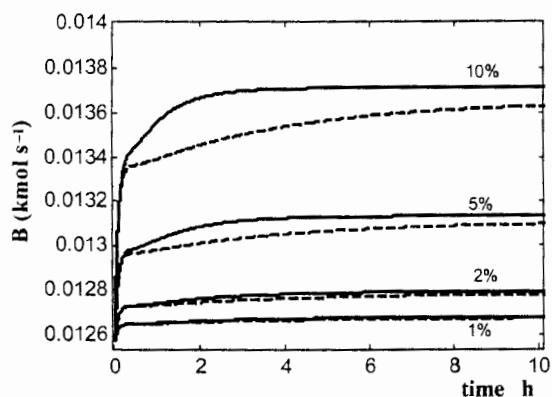


Figure 6. Dynamic response of bottoms flowrate (B) to different magnitude of increase in  $F_B$ . (---) Linear model; (—) nonlinear model.

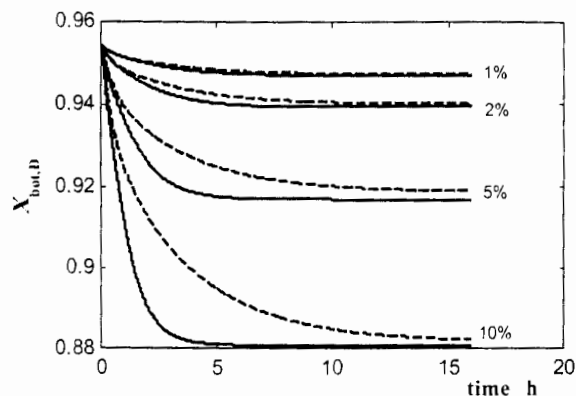


Figure 7. Dynamic response of the composition of component D to different magnitude of increase in  $F_B$ . (---) Linear model; (—) nonlinear model.

compares the dynamic performance of the composition of product D for the linear model to that of nonlinear model at different magnitude of increase in  $F_B$ . The two models show a decrease in the concentration of D due to increase in concentration of reactant B in the bottoms.

In all the dynamic comparisons that are carried out between the two models, the linear model shows a good performance at small magnitude of disturbance and the deviation between linear and nonlinear models increases with increase in the magnitude of disturbances (see Table 3). The details of linear model accuracy are discussed in the next main section 7.

### Openloop Model with Internal Composition Controller (OL + IC)

Several papers have reported the use of internal composition measurement in the closeloop control of reactive distillation with multiple feeds to maintain reactants stoichiometry (Al-Arfaj and Luyben, 2000). The inclusion of internal composition controller (to balance the reaction stoichiometry) is used to demonstrate the enhancement of openloop performance of both linear and nonlinear models. The concentration of reactant A in the first tray of reactive zone (numbered from the bottoms) is controlled by manipulating the fresh feed flowrate of reactant A ( $F_A$ ). The internal composition control equation is given as

$$F_A = F_{Ass} + Kc(X_{set} - X_{nfl,A}) \quad (18)$$

where  $F_{Ass}$  is  $F_A$  at the steady state,  $Kc$  is the controller gain and  $X_{set}$  is the internal composition setpoint. The controller used is  $P$ -only controller because the objective is to maintain reactant A inventory and not to fix the composition at that stage. A relay feedback test (Luyben, 1987) on both the linear and nonlinear process models is employed to obtain the ultimate gain. The controllers are tuned using the Tyreus-Luyben tuning method (Tyreus and Luyben, 1992). The controllers gains for the two process models are:

$$Kc \text{ (linear model)} = 1.52 \quad (19)$$

$$Kc \text{ (nonlinear model)} = 1.28 \quad (20)$$

Figure 8 shows the steady state composition profiles for both linear and nonlinear models when the internal



