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The use of dilute acetic acid for butyl acetate production in a reactive distillation: Simulation and control studies

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Abstract−The recovery of dilute acetic acid, which is widely found as a by-product in many chemical and petrochemical industries, becomes an important issue due to economic and environmental awareness. In general, separation of acetic acid in aqueous solution by conventional distillation columns is difficult, requiring a column with many stages and high energy consumption. As a result, the primary concern of the present study is the application of reactive distillation as a potential alternative method to recover dilute acetic acid. The direct use of dilute acetic acid as reactant for esterification with butanol to produce butyl acetate in the reactive distillation is investigated. Simulation studies are performed in order to investigate effect of the concentration of dilute acetic acid and key process parameters on the performance of the reactive distillation in terms of acetic acid conversion and butyl acetate production. In addition, three alternative control strategies are studied for the closed loop control of the reactive distillation. The control objective is to maintain the butyl acetate in a bottom product stream at the desired purity of 99.5 wt%.

Key words: Dilute Acetic Acid, Reactive Distillation, Butyl Acetate, Simulation, Control

INTRODUCTION

The aqueous solution of acetic acid is normally found as a by-product from many chemical and petrochemical processes. Examples of such relevant processes include the production of cellulose acetate, an ester group of cellulose used in lacquers and the photographic film process, which is typically associated with a 35 wt% aqueous solution of acetic acid as a waste stream. Other important processes involve the synthesis of terephthalic acid and glyoxal, which have by-products of dilute acetic acid streams of typically 70 wt% and 13-20 wt%, respectively [1,2].

Recovery of dilute acetic acid, therefore, becomes an important issue due to economic and environmental awareness. The conventional process applied to separate acetic acid from its aqueous solution involves a dehydration process by using a distillation column, and then the high-purity acetic acid obtained is used as a raw material in the synthesis of many valuable chemical compounds. Due to the special properties of acetic acid, separation with distillation is difficult and expensive, requiring a column with many stages and high energy consumption. An alternative process for acetic acid recovery is an extraction by using a suitable solvent; however, it is limited by phase separation [1,3]. Furthermore, additional steps to recover and recycle the solvent are necessary.

Recently, a number of researches have been focused on the implementation of reactive distillation as a promising alternative to recover dilute acetic acid. In comparison with the traditional approach of separation followed by reaction processes, performing the chemical reaction and separation in a single reactive distillation column offers advantages not only to separate acetic acid from its aqueous solution but also to produce a valuable product at the same time, thereby reducing capital and energy costs. Following this approach, the direct utilization of dilute acetic acid as a reactant for the production of a high-valued ester, a common solvent used in chemical industries, has received much attention [2,4]. In general, the esterification of acetic acid and different alcohols is represented by the following reaction:

$$\text{Acetic Acid}+\text{Alcohol}\Leftrightarrow\text{Acetate}+\text{Water} \tag{1}$$

Since the esterification reaction is equilibrium limited, the use of the reactive distillation is an attractive method by removing products from the reaction mixture, which leads to an increase in the reactant conversion. In addition, it was reported that a ternary azeotropic mixture of acetate, alcohol and water is found [5,6], thus resulting in difficulties in downstream separation if a traditional production process is utilized. However, the reactive distillation may be effective in order to deal with these difficulties.

Generally, the design and operation of reactive distillation systems are considerably more complex than those of conventional reactors or distillation. Nonlinear phenomena, which are caused by interactions of reaction kinetics, phase equilibrium and mass transfer, and strong influences of various operating parameters, lead to the complicated behavior of the systems [7]. Although a large number of research efforts have been conducted to investigate and design reactive distillations with various systems [8-11], detailed design of reactive distillation for mixtures exhibiting phase-split behavior in a decanter is less obvious. Another important issue to be considered for the successful application of reactive distillation col-

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umns to recover dilute acetic acid is the design of control systems to maintain the columns at a desired condition. However, there is limited research concerning the closed-loop control of reactive distillations [12]. For examples, Al-Arfaj and Luyben [13] evaluated six alternative control structures for an ideal two-product reactive distillation column. In all the schemes, a composition analyzer was used in the reactive zone to maintain stoichiometric balance. Wang et al. [14] studied a reactive distillation column for butyl acetate production from a pure acetic acid under steady-state condition and designed the control strategy of the column. Product quality was maintained by controlling the temperature of the stage inside the column.

In this work, the implementation of a reactive distillation to recover dilute acetic acid via esterification with butanol for the production of butyl acetate, a relatively important solvent, is investigated. Simulation studies of the effect of dilute acetic acid concentration on the reactive distillation performance in terms of acetic acid conversion and butyl acetate production are carried out by using HYSYS commercial software. The effects of key design and operating variables on the performance of the reactive distillation are also evaluated in order to determine an optimal configuration of the reactive distillation column. Finally, the closed-loop control of the reactive distillation column based on the obtained suitable column configuration and specifications is considered.

SIMULATION OF REACTIVE DISTILLATION

A steady-state simulation of a reactive distillation for the production of butyl acetate from esterification of dilute acetic acid and butanol is performed by using HYSYS commercial software. A rigorous equilibrium stage model and NRTL model for describing thermodynamic properties and phase equilibriums are used [15,16]. The configuration of a reactive distillation column under the study is shown in Fig. 1. There are three zones in the column. The rectifying and stripping zones operate exactly as a nonreactive distillation column in order to purify top and bottom products, respectively. Butyl acetate (BuOAc) and water (H_2O) are formed in the reactive zone where the esterification of acetic acid (HOAc) and butanol (BuOH) as in Eq. (2) occurs. The kinetic expressions of this reac-

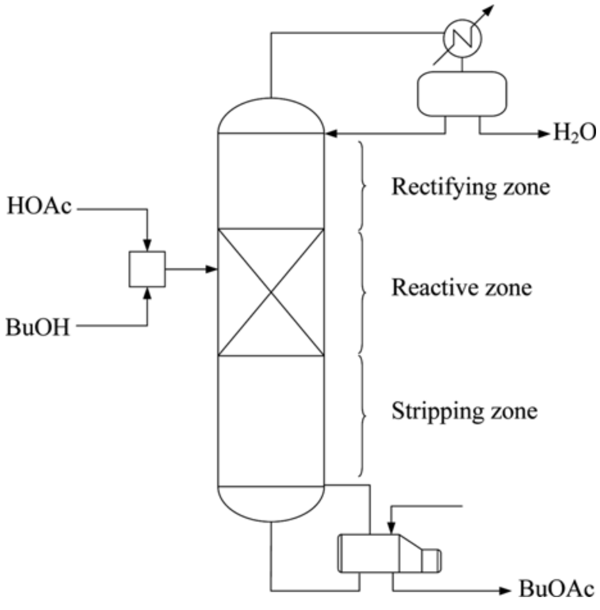

Fig. 1. Configuration of a reactive distillation column for butyl acetate process.

Table 1. Specifications of reactive distillation column

Feed conditions		
	HOAc	BuOH
Flow rate (kmol/h)	100	100
Temperature (°C)	25	25
Pressure (atm)	1.2	1.2
Column specifications		
Number of rectifying stages	11	
Numver of reactive stages	13	
Number of stripping stages	15	
Overhead pressure (atm)	1	
Column pressure drop (atm)	0.134	

Table 2. Comparison of experimental data and simulation results

	Experimental data [17]	Simulation results (HYSYS)
Bottom flow (mol/h)	42	41.9
Distillate flow (mol/h)	27	27.1
x_B (HOAc)	0.171	0.172
x_B (BuOH)	0.287	0.298
x_B (BuOAc)	0.517	0.527
x_B (H_2O)	0.025	0.003
x_D (HOAc)	0.143	0.1586
x_D (BuOH)	0	0
x_D (BuOAc)	0.008	0.0154
x_D (H_2O)	0.849	0.826
Reboiler duty (watt)	1070	950
X_{BuOH} (%)	64.58	64.31

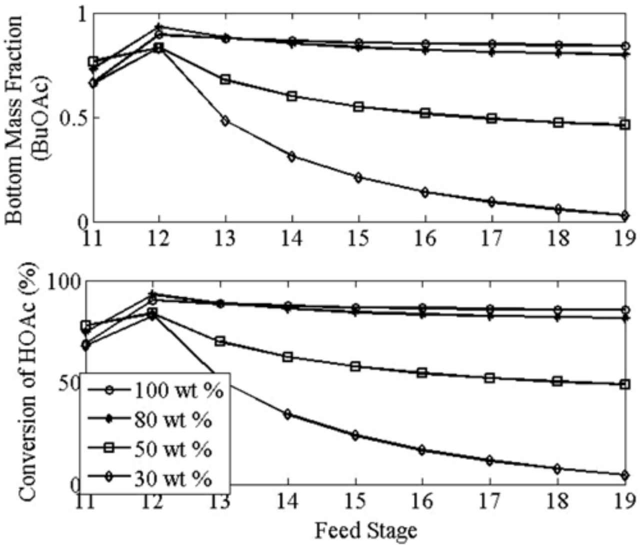

Fig. 2. Effect of feed location at different acid concentrations.

