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คณะวิศวกรรมศาสตร์ จุฬาลงกรณ์มหาวิทยาลัย
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MODEL PREDICTIVE CONTROL OF A REACTIVE DISTILLATION FOR BUTYL ACETATE PRODUCTION

Mr. Teerasit Komkrajang

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By	Mr. Teerasit Komkrajang
Field of Study	Chemical Engineering
Thesis Advisor	Assistant Professor Amornchai Arpornwichanop, D.Eng.

Accepted by the Faculty of Engineering, Chulalongkorn University in Partial Fulfillment of the Requirements for the Master's Degree

……………………………………………Dean of the Faculty of Engineering
(Associate Professor Boonsom Lerdhirunwong, Dr.Ing.)

THESIS COMMITTEE

Paisan Kittisupakorn
…………………………………………….. Chairman
(Associate Professor Paisan Kittisupakorn, Ph.D.)

Amornchai Arpornwichanop
…………………………………………….. Thesis Advisor
(Assistant Professor Amornchai Arpornwichanop, D.Eng.)

…………………………………………….. External Member
(Assistant Professor Worapon Kiatkittipong, D.Eng.)

Soorathep Kheawhom
…………………………………………….. Member
(Soorathep Kheawhom, Ph.D.)

ธีรสิทธิ์ โกมกระจ่าง: การควบคุมแบบโมเดลพรีดิกทีฟของการกลั่นแบบมีปฏิกิริยาสำหรับการผลิตบิวทิลอะซิเตต (MODEL PREDICTIVE CONTROL OF A REACTIVE DISTILLATION FOR BUTYL ACETATE PRODUCTION) อ. ที่ปรึกษา: ผศ.ดร. อมรชัย อาภรณ์วิชานพ, 105 หน้า

การประยุกต์ใช้หอกลั่นแบบมีปฏิกิริยาสำหรับกระบวนการผลิตบิวทิลอะซิเตตได้รับความสนใจมากขึ้นตลอดทศวรรษที่ผ่านมาเนื่องจากเทคโนโลยีนี้สามารถลดได้ทั้งต้นทุนในการดำเนินการและต้นทุนในการผลิต อย่างไรก็ตามการควบคุมหอกลั่นแบบมีปฏิกิริยาเป็นสิ่งที่ท้าทายเนื่องจากระบบดังกล่าวมีความซับซ้อน ซึ่งเป็นผลมาจากการรวมการเกิดปฏิกิริยาและการแยกสารเข้าไว้ด้วยกันภายในหน่วย ๆ เดียว โดยปกติการใช้ตัวควบคุมแบบพีไอดีไม่สามารถจัดการกับกระบวนการที่มีความไม่เป็นเชิงเส้นสูงได้ ดังนั้นในงานวิจัยนี้จึงนำเอาการควบคุมแบบโมเดลพรีดิกทีฟเข้ามาประยุกต์ใช้กับกระบวนการผลิตบิวทิลอะซิเตต โดยในงานวิจัยนี้ได้พัฒนาแบบจำลองแบบพลวัตของหอกลั่นแบบมีปฏิกิริยาสำหรับกระบวนการผลิตบิวทิลอะซิเตตโดยอาศัยสมการดุลมวลสารและพลังงาน สมการสมดุลวัฏภาค และสมการแสดงอัตราการเกิดปฏิกิริยา ความถูกต้องของผลการจำลองกระบวนการได้ถูกเปรียบเทียบกับงานวิจัยที่เกี่ยวข้อง และผลลัพธ์ที่ได้ส่วนใหญ่สอดคล้องกันดี โครงสร้างการควบคุม 2 รูปแบบโดยใช้ตัวควบคุมแบบโมเดลพรีดิกทีฟได้ถูกเสนอและสมรรถนะของโครงสร้างการควบคุมทั้งสองได้ถูกเปรียบเทียบและอภิปรายในกรณีที่มีการรบกวนระบบและการเปลี่ยนแปลงค่าเป้าหมาย นอกจากนี้สมรรถนะของตัวควบคุมแบบโมเดลพรีดิกทีฟได้ถูกประเมินและเปรียบเทียบกับตัวควบคุมแบบพีไอ