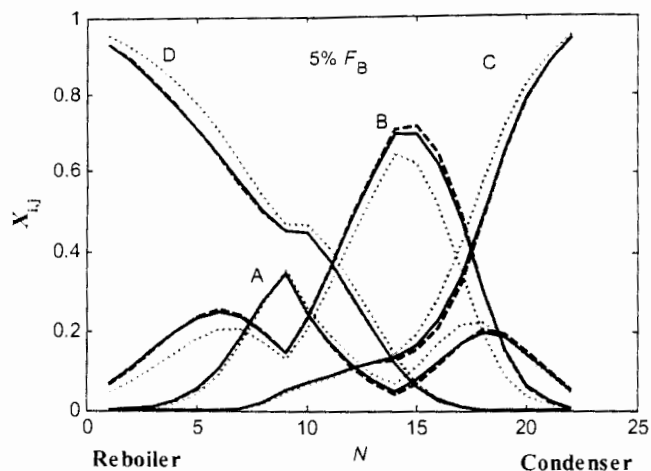


Figure 8. OL + IC: composition profiles with 5% increase in  $F_B$ . (---) Base steady state profile; (---) linear model; (—) nonlinear model.

composition controller is included. The source of disturbance is 5% increase in feed  $F_B$ . The linear model demonstrates a better performance and approximation of nonlinear model when compared with the same results shown in Figure 5, where no internal composition controller is used.

Figure 9 compares the dynamic response of the internal composition controller ( $X_{nfl,A}$  and  $F_A$ ) for both linear and nonlinear models. The disturbances are various magnitudes of positive step changes in  $F_B$ . The increase in the amount of reactant B fed into the column reduces the internal composition of reactant A. Controllers based on the two models

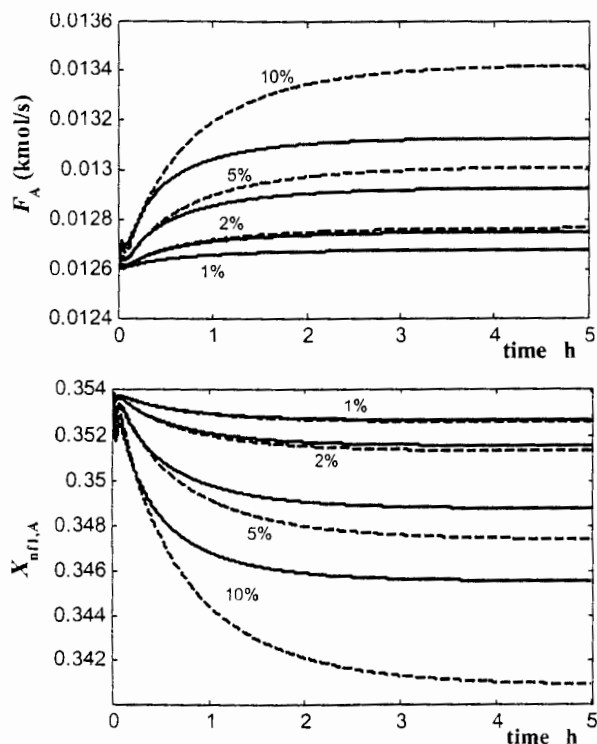


Figure 9. OL + IC: dynamic response of flowrate ( $F_A$ ) and composition of reactant A on tray  $n/1$  to different magnitude of increase in  $F_B$ . (---) Linear model; (—) nonlinear model.

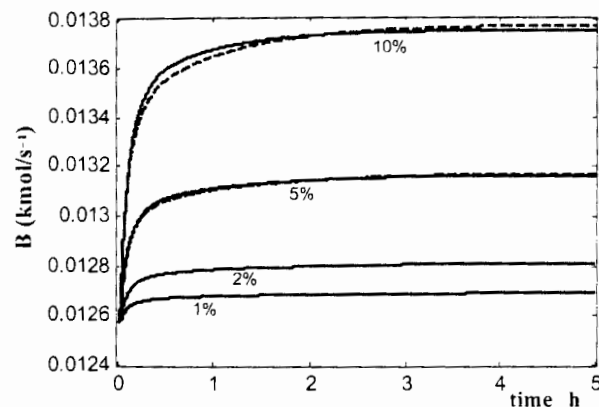


Figure 10. OL + IC: dynamic response of bottoms flowrate ( $B$ ) to different magnitude of increase in  $F_B$ . (---) Linear model; (—) nonlinear model.

respond adequately by increasing the feed flowrate  $F_A$  to counteract the gradual buildup of reactant B in reactive zone. The response time of the two models is comparable at lower magnitudes of disturbance. However, as the magnitude of disturbance increases linear model responds slower and predicted higher amount of  $F_A$  than that of the nonlinear model. The deviation between the two models increases with increase in disturbance magnitude.