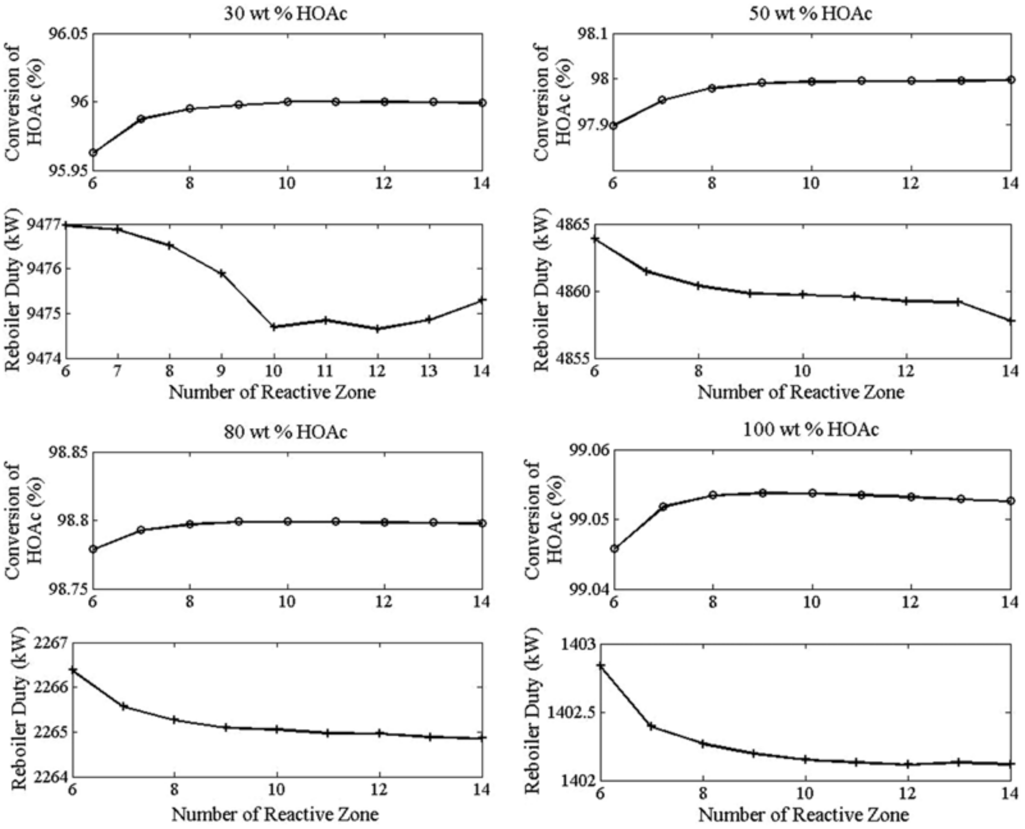

Fig. 3. Effect of the number of reactive stages at different acid concentrations.

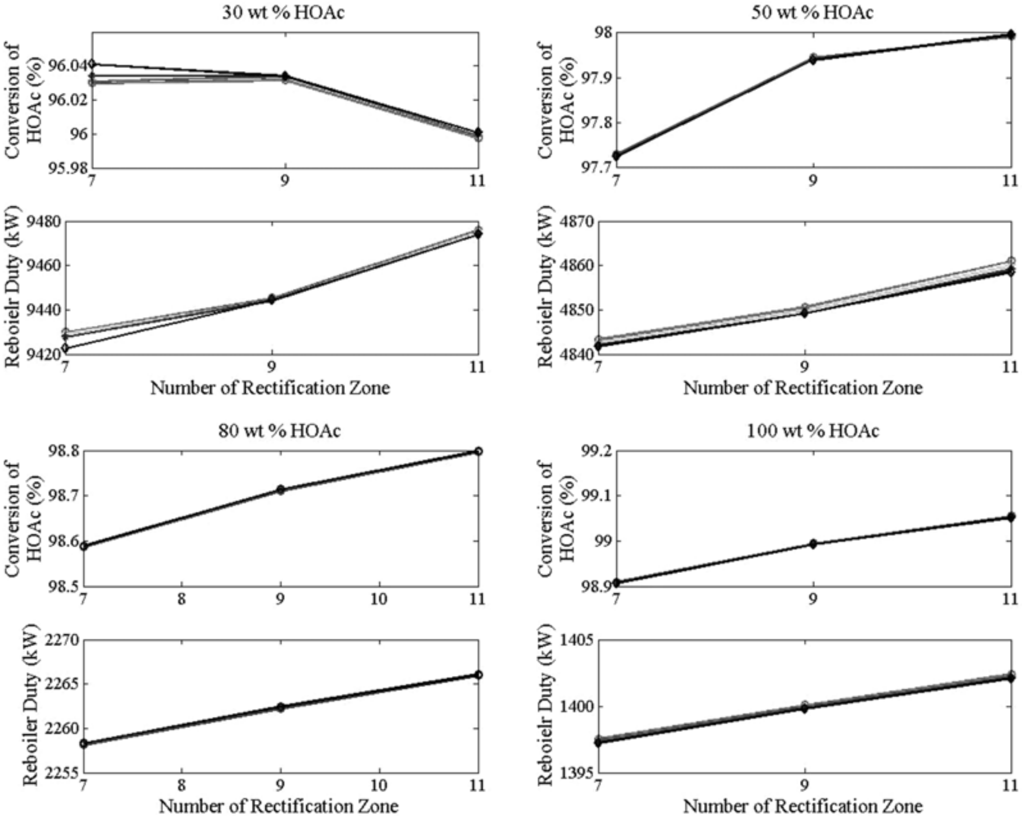

Fig. 4. Effect of the number of non-reactive stages at different acid concentrations.

tion in the presence of Amberlyst 15 as catalyst reported by Steinigeweg and Gmehling [17] are used in this study (Eqs. (3)-(5)).

$$\mathrm{HOAc}+\mathrm{BuOH}\xleftrightarrow{H^+}\mathrm{BuOAc}+\mathrm{H_2O} \tag{2}$$

$$r=k_f\left(a_{HOAc}a_{BuOH}-\frac{a_{BuOAc}a_{H_2O}}{K_{eq}}\right) \tag{3}$$

$$k_f=6.1084\times10^4\exp\left(-\frac{56.67\ \mathrm{kJ/mol}}{RT}\right) \tag{4}$$

$$K_{eq}=0.6206\exp\left(-\frac{10.99\ \mathrm{kJ/mol}}{RT}\right) \tag{5}$$

The molar feed ratio of HOAc to BuOH is kept constant at 1 : 1 following the stoichiometric ratio of the reaction. BuOH and HOAc feeds are mixed in a single stream and then introduced to the column at the same stage. The overhead vapor having a composition close to a ternary azeotropic mixture of H_2O, BuOH, and BuOAc is condensed in a condenser and then separated into two phases in a decanter [5,6]. The aqueous phase is completely withdrawn from the decanter, whereas the organic phase is totally recycled back to the column. BuOAc product is removed from the bottom of the column as it is the highest boiling point component in the system. The specifications of the reactive distillation column at standard condi-

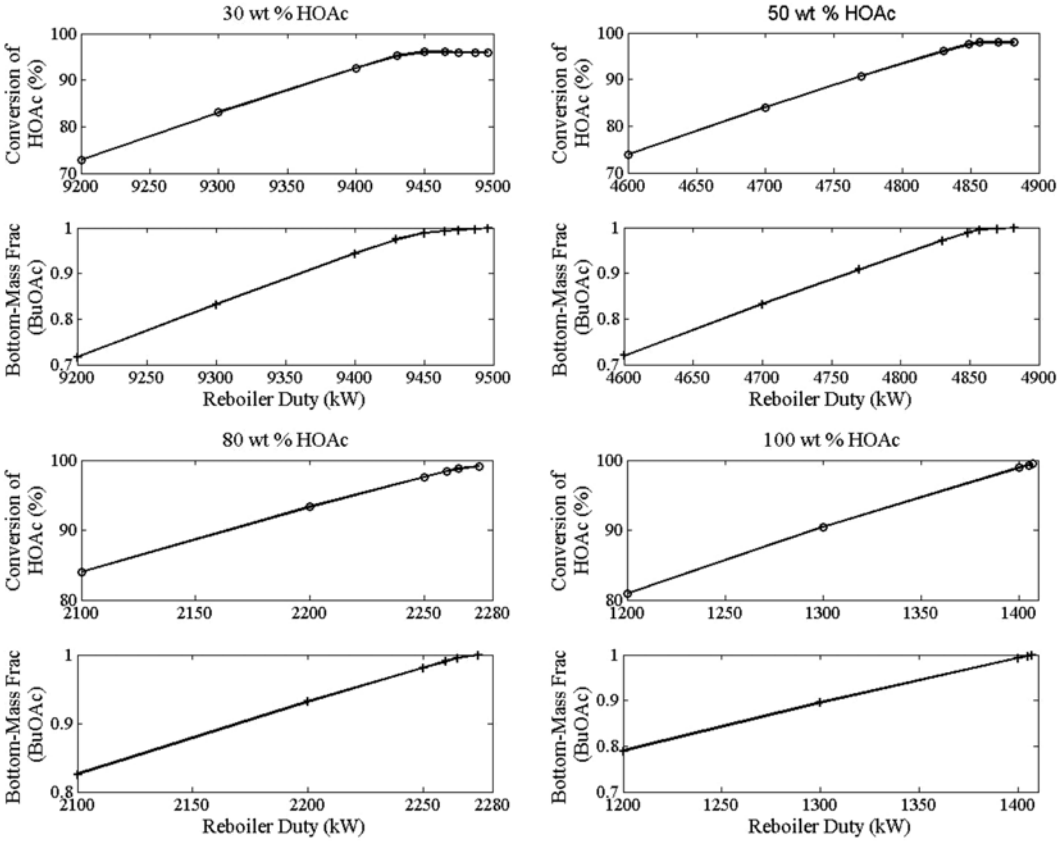

Fig. 5. Effect of reboiler duty at different acid concentrations.