ภาควิชา วิศวกรรมเคมี
ลายมือชื่อนิสิต ธีรสิทธิ์ โกมกระจ่าง

สาขาวิชา วิศวกรรมเคมี
ลายมือชื่ออาจารย์ที่ปรึกษา อมรชัย อาภรณ์วิชานพ

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Application of a reactive distillation for butyl acetate production has been received increasing attention over last decades because it can reduce both operating costs as well as capital investments. However, the control of reactive distillation is a challenging problem due to its complex dynamics which results from combining the reaction and separation into single unit. Conventional PID control cannot always handle its high nonlinearity. Therefore, this research considers the application of a model predictive control (MPC) to a butyl acetate reactive distillation column. A dynamic model of the reactive distillation for butyl acetate production is developed based on material and energy balances, vapor-liquid equilibrium and reaction kinetics. The accuracy of simulation results is compared with the literature and a good agreement is observed. Two alternative control structures based on a MPC controller are proposed and the performances of both the control structures for disturbance rejection and set-point tracking are investigated and discussed. Finally, the performance of MPC controller is evaluated and compared to that of a conventional PI controller.

Department ...Chemical Engineering.... Student's signature ..Teerasit Komkrajang...

Field of study Chemical Engineering..... Advisor's signature Amornchai Arpornwichanop

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NOMENCLATURE

a_i	activity of the component i in the liquid phase	[-]
B	bottom flow rate	[mol/s]
D	distillate flow rate	[mol/s]
E_A	activation energy for the equilibrium constant of esterification	[kJ/kmol]
F	feed flow rate	[mol/s]
F_r	feed ratio of acetic acid flow to butanol flow	[-]
H	molar enthalpy	[J/mol]
K_P	proportional gain in PI controller	
L	liquid flow rate	[mol/s]
m_{cat}	mass of catalyst	[kg]
P	pressure	[Pa]
P^0	vapor pressure	[Pa]
Q	heat transferred from stage to surroundings	[J/s]
Q_c	condenser duty	[J/s]
Q_r	reboiler duty	[J/s]
r	reaction rate	[mol/s]
R	reflux rate	[mol/s]
s	laplace operator	[-]
T	temperature	[K]
U	liquid molar holdup	[mol]
V	vapor flow rate	[mol/s]
x_i	mole fraction of component i in liquid phase	[-]
y_i	mole fraction of component i in vapor phase	[-]
z_i	mole fraction of component i in feed	[-]

Abbreviations

CV	controlled variable	[-]
IAE	integral absolute error	[-]
MV	manipulated variable	[-]

Greeks Symbols

γ_i	activity coefficient of component i	[-]
ρ	molar density	[mol/m^3]
τ_I	integral time in PI controller	[min]

CHAPTER I

INTRODUCTION

1.1 Introduction

Butyl acetate is widely used in chemical industries as an intermediate in organic synthesis. It is used in large quantities as a solvent for paint and coatings. Because of its lower impact on the environment, the production of butyl acetate has been growing in the last decade. Generally, butyl acetate is produced directly from acetic acid and butanol via the esterification reaction. This reversible reaction needs to be catalyzed by acidic agents such as sulphuric acid, p-toluenesulfonic acid or some solid acidic catalysts. However, using liquid acidic catalysts brings to many operation problems such as corrosion and difficulties of product-catalyst separation and disposal of waste from reaction. Therefore, a solid acidic catalyst is presently applied as it is more environmental friendly compared with common homogeneous processes.

Traditional processes for the production of butyl acetate consists of a packed-bed reactor where esterification of acetic acid and butanol is performed, connected with a train of distillation columns. Since esterification is a reversible reaction, the conversion of acetic acid in the reactor is limited by equilibrium. In addition, separation of butyl acetate as desired product from solution mixture is a difficult task, requiring the column with many stages and high energy consumption. In practice, the distillation train could be much complex if one or more azeotropes are formed in the mixture.

Recently, there are a number of research efforts concerning the use of a reactive distillation as a potential alternative for equilibrium-limited reactions. It combines both the chemical reaction and the separation into single unit operation. This technology offers several advantages over the traditional approach of reaction process followed by separation. Reactive distillation offers distinct benefits via the direct removal of the reaction products by distillation and therefore, it can increase both the reactant conversion and selectivity, leading to significantly lower investment and operating costs. Furthermore, higher energy efficiency could be achieved in case of exothermic reaction systems because the heat generated by the reactions reduces the reboiler heat input to the column.