Figure 10 shows the dynamic response of bottoms flowrate ( $B$ ) for both linear and nonlinear models to different magnitude of changes in  $F_B$ . Figure 11 compares the dynamic performance of the composition of product D for a linear model to that of nonlinear model at different magnitude of changes in  $F_B$ . The results show a good approximation of rigorous model by the linear model.

#### ACCURACY OF LINEAR MODEL

In this section, the accuracy of a linear model with and without internal composition controller is quantified using the error index defined in the Error Index Section. The quantification of model error based on various magnitudes of disturbance in feed flowrate  $F_B$ , feed composition of reactant A (reactant B in  $F_A$ ) and change in vapour

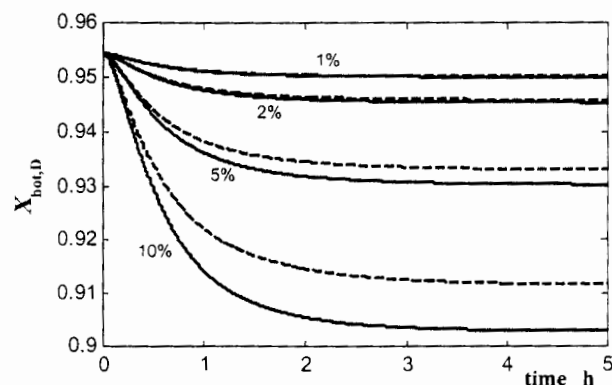


Figure 11. OL + IC: dynamic response of the composition of component D to different magnitude of increase in  $F_B$ . (---) Linear model; (—) nonlinear model.



boilup ( $V_m$ ) are studied. Three major categories were used to classify the ARE of the system.

- (1) Bulk steady state: in this category, the ARE of a given variable is averaged out over the number of stages. For example, the bulk steady state temperature is the sum of temperature in all the stages divided by the total number of stages.
- (2) Individual steady state: this is the ARE of a given variable at steady state.
- (3) Individual dynamic variable: this is the average ARE of a given variable over the time required to reach steady state.

Table 3 presents the summary of the average relative error of the system without internal composition inventory. Figure 12 shows the ARE of the bulk steady state composition profiles with disturbance in  $F_B$ . The results indicate that an approximation of the rigorous model with the linearized model without internal composition controller could be acceptable when the magnitude change in feed flowrate is below 6%. Similar conclusion could be reached when the ARE of the system are quantified based on individual dynamic and steady state of the system variables (see Figure 13). The ARE of the bulk composition in the OC + IC scenario is around 10% when  $F_B$  is increased by 10% while it is ranging between 20–40% if the internal composition is not included. The ARE of the system is significantly reduced when the internal controller is included in the openloop system (see Figures 12(b) and 13), which suggest that the performance of linear system is acceptable with disturbance magnitude more than 10% if the ARE tolerance is less than 20%. The details of average relative error for the OL + IC scenario are presented in Table 4.

Figure 14 compares the impact of disturbance from different system variables (i.e.,  $F_B$ ,  $Z_a$  and  $V_m$ ) on the