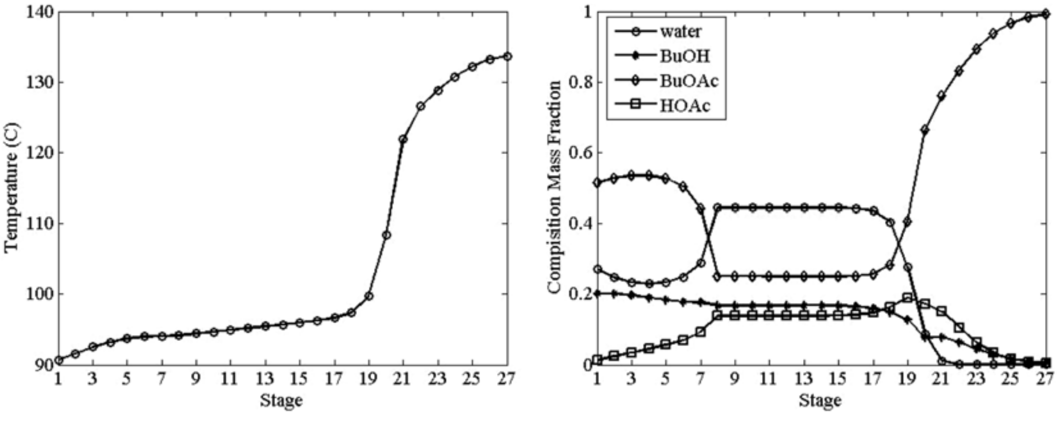

Fig. 6. Temperature and composition profiles at nominal operating conditions.

tion are given in Table 1 [14].

Note that in order to use the reactive distillation model from HYSYS for simulating a reactive distillation with confidence, the reliability of the model is verified by comparing the simulation results with the experimental data from Steinigeweg and Gmehling [17]. With the same standard experimental conditions reported in their paper, results of the simulation run in comparison to the experimental data are shown in Table 2, indicating that the simulation results are in good agreement with the experimental data.

STEADY STATE ANALYSIS

Reactive distillation columns behave substantially differently from conventional distillation columns due to the interactions between chemical reaction and vapor-liquid equilibrium. In this section, the effects of the concentration of feed acetic acid and key design and operating variables are discussed. It is noted that the specifications of the reactive distillation column as shown in Table 1 for the synthesis of BuOAc are for a preliminary configuration. The major design parameters include feed location and a number of reactive and nonreactive stages. After investigating the influence of these parameters, a suitable configuration of the reactive distillation column will be determined and used for a control study.

1. Effect of Feed Location

The feed location of HOAc and BuOH is a very important parameter in the operation of the reactive distillation column. For the system to be operated optimally, provision should be made for maximum contact area between the reactants so that the column is more used as a reactor and not as a distillation unit only. Fig. 2 shows the effect of the feed location on the HOAc conversion and mass fraction of BuOAc in the bottom stream at different concentration of feed acetic acid. In all simulations, the reboiler duty is fixed at 1,300, 2,200, 4,700 and 9,300 kW for the 100 wt%, 80 wt%, 50 wt% and 30 wt% of HOAc concentration in the feed stream, respectively. At the specified heat duty, the product butyl acetate of 90 wt% at the bottom stream is obtained and used as a basis for comparing the effect of the feed location. The results show that when the feed location is shifted down from the top stage of the reactive section, the conversion of reactants decreases, leading to a decrease in the BuOAc concentration in the bottom product stream. This result is more evident if acetic acid with lower concentration is applied. From Fig. 2, the feed location at stage 12 provides the highest conversion of HOAc and bottom product purity. It can be concluded that the most effective approach is to feed both the reactants into the column on the top of the reactive section. Therefore, all the rest of the simulations are performed by determining the feed location at this stage.

Table 3. Optimal steady-state operating conditions for the reactive distillation fed by 80 wt% HOAc

	Bottom	Distillate
Flowrate (kmol/h)	49.5113	92.1637
Composition (Mass Fraction)		
H_2O	0	0.9490
HOAc	0.0017	0.0183
BuOH	0.0031	0.0190
BuOAc	0.9953	0.0137

2. Effect of Reactive Stages

In order to increase the conversion of HOAc, the optimum num-

Table 4. Compositions and flow rate of distillate and bottom streams

Feed conditions at 80 wt% HOAc		
Feed	HOAc	BuOH
Temperature (°C)	25	25
Pressure (atm)	1.259	1.259
Feed flow (kmol/h)	91.67	50
Feed stage	8	8
Column specifications		
Number of rectifying stages	7	
Numver of reactive stages	13	
Number of stripping stages	7	
Overhead pressure (atm)	1	
Column pressure drop (atm)	0.248	

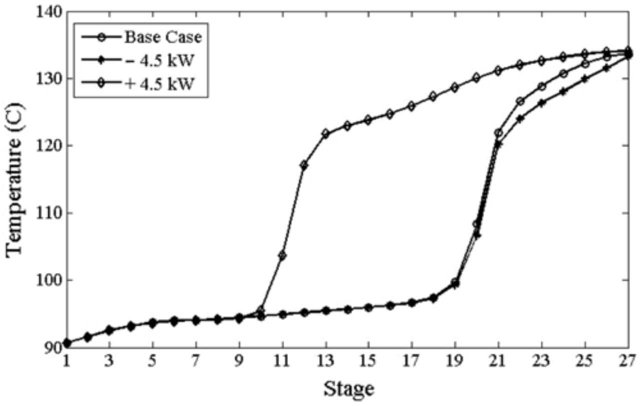

Fig. 7. Effect of reboiler heat duty on column temperature profiles.

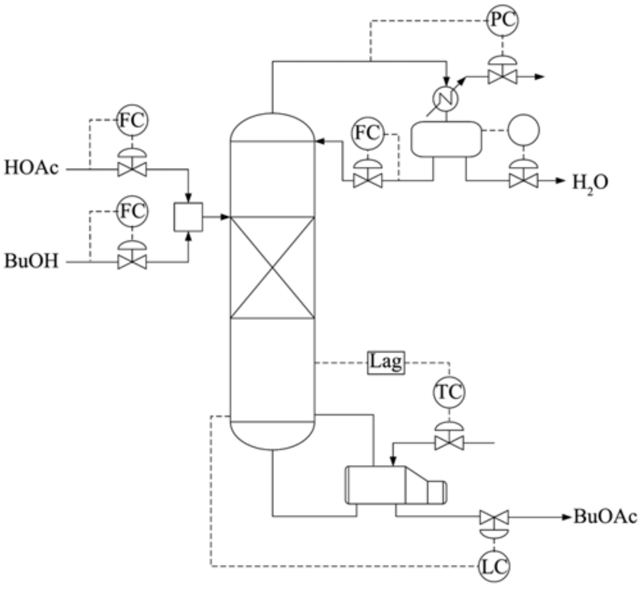

Fig. 8. Control structure 1: constant reflux flow.

ber of stages in the reactive section should be carefully provided. In all simulations presented here, the product purity of BuOAc at the bottom is at 99.5 wt%. The number of reactive stages varies from 6 to 14 stages. Fig. 3 shows the influence of the number of reactive stages on the conversion of HOAc and the required reboiler heat duty at different HOAc feed concentrations. For all cases under the conditions studied, increasing the number of reactive stages slightly increases the conversion of HOAc as more reactive stages pronounce the esterification of *n*-butanol and acetic acid. In addition, it was found that an increase in reactive stage is less sensitive to the reboiler heat duty.

3. Effect of Non-Reactive Stages

Fig. 4 shows the influence of the number of rectification stages on conversion, reboiler duty and product stream at different concentration of HOAc in feed stream. In this study, a number of stages in the reactive section are fixed at 13 and the product specification is also set to 99.5 wt% of BuOAc. For 100 wt%, 80 wt% and 50 wt% of HOAc, increasing the number of rectifying stages slightly increases both the conversion of HOAc and the reboiler duty. Since more *n*-butanol and acetic acid from the decanter are returned to the column and to the reactive section, higher conversion of HOAc is observed. It is noted that an increase in rectifying stages required more reboiler heat duty to obtain BuOAc of 99.5 wt%. However, for 30 wt% of HOAc, an increase in a number of rectifying stages results in a slight decrease of conversion of HOAc and an increase of reboiler heat duty. The results indicate that no further separation stages in the rectifying section are required because the decanter ensures a sufficient separation of water from the organic compounds because of the low solubility of BuOH and BuOAc in water. Considering the influence of the number of stripping stages (not shown