Although reactive distillation has many advantages over conventional process, its operation and control is much more difficult. In the conventional process, the reactant conversion can be controlled separately in the reaction vessel by maintaining the reactor process variables at desirable conditions. Consequently, the product purity is controlled independently in the separation unit. On the other hand, the interaction of reaction and separation makes the reactive distillation column exhibit complex behavior such as multiplicity in steady-state solutions, high process nonlinearity and strong interactions between process variables. These complexities make the control of reactive distillation extremely difficult and may reduce the flexibility of the system. In the past years, there are many papers studied the open-loop dynamics of reactive distillation; however, only a few papers have discussed the closed-loop control of reactive distillation columns. Many researchers proposed various methods of controlling reactive distillation column using conventional proportionl-integral-derivative (PID) controller. This controller is still widely used in reactive distillation processes because of its simplicity and robustness. However, its performance is usually not sufficient for nonlinear characteristics, which is actually inherent in these chemical processes.

In this work, we focus on the control of a reactive distillation for the production of butyl acetate from esterification of acetic acid and butanol. Two alternative control structures are proposed to maintain the reactant conversion and product purity at desired condition through two-point temperature control and one-point temperature control with the direct control of reactant composition using MPC controller. The performances of both structures are investigated via the close-loop dynamic simulations in the face of various disturbances including feed flow rate change, feed composition change and set-point change. In addition, the performance of MPC for an appropriate control structure is compared with two single-loop PI control strategies under the cases of set point change and disturbance rejection.

1.2 Objective of research

The objective of this work is focused on the control of a reactive distillation for butyl acetate synthesis by using a model predictive control (MPC) algorithm.

1.3 Scopes of research

In this study, a mathematical model of a reactive distillation for butyl acetate production under an unsteady state condition is developed based on mass and energy balances. The developed model is employed for dynamic simulation of the reactive distillation and investigation of the effect of operating conditions, i.e., feed composition, reflux rate, reboiler duty on its performance. Since interaction of reaction and separation makes the reactive distillation column exhibit complex behavior, thus leading to a challenging control problem. As a result, a model predictive control (MPC) as an advanced model-based control algorithm, is proposed to control the reactive distillation. The control performance of MPC is evaluated under set point tracking and disturbance rejection conditions and compared with a conventional PID control technique.

CHAPTER II

LITERATURE REVIEWS

This chapter provides a review of the study related to the dynamic simulation and control of a reactive distillation for butyl acetate production. The previous studies of the application of reactive distillation for butyl acetate synthesis are discussed in section 2.1. Some relevant works have been done by researchers to study modeling and simulation of reactive distillation and this is summarized in section 2.2. Finally, the work of control of reactive distillation is considered in section 2.3

2.1 Reactive distillation for butyl acetate synthesis

Several attempts have been made to improve the performance of the reaction. Different strategies for the simultaneous removal of product during the course of the reaction have been received much attentions in the past. Reactive distillation has been one of such important methods to achieve this objective. Both reaction and distillation are performed in a single unit to enhance the overall performance. It not only reduces the capital cost but saves energy consumption as well.

The synthesis of butyl acetate through an esterification process has been carried out for many years. One of the earliest studies is by Leyes and Othmer (1945) and Hatig and Regner (1971), who investigated this reaction in the presence of sulfuric acid as a homogeneous catalyst. They studied the production of butyl acetate by a new method-reactive distillation but they cannot produce high-purity butyl acetate by using this method, because of lack of sufficient knowledge on the complex phase equilibrium of the system containing water, acetic acid, butanol and butyl acetate

In recent years, the solid heterogeneous catalysts are gaining attention because of their obvious engineering advantages such as ease of separation and fewer disposal and corrosion problems. Cation-exchange resin is considered to be an important heterogeneous catalyst for liquid-phase reactions like esterification, etherification, etc.

Zheng and Zeng (1997, 1998) studied the kinetics of esterification in the presence of a strong cation-exchange resin. They investigated the influence of various parameters and proposed the rate equation. Liao and co-workers (1993, 1995, 1997) also studied this reaction in the presence of solid acid cation-exchange resins.

In a broad study on heterogeneous catalysis of esterification, Kekre and Gopala (1969) investigated the influence of the mole ratio of the reactants, temperature, catalyst concentration, resin particle size and cross-linkage of the cation exchange resin. Interest in the heterogeneously catalyzed production of butyl acetate has been continuing as can be seen in Liao et al. (1993), Liao and Tong (1995), Liao and Zhange (1997), Janowsky et al. (1997), Zheng and Zeng (1997, 1998) and Steinigeweg and Gmehling (2002).