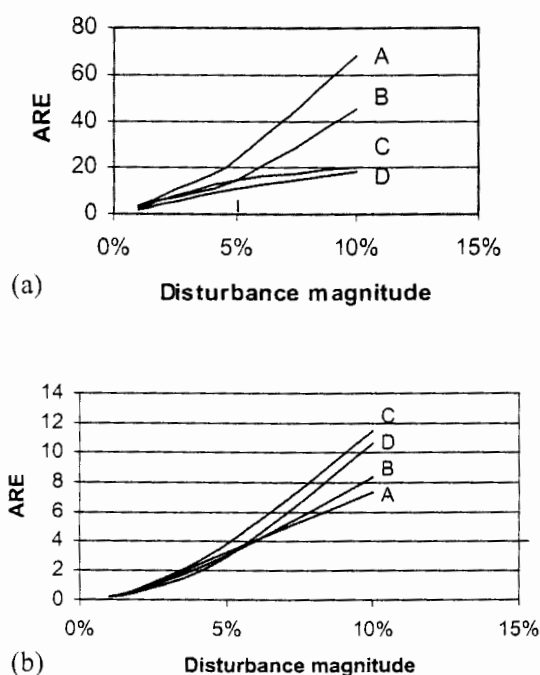


Figure 12. The ARE of the bulk steady state composition profiles with disturbance in  $F_B$ . (a) OL; (b) OL + IC.

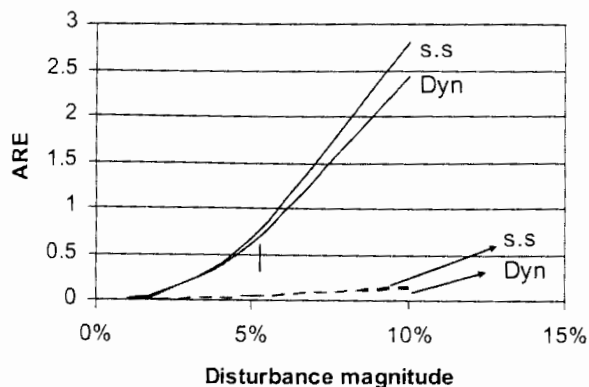


Figure 13. The ARE of the individual dynamic (Dyn) and steady state (s.s.) of bottoms flowrate (B) with disturbance in  $F_B$ . (—) OL; (---) OL + IC.

performance of the linearized model using their ARE. We have used the bulk steady state temperature profiles for this comparison because it represents the cumulative effect of system dynamics. Introduction of disturbance from the feed composition (reactant B in  $F_A$ ) is shown to have higher ARE than from feed flowrate ( $F_B$ ). This suggests that disturbance in feed composition affect the internal composition and increases the system nonlinearity more than that made by disturbing the system from feed flowrate. Exciting the system by changing the vapour boilup shows the highest trend of error because it impacts both the reaction kinetics and the separation capacity of the system, and thus, the system nonlinearity.

#### Effect of $K_c$ on the Accuracy of Linear Model

The accuracy of a linear processes model when compared its performance to that of a nonlinear process model could depend on the value of the controller gain. To illustrate this, the effect of internal composition controller gain ( $K_c$ ) on the accuracy of linear model is explored at different values of  $K_c$ . Figure 15 shows the ARE of the individual dynamics of  $X_{bot,D}$  and  $X_{dis,C}$  for different values of controller gain. The result indicates that using a controller gain smaller in magnitudes than that of a well tuned controller gain ( $K_c = 1.52$ ) will result in higher value of ARE. On the other hand, using the higher value of  $K_c$  does not

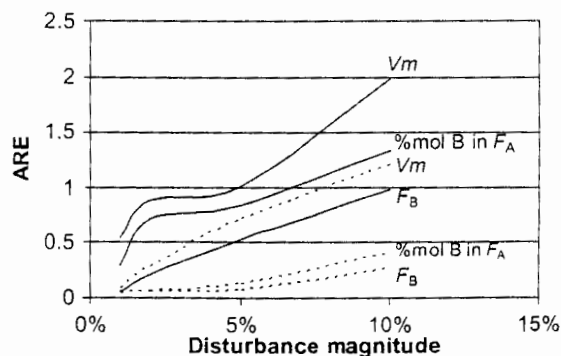


Figure 14. The ARE of the bulk steady state temperature profiles with disturbances in  $V_m$ ,  $F_B$ , %mol B in  $F_A$ . (—) OL; (---) OL + IC.