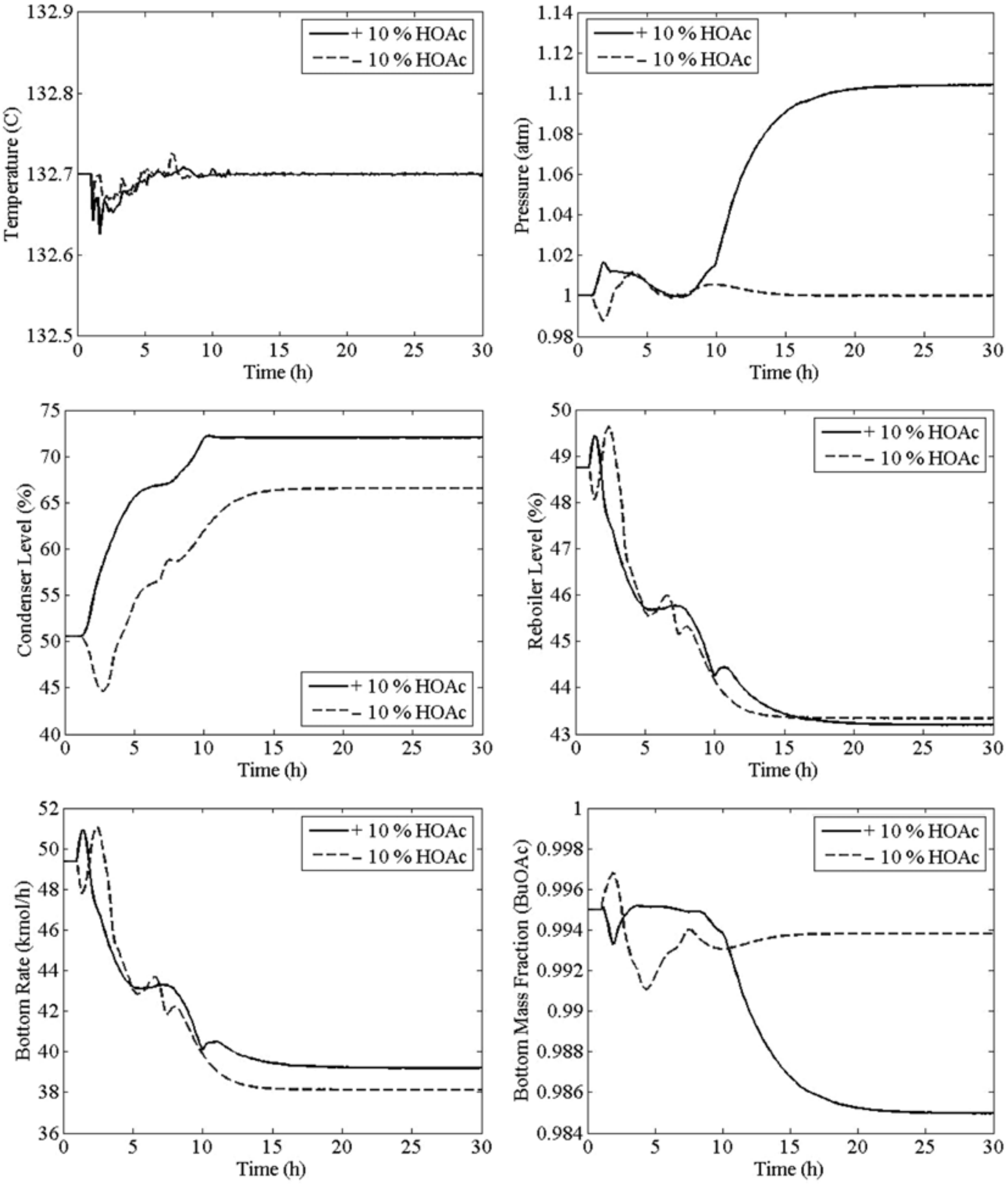

Fig. 9. Control responses of CS1 with ±10% HOAc feed rate.

in the figure), increasing the number of stripping stages has no significant effect on the conversion of HOAc and reboiler duty for all the values of feed HOAc concentration.

4. Effect of Reboiler Duty

Fig. 5 shows the conversion of HOAc and the mass fraction of BuOAc in the bottom stream as a function of reboiler heat duty at different acid concentration. Considering the case in which 30 wt% acetic acid is fed to the column, it is observed that increasing the reboiler duty increases the BuOAc purity in the bottoms. It is even possible to get higher conversion of HOAc with an increase in reboiler duty. For other cases, the conversion of HOAc and the mass fraction of BuOAc in the bottom stream show a similar trend. This is expected because an increase of the heat duty increases the temperature in the reactive section, resulting in the increased conversion of HOAc. It should be noted that as the concentration of HOAc in feed stream decreases from 100 to 30 wt%, higher reboiler duty is required due to the presence of higher amount of water in fresh feed.

5. Suitable Configuration of Reactive Distillation Column

From the above studies on the influence of HOAc concentration and key design and operating variables, the HOAc concentration in the feed stream is found to be a very important variable in the design of a reactive distillation column. It has an impact on design parameters, i.e., feed location, reboiler duty and total stages of column. As HOAc concentration is decreased from 100 wt% to 30 wt%, more energy consumption is required for the production of BuOAc with the purity of 99.5 wt%. For example, with 30 wt% and 50 wt% HOAc, reboiler heat duty of 9,400 and 4,800 kW is required. Considering the required heat duty, the use of 80 wt% HOAc as a reactant directly for the synthesis of BuOAc in a reactive

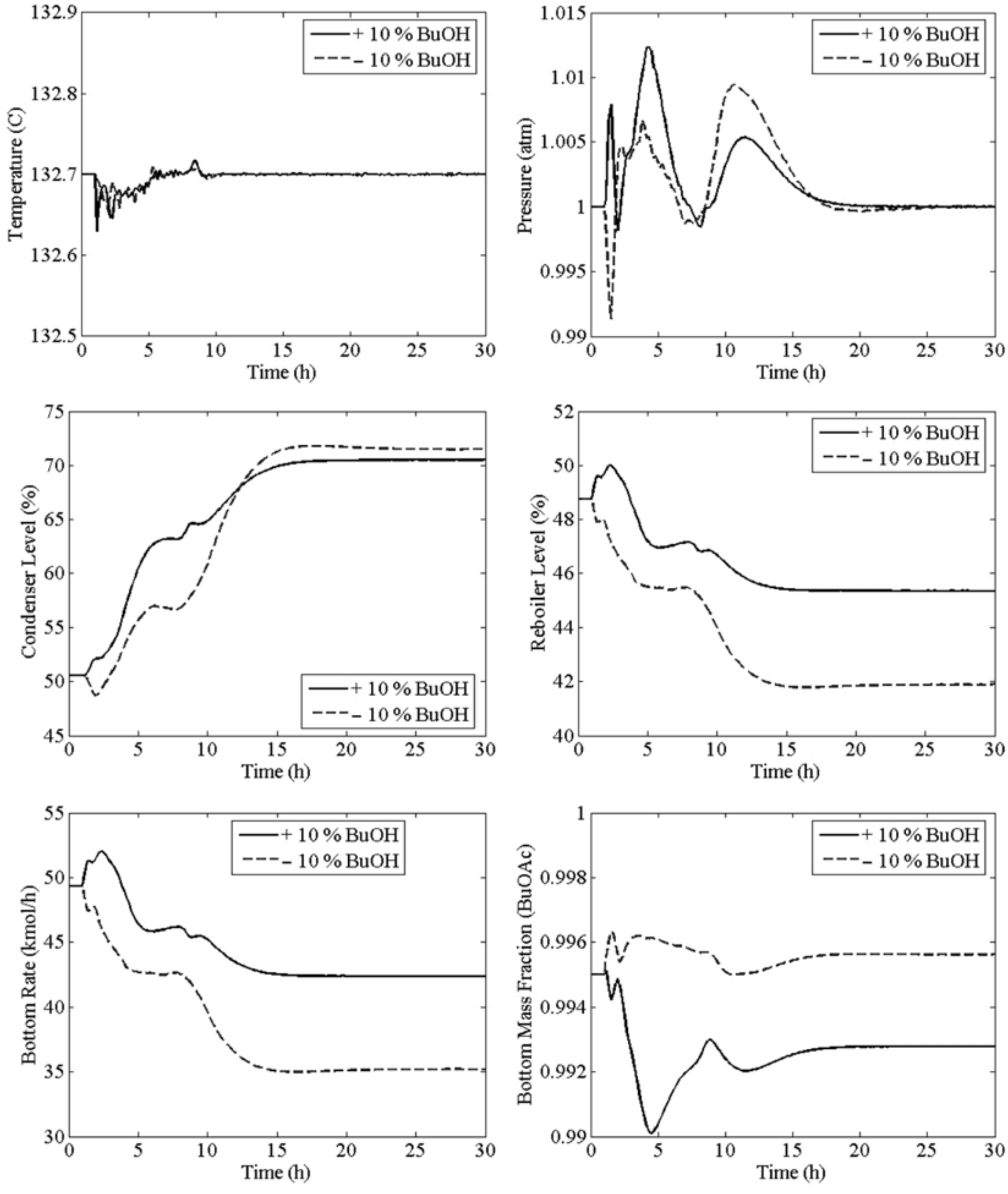

Fig. 10. Control responses of CS1 with ±10% BuOH feed rate.