Reaction kinetics of butyl acetate synthesis is usually described by second order pseudo-homogeneous models (Leyes and Othmer, (1945), Kekre and Gopala, (1969), Steinigeweg and Gmehling, (2002), Gangadwala et al., (2003)). The side reactions of the butyl acetate formation can generally not be studied together with the main reaction, as they proceed at much lower speed, so that only very little amounts of side products are formed in typical reaction kinetic studies of the main reaction. This explains why almost no quantitative data on the side reactions of interest are found in many literatures.

Janowsky et al. (1997) mentioned the side product formation such as di-butyl ether from butyl acetate synthesis in a reactive distillation column. They could mitigate completely the side reaction by diminishing the pressure of the column, but not the formation of di-butyl ether.

Zhicai et al. (1998) described the development of a process and related equipment to produce high-purity butyl acetate from acetic acid and butanol by combining eesterification with separation by distillation. They also proposed a mathematical model to simulate the above esterfication-distillation process incorporating the relaxation method and utilizing the chemical theory to calculate fugacity coefficients for computation of phase equilibrium data. The simulation results agree well with experiments.

Hinaka et al. (1999) studied the synthesis of butyl acetate using a reactive distillation column in which solid acidic catalysts are packed inside a catalytic zone. Two sets of experiments consisting of a reactive distillation column and a primary reactor connected with the reactive distillation column were performed. ASPEN PLUS software was also used for the simulation of butyl acetate synthesis. A comparison of experimental data with the simulation results showed good agreement.

Steinigeweg and Gmehling (2002) presented a systematic approach to the development of a heterogeneously catalyzed reactive distillation process for the production of butyl acetate including reliable thermodynamic and kinetic data. In this investigation, a strongly acidic ion-exchange resin (Amberlyst-15, Rohm & Haas) was used as the catalyst. They studied the influence of various operating conditions, i.e., reboiler duty, total feed flow and reactant ratio, on the performance of a reactive distillation in the production of butyl acetate. A comparison of the experiment data with the simulation result calculated by ASPEN PLUS indicated that an equilibrium stage model is capable of describing the behavior of the reactive distillation column quantitatively.

Gangadwala et al. (2003) studied the kinetics of the etherification of butanol in the presence of several acidic ion-exchange resins including Amberlyst 15. They found that the etherification practically does not occur at low temperatures (60–90 ◦C) in conventional batch reaction experiments with a charge of acetic acid and butanol. They also performed separate kinetic experiments with pure butanol as charge at higher temperatures (113–126 ◦C) and high catalyst loading in the range 7–17 g of catalyst/gmol of butanol which allowed to develop a kinetic model of the etherification based on the Langmuir–Hinshelwood–Hougen–Watson approach. There was no data on the kinetics of the formation of isomers of butene under the conditions used for heterogeneously catalyzed reactive distillation of acitic acid with butanol. However, the formation of isomers of butene can influence the formation rate of di-butyl ether as di-butyl ether is produced not only by self-condensation of butanol but also as the product of the addition of butanol to isomers of butene.

Another work focusing on the production of butyl acetate by a reactive distillation was carried out by Gangadwala et al. (2004). An equilibrium stage model based on the intrinsic kinetics of the esterification and the unwanted side-reaction etherification over the Amberlyst-15 catalyst was developed and validated with experiment data reported in literature. With the proposed model, three different configurations of the reactive distillation column are investigated with regard to the impact of important design variables on the column performance. . It was found that the column with a reactive section and nonreactive stripping and rectifying sections is most suitable and optimum values of various design parameters such as reboiler duty, catalyst loading, catalyst section length and location, and feed tray location were determined.

Blagova et al. (2006) studied side reaction kinetics of the heterogeneously catalyzed esterification of butanol with acetic acid in and isothermal fixed bed reactor at temperature between 100 and 120 °C using three different ion-exchange catalysts. Regarding the formation of side products, Purolite CT 269 and Amberlyst 48 give similar results: side reactions proceed to significant extent. For Amberlyst 46, however, side reactions were found to be almost negligible. They concluded that the esterification occurs mainly on or near the external surface of catalysts particles whereas side reactions occur mainly in the pores. Their work showed that surface-sulfonated catalysts like Amberlyst 46 are very attractive for the production of esters by reactive distillation.

2.2 Modeling and simulation of reactive distillation

Two fundamentally different approaches to simulating distillation systems have been developed: the equilibrium stage model and the non-equilibrium transfer rate model (Kister, 1