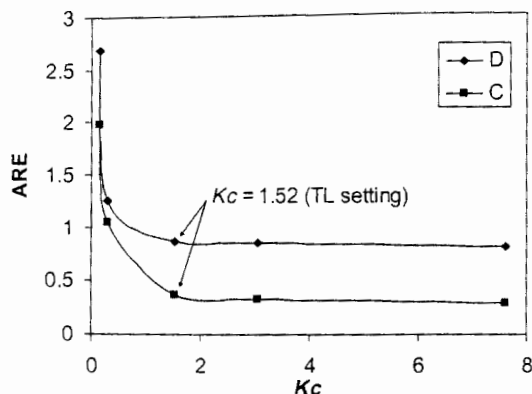


Figure 15. The ARE of the individual dynamic (Dyn) of  $X_{dis,C}$  and  $X_{bot,D}$  showing the effect of internal composition controller gain on the accuracy of linear model.

significantly reduce the ARE. Therefore, it can be concluded that a well tuned controller gain, which is as high as the margin of stability allows is a necessity for a reasonable performance of a linear process model when an internal composition controller is included.

### Summary

Critical comparison of the performance of the openloop linear model with and without the internal composition controller reveals the following important points.

- (1) The performance of linear model is improved with the inclusion of internal composition controller, which suggests the degree of nonlinearity in nonlinear model is reduced when the stoichiometry balance of reaction in the reactive zone is maintained. The average relative error of the linear model to nonlinear model is reduced even at higher magnitude of disturbance when the internal composition controller is included (see Tables 3 and 4).
- (2) The settling time of linear model with internal controller is shorter than that without the internal controller, which is an indication of better system stability.
- (3) For implementation purposes, it is recommended to use the linearized model whenever the ratio of disturbance magnitude to the tolerable model error is not greater than 1 and that the system is openloop stable under that magnitude of change. For example, if the tolerable model error is 20% then the linearized model could be used for disturbance magnitude up to 20%.
- (4) It is expected that the closeloop performance (with either single end or dual end quality control) based on a linearized model will be reasonably close to the nonlinear model.

### CONCLUSION

In this study, we have compared the openloop performance of a linearized dynamic model of a generic reactive distillation system with that of nonlinear model. The approximate linear model nicely averages the process nonlinearities when the magnitude of input change is small and becomes inadequate as the deviation from the base steady

states increases with increase in the magnitude of disturbance. The effect of various step input changes on the performance of the approximate model was explored. The linear model could be used to approximate the behaviour of the system if the magnitude of the disturbance is less than 6% when there is no internal composition controller.

It is shown that the performance and robustness of linear model is enhanced with inclusion of internal composition inventory control in the openloop model of the system. When the internal composition controller is included, the linearized model could be used to approximate the nonlinear model up to a disturbance magnitude equals to the tolerable model error provided that the system is openloop stable. If the system shifts from the base state to another state under the influence of a disturbance, then linearizing around the base state will result in a model that provides false conclusion.

It is found that the accuracy of a linear processes model when compared its performance to that of a nonlinear process model could depend on the value of the controller gain. A poor design of the internal composition controller gain could decrease the accuracy of the linear process model. It is recommended that the controller gain should be well designed and tuned to obtain a reasonable performance of the linear process model when an internal composition controller is included.