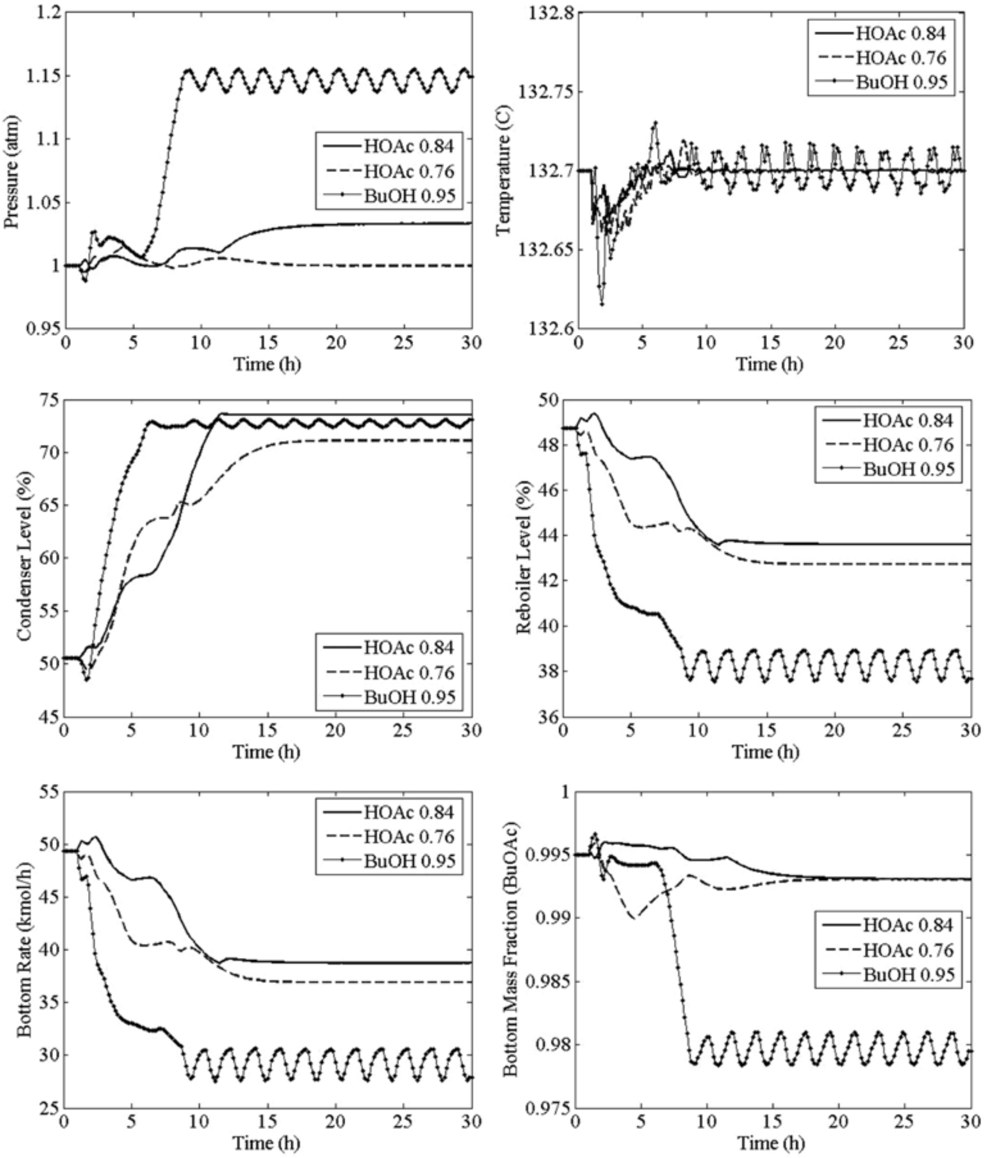

Fig. 11. Control responses of CS1 with step change in composition of feed stream.

distillation column seems to be practical. Fig. 6 shows temperature and composition profiles within the column fed by BuOH and 80 wt% HOAc at the stoichiometric ratio. At the top of the column, a heterogeneous azeotropic mixture between water, BuOH and BuOAc is formed and then separated into the aqueous and organic phases in the decanter. The results in Table 3 show that the distillate stream contains mostly water (94.9 wt%), whereas the major component of the bottom stream is BuOAc (99.5 wt%). Table 4 presents a suitable configuration of the reactive distillation column fed by 80 wt% HOAc and pure BuOH, which is used for further control study.

CONTROL OF REACTIVE DISTILLATION

Based on the optimal configuration of a reactive distillation column for butyl acetate production from dilute acetic acid (80 wt%), the control system of the reactive distillation is studied with the objective to maintain the BuOAc purity of 99.5 wt% at the bottom

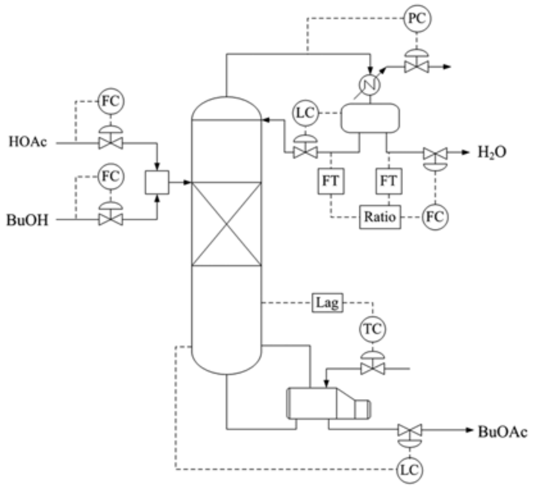

Fig. 12. Control structure 2: constant reflux ratio.

product stream. Since the aqueous phase consisting of mainly water is moved from the overhead decanter as distillation and water composition is determined by the liquid-liquid equilibrium, only the composition of BuOAc at the bottom stream is controlled.

Three alternative control structures are studied for controlling the reactive distillation column with a single mixed feed of dilute HOAc and pure BuOH introduced at the 8^{th} stage from the top of the column. The first and the second schemes are based on tray temperature control for regulating the product at a desired specification. This control strategy is known as an inferential control technique and is widely used in industries because the cost of an on-line composition analyzer and maintenance is expensive. The third control scheme uses a composition analyzer to directly control the product quality. Even though additional investment is required for the analytical equipment, a great improvement in process operation can be achieved [18]. It should be noted that all the control loops in the proposed control schemes are single-input single-output structures based on proportional-integral-derivative (PID) controllers. According to the actual applications in industries, the P controller is used in level control loop, the PI controllers are used in flow and pressure control loops and the PID controller is used in temperature control loop. The controllers are tuned based on the Tyreus-Luyben tuning method. All valves are designed to be half open at steady state. As a disturbance can cause output variables to move away from their desired set point, the control structure should account for all potential disturbances having a significant effect on the process. In this study, we assume the disturbances consist of step changes in the fresh feed flow rate (±10% of HOAc and BuOH feed flow rate) and the feed composition of HOAc and BuOH.

1. Selection of Tray Location for Temperature Control

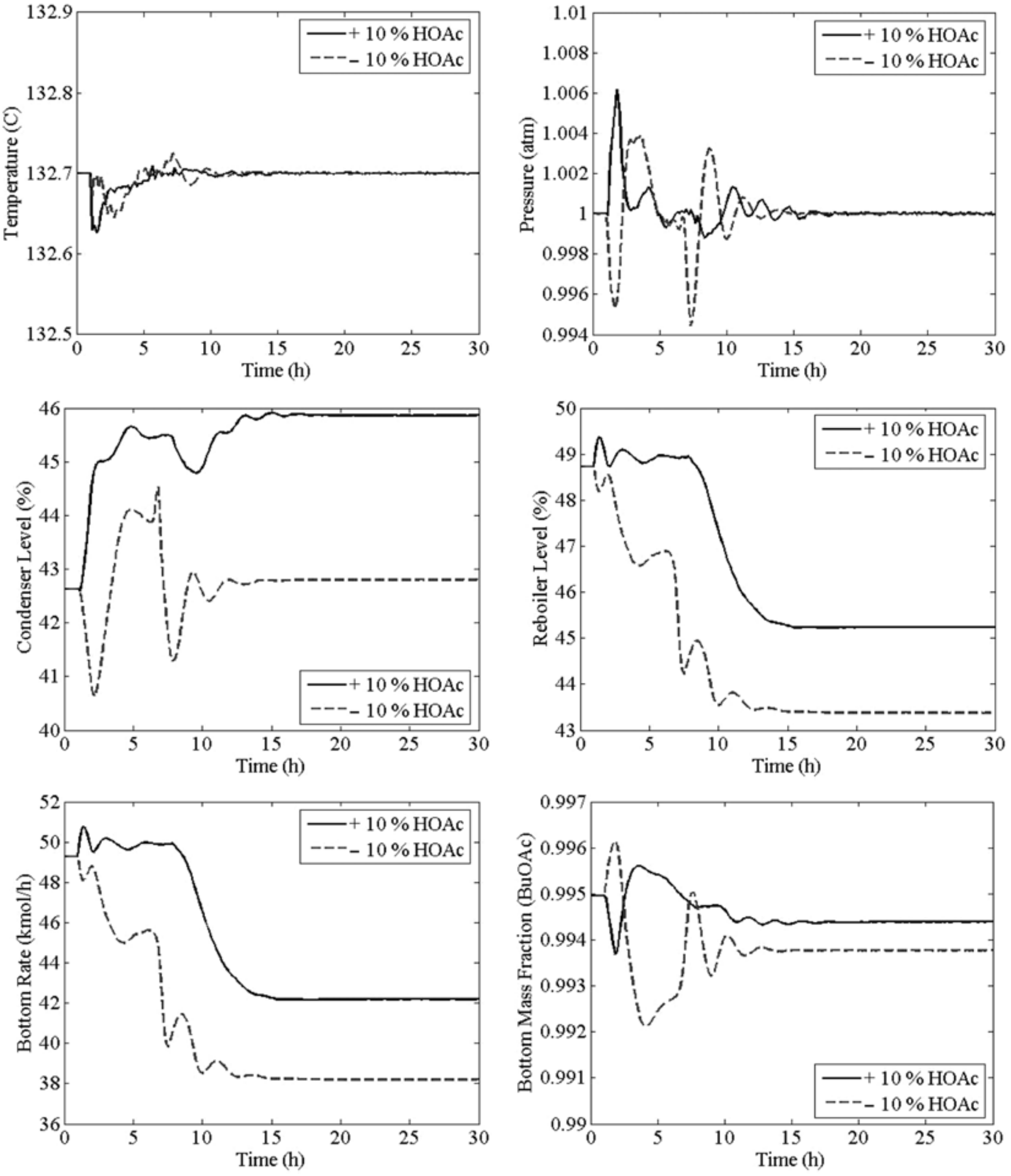

Fig. 13. Control responses of CS2 with ±10% HOAc feed rate.