### NOMENCLATURE

A	reactant component
A	matrix of state variables of the linearized process
$A_{vp,j}$	vapour pressure constants for component $j$
B	matrix of inputs of the linearized process
$B_{vp,j}$	vapour pressure constants for component $j$
B	reactant component
B	bottoms flow rate, $\text{kmol s}^{-1}$
C	matrix of outputs of the linearized process
C	product component
d	disturbance variable vector
$d_s$	disturbance variable at the steady state
D	product component
D	distillate flow rate, $\text{kmol s}^{-1}$
E	matrix of disturbances of the linearized process
$F_A$	fresh feed flow rate of reactant A, $\text{kmol s}^{-1}$
$F_B$	fresh feed flow rate of reactant B, ( $\text{kmol s}^{-1}$ )
Kc	controller gain
$K_F$	specific reaction rate of the forward reaction, $\text{kmol s}^{-1} \text{ kmol}^{-1}$
$K_B$	specific reaction rate of the reverse reaction, $\text{kmol s}^{-1} \text{ kmol}^{-1}$
$M_i$	liquid holdup in all stages, kmol
n	number of data points
N	total number of stages including reboiler and reflux drum
Nc	number of component
nf1	first tray of reactive tray
nf2	last tray of reactive tray
$N_r$	number of reactive trays
$N_{RX}$	number of reactive trays
$N_S$	number of stripping trays
$N_T$	total number of trays
P	total pressure, bar
R	reflux flow rate, $\text{kmol s}^{-1}$
$R_{i,j}$	rate of production of component $j$ on tray $i$ , $\text{kmol s}^{-1}$
$T_i$	temperature in tray $i$ , K
U	input variable vector
$U_s$	disturbance variable at the steady state
$V_i$	vapour flow rate from tray $i$ , $\text{kmol s}^{-1}$
$V_m$	vapour boil up from reboiler, $\text{kmol s}^{-1}$
$V_n$	vapour flow rate in rectifying section $\text{kmol s}^{-1}$

$X_{dis,C}$	composition of C in the distillate
$X_{bol,D}$	composition of D in the bottoms
$X_{nfl,A}$	composition of A in tray $n/1$
$X_{nfl,B}$	composition of B in tray $n/2$
$X$	state vector
$X_s$	steady state values
$X(t)$	the state variable at time $t$
$x_{ij}$	liquid composition of component $j$ on tray $i$
$Z_a$	composition of reactant A fresh feed
$Z_b$	composition of reactant B fresh feed
$\alpha_j$	relative volatility of component $j$ with respect to heavy component
$\Delta H_v$	heat of vaporization, cal mol <sup>-1</sup>
$\lambda$	heat of reaction, cal mol <sup>-1</sup>

## REFERENCES

- Al-Arfaj, M.A. and Luyben, W.L., 2000, Comparison of alternative control structures for an ideal two-product reactive distillation column, *Ind Eng Chem Res*, 39: 3298–3307.
- Al-Arfaj, M.A. and Luyben, W.L., 2002, Comparative control study of ideal and methyl acetate reactive distillation, *Chem Eng Sci*, 24(57): 5039–5050.
- Al-Arfaj, M.A. and Luyben, W.L., 2004, Plantwide control for TAME production using reactive distillation, *AIChE J*, 50: 1462–1473.
- Fuentes, C. and Luyben, W.L., 1983, Comparison of energy models for distillation columns, *Ind Eng Chem Res*, 21: 321–325.
- Luyben, W.L., 1987, Derivation of transfer functions for highly nonlinear distillation Columns, *Ind Eng Chem Res*, 26: 2490–2495.
- Luyben, W.L., 2000, Economic and dynamic impact of the use of excess reactant in reactive distillation systems, *Ind Eng Chem Res*, 39: 2935–2946.
- Malone, M.F. and Doherty, M.F., 2000, Reactive distillation, *Ind Eng Chem Res*, 39: 3953.
- Marquardt, W. and Amrhein, M., 1994, Development of a linear distillation model from design data for process control, *Comp Chem Engng*, 18: S349–S353.
- Olanrewaju, M.J. and Al-Arfaj, M.A., Development and application of linear process model in estimation and control of reactive distillation, *Comp Chem Engng*, (submitted).
- Skogestad, S. and Morari, M., 1987, The dominant time constant for distillation columns, *Comp Chem Eng*, 11: 607–617.
- Sundmacher, K. and Kienle, A. (eds), 2003, Reactive distillation: status and future directions (Wiley-VCH, Weinheim, Germany).
- Taylor, R. and Krishna, R., 2000, Review modeling reactive distillation, *Chem Eng Sci*, 55: 5183–5229.
- Tyreus, B.D. and Luyben, W.L., 1992, Tuning PI controllers for integrator/dead time processes, *Ind Eng Chem Res*, 31: 2625–2628.

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