Since column temperature is closely related to the product quality, the indirect control of product compositions by controlling the temperature of the distillation column is commonly found in industrial applications. A typical procedure for selecting a tray location for temperature control is applied in this work [19]. This procedure attempts to obtain high steady-state gain (relation of manipulated and controlled variables) and to avoid problems with nonlinearity by selecting the control tray where both positive and negative changes in a manipulated variable produce large and equal changes in a controlled variable. Fig. 7 shows the steady state temperature profile when reboiler heat duty as a manipulated variable is changed. Based on the criterion mentioned earlier, the temperature at the 24th stage is selected as the controlled variable of the reactive distillation.

2. Control Structure 1 (CS1)

The typical control structure of reactive distillation is shown in Fig. 8. The fresh feeds of HOAc and BuOH are flow-controlled. The column pressure is controlled by manipulating the condenser heat removal. The base level and reflux drum level are controlled by bottom product flow rate and distillate flow rate, respectively. The organic reflux flow is flow-controlled; it is kept constant at the nominal steady state condition. Temperature control of the 24th stage is implemented by manipulating the reboiler duty.

When the flow rate of HOAc is increased by 10%, it can be seen from Fig. 9 that CS1 is able to maintain temperature at the desired conditions; the controlled stage temperature can quickly be settled at its set point value. However, CS1 cannot control the column pressure; the pressure is moved from the desired value (1 atm) to a new steady state (1.105 atm). This can be explained by the limitation of the condenser in which the maximum removal of heat is limited at 3,500 kW. It can be seen that the condenser level increases to 72.01%

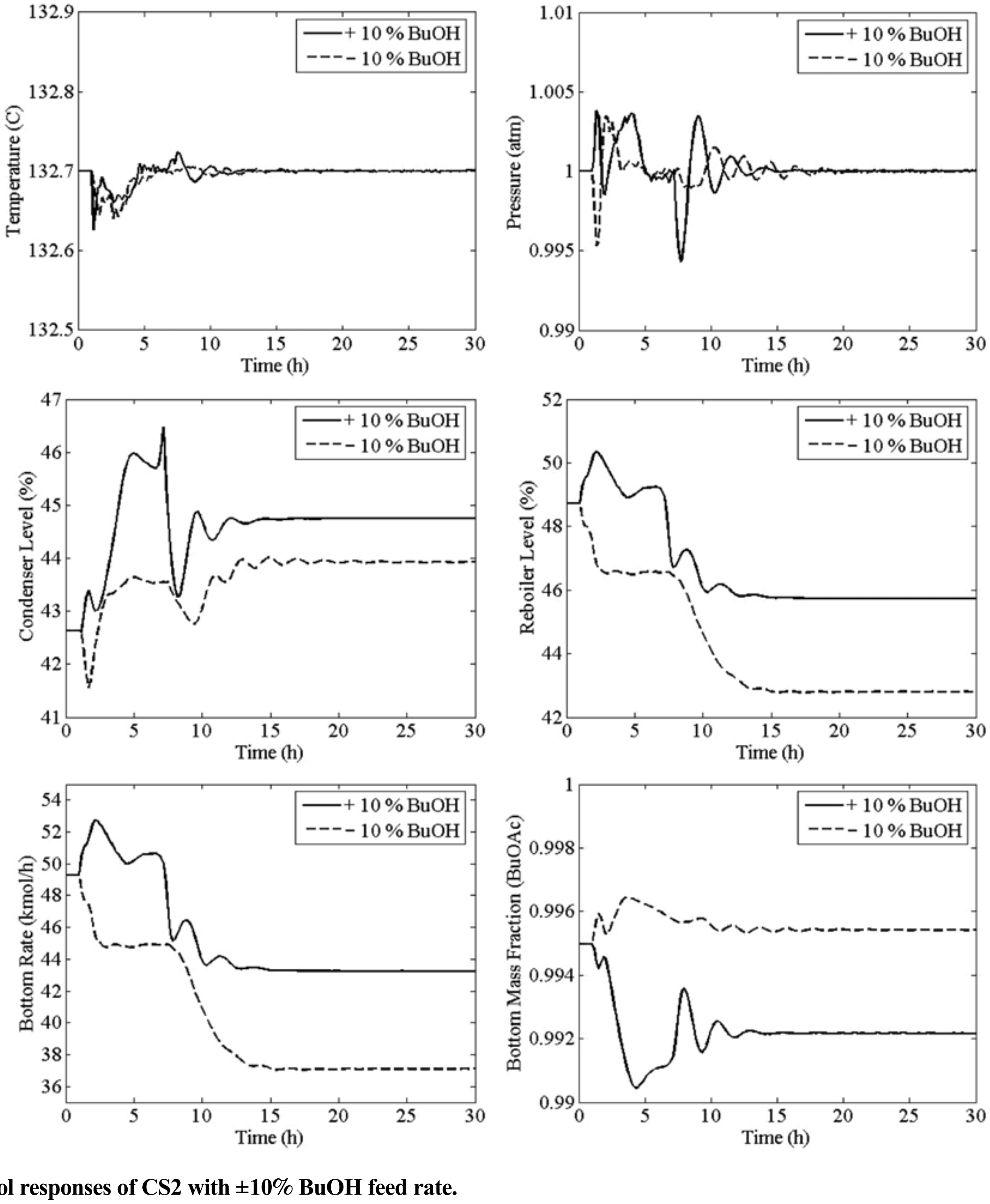

Fig. 14. Control responses of CS2 with ±10% BuOH feed rate.

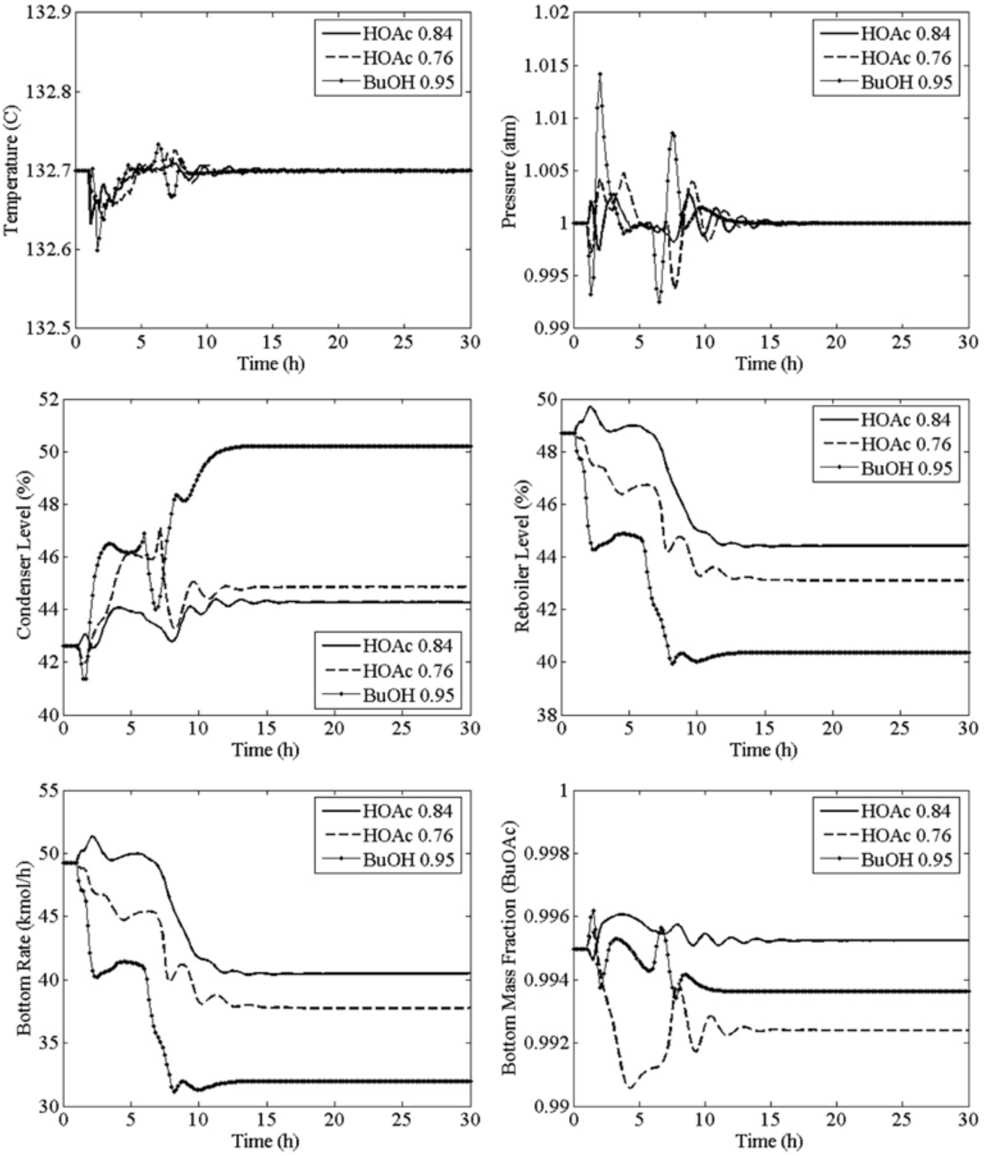

Fig. 15. Control responses of CS2 with step change in composition of feed stream.

and the purity of BuOAc at the bottom decreases to 98.48 wt%. For the case of a 10% decrease in HOAc feed rate (Fig. 9), both the temperature and pressure are controlled at the desired set point in 15 and 10 h, respectively. The BuOAc purity at bottom is 99.38 wt% at bottom rate of 38.12 kmol/h.

Fig. 10 shows the responses of the process disturbed by ±10% BuOH feed rate. It is found that CS1 can handle these changes. With 10% increase of BuOH feed flow, a product purity of BuOAc at 99.28 wt% and product rate at 42.38 kmol/h are obtained, whereas with 10% decrease of BuOH feed flow rate, the purity of BuOAc and the product flow are 99.56 wt% and 35.19 kmol/h, respectively.

Fig. 11 shows the control response of CS1 for case of the step changes in feed composition (±5 wt% of HOAc and −5 wt% BuOH in fresh feed). When the concentration of HOAc is decreased from 80 wt% to 76 wt%, the results show that a purity of BuOAc of 99.30 wt% is obtained at a flow rate of 36.69 kmol/h. In this case, both the column temperature and pressure can immediately be controlled

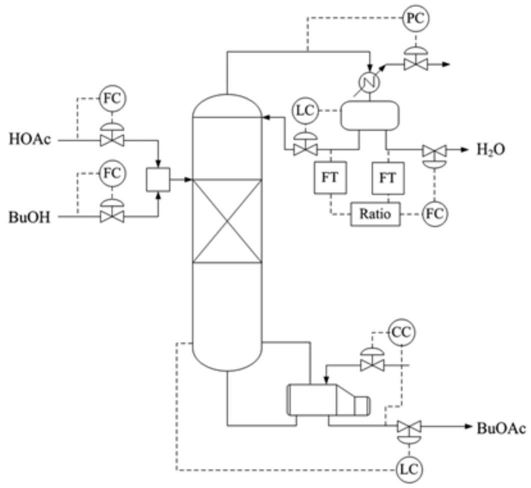

Fig. 16. Control structure 3: direct product control.

back to their set point. However, when the concentration of HOAc in the feed stream is increased from 80 wt% to 84 wt%, CS1 is not able to maintain the column pressure due to the limited capability of the condenser, whereas it can control the temperature at the set point. For a decrease in the concentration of BuOH (95 wt% BuOH in fresh feed), CS1 cannot handle this disturbance; the system becomes unstable.

From the control responses in Figs. 9 to 11, it can be concluded that CS1 cannot handle two unmeasured disturbances in the increase of HOAc feed rate and the decrease of BuOH feed composition. Also, the condenser level is observed to increase in all disturbances. Since the column temperature is controlled by reboiler duty, the increase of reboiler heat duty results in a high vapor flow rate at the top of the column, leading to increased pressure at the top of the column. To maintain the column pressure at the desired value, higher condenser heat duty is needed to condense more overhead vapors, resulting in high liquid level in the reflux drum. However, since the organic reflux flow is fixed, the increase in distillate rate is the only way to maintain condenser liquid level. As a result, this control structure cannot cope with the liquid level in the reflux drum.

3. Control Structure 2 (CS2)

Fig. 12 shows a schematic diagram of CS2. In this control structure, the fresh feeds of HOAc and BuOH are still flow-controlled and the column pressure is controlled by manipulating the heat removal of the condenser. Product stream flowrate at the bottom is employed to control the base level. As in CS1, the product composition is indirectly controlled by the control of the temperature at the 24th stage using reboiler duty as a control variable. However, the difference between CS1 and CS2 is in organic reflux flow and condenser level controlled-loops; the reflux drum level is controlled by

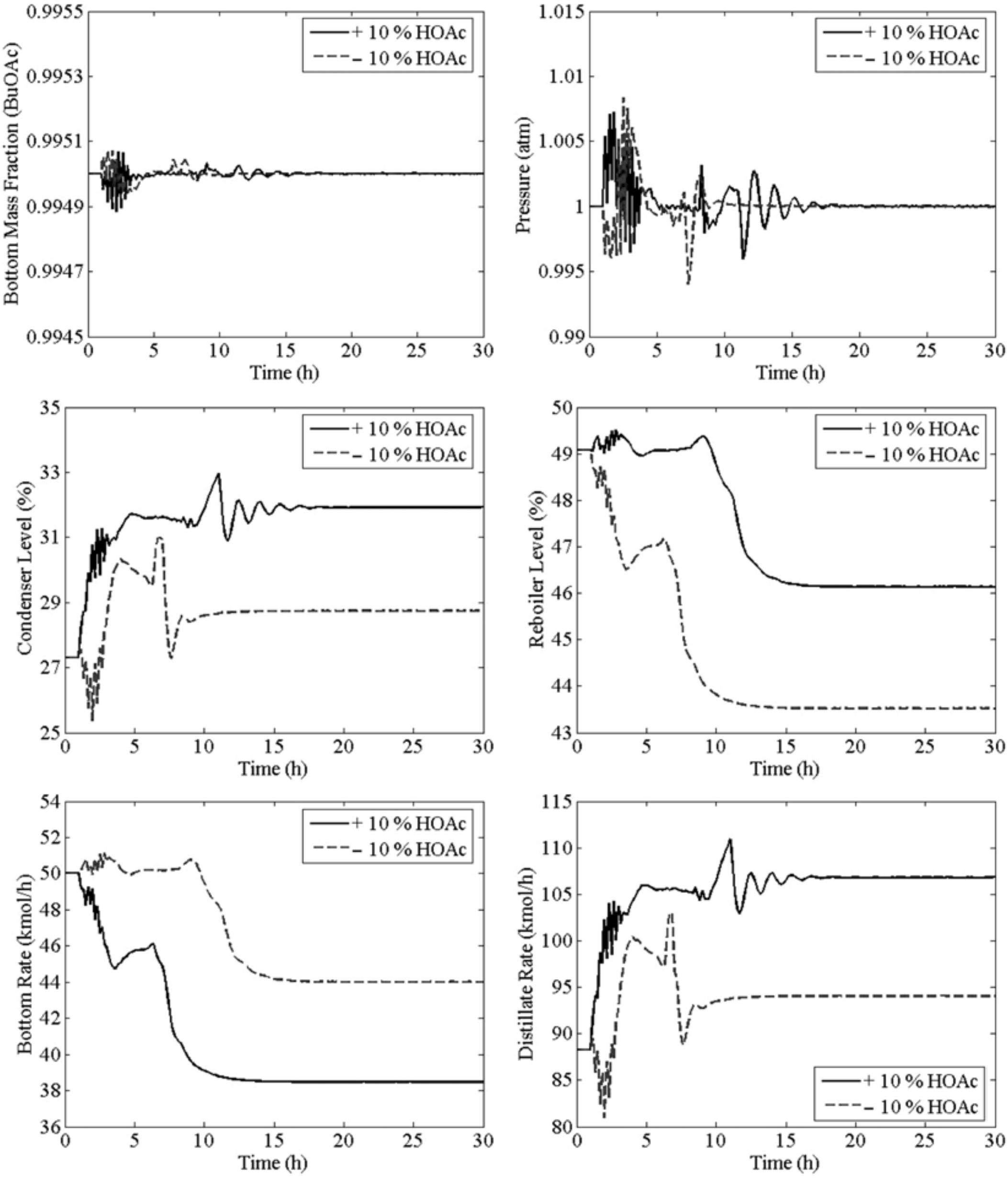

Fig. 17. Control responses of CS3 with ±10% HOAc feed rate.

manipulating the reflux flow, and a constant reflux ratio (equal to the steady state value) is maintained by adjusting the distillate flowrate. As a result, in CS2, the reflux ratio can be controlled at a desired value.

The response of the column under a change of ±10% HOAc feed rate is shown in Fig. 13. From the figure, the 24^{th} stage temperature and pressure can be controlled at the desired set point while the liquid level in reflux drum is not higher than 50%. However, the BuOAc product purity is lower than its specification (<99.5 wt%). When a step change in BuOH feed rate (±10%) is introduced, the PID controllers can manage this disturbance quite well; the temperature is controlled at the set point with shorter settling time (12 h) (Fig. 14). However, the column pressure takes a long operating time to reach the desired target (1 atm). Under a disturbance of +10% and −10% of BuOH feed flowrate, the BuOAC products obtained are 99.21 and 99.54 wt%, respectively.

The control performance of CS2 for step changes in feed composition is shown in Fig. 15. The CS2 can handle these changes. The controllers are able to maintain the temperature and pressure at the desired set point and the condenser liquid level is not higher than 51%. However, the product purity of BuOAc at the bottom stream is still lower than the desired value.

From Figs. 13 to 15, it can be seen that in CS2, since the change in the organic reflux rate is allowed, the condenser level can be maintained within the specified value (<55%). In addition, CS2 is able to control the temperature and pressure at desired set point in all case studies. Considering the product specification of BuOAc, although CS2 can accurately control the temperature of the 24^{th} stage, the product composition is changed from the design value, indicating the limitation of CS2. Therefore, CS3 is designed for the control of

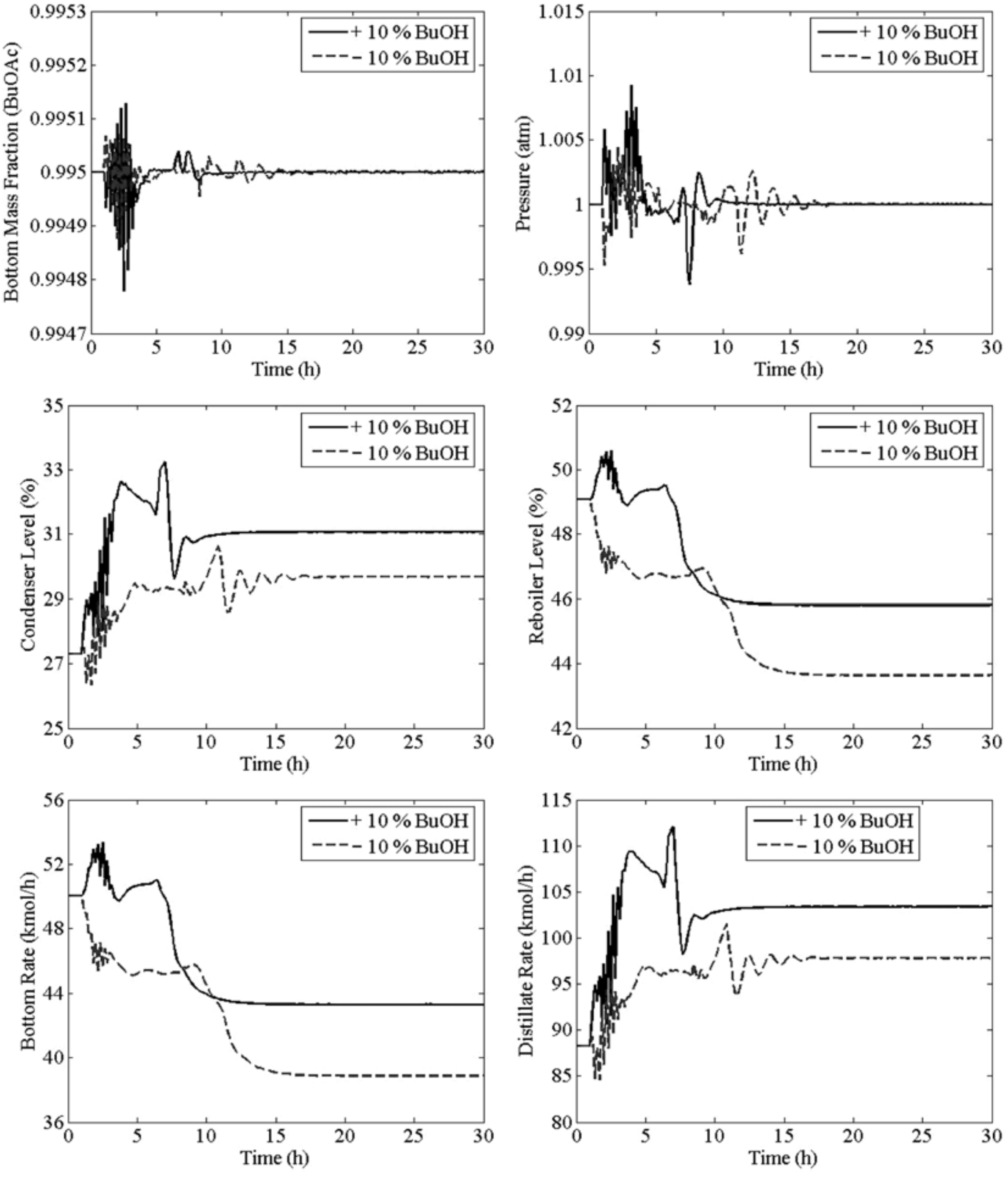

Fig. 18. Control responses of CS3 with ±10% BuOH feed rate.

reactive distillation with respect to the BuOAc purity at 99.5 wt% as a desired target.

4. Control Structure 3 (CS3)

In this control scheme, all important control loops are the same as in CS2 except the composition control loop. Instead of using an indirect composition control approach, the direct composition control is selected (Fig. 16). In CS3, the composition of the bottom product is assumed to be measurable and is controlled by manipulating the vapor boilup. It is noted that several types of instruments are available for composition measurement. Among these, a gas chromatograph is the most widely used. Although the time required for composition analysis is a major problem, the equipment has presently been enhanced and can be used for automatic control [18].

Figs. 17 and 18 show the control response when the flow rate of HOAc and BuOH is increased and decreased by 10%, whereas Fig. 19 demonstrates the response for the step changes in feed composition. It can be seen that the CS3 is able to maintain the composition and pressure at the desired set point in all simulations. However, an oscillation in the process response is observed at the early period time. The dynamic response of the composition control loops in CS3 is slower than the others because of the lag time in the bottom composition to the reboiler duty control loop.

CONCLUSIONS

This study concentrates on the direct use of dilute acetic acid for the synthesis of butyl acetate from esterification with butanol in a reactive distillation. The influence of important design parameters on the conversion of acetic acid is evaluated at different concentration of feed acetic acid. The results show that the maximum con-

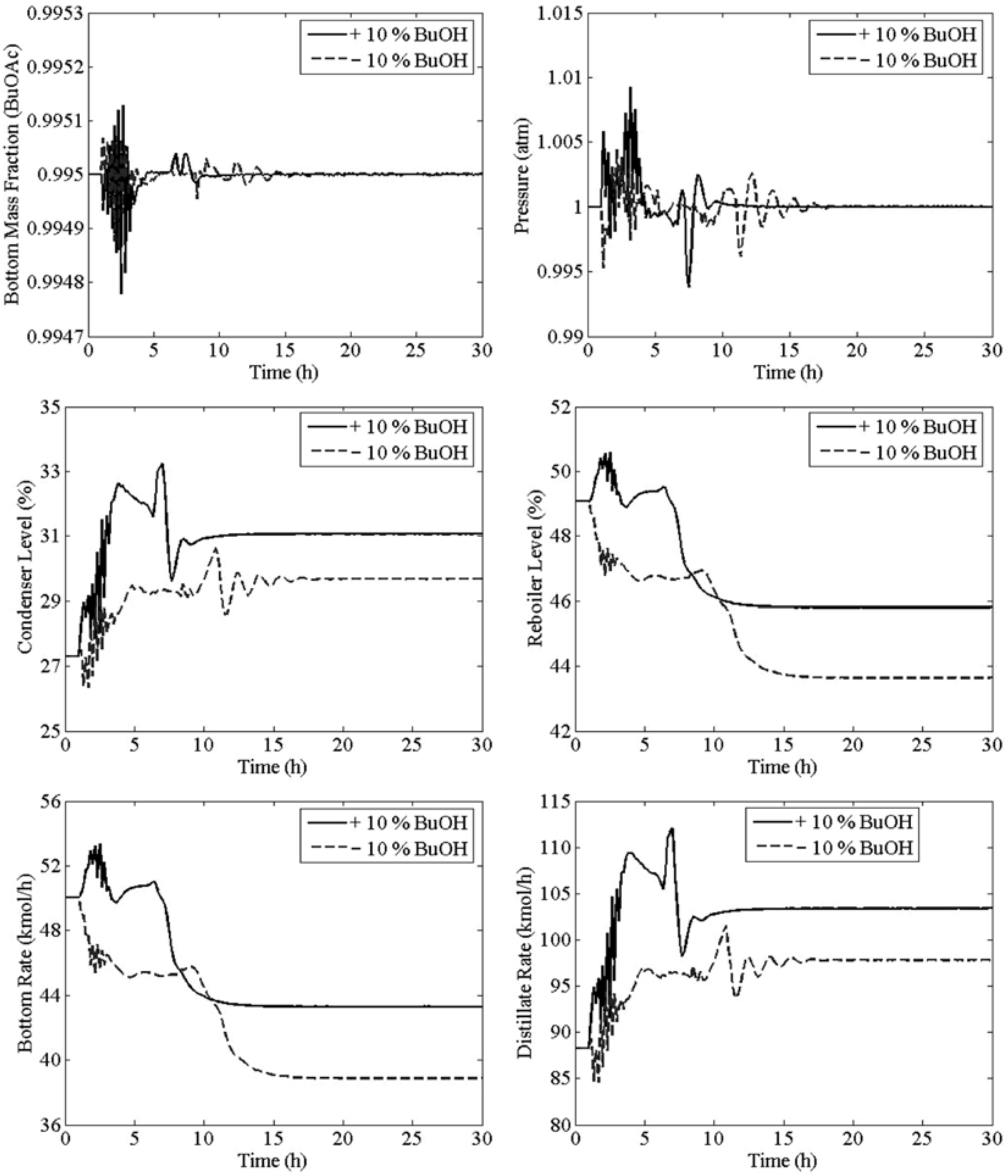

Fig. 19. Control responses of CS3 with step change in composition of feed stream.

version of acetic acid and purity of butyl acetate at bottom stream can be achieved when both acetic acid and butanol feeds are introduced to the column at the top stage of the reactive section. Changing a number of stages in the reactive section and non-reactive section has slight effect on the performance of the reactive distillation under the studied conditions. When acetic acid with lower concentration is used, a higher reboiler heat duty is required in order to obtain butyl acetate at 99.5 wt% purity. Regarding the required heat duty, the use of 80 wt% acetic acid as a reactant for the synthesis of butyl acetate in a reactive distillation column seems to be practical, and the optimal design of the column consists of 7 rectifying, 13 reactive, and 7 stripping stages.

Based on the optimum configuration of a reactive distillation column for the production of butyl acetate from dilute acetic acid (80 wt%) and the obtained steady state conditions, three alternative control structures are studied for controlling the reactive distillation column at desired steady state conditions under unmeasured disturbances in feed flow rate and concentration of acetic acid and butanol. The first two control structures (CS1 and CS2) employ the indirect control of bottom product composition with fixed reflux flow rate and reflux ratio, respectively. Tray temperature is used to infer product composition. The third control structure uses the direct control of the purity of butyl acetate based on an internal composition measurement. The results show that the use of the composition measurement is required to achieve the product with desired specification.

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NOMENCLATURE

a_i : activity of component i [-]
BuOAc : butyl acetate [-]
BuOH : butanol [-]
H_2O : water [-]
HOAc : acetic acid [-]
k_f : the forward rate constant [mol/(g s)]
K_{eq} : the reaction equilibrium constant [-]
T : temperature [K]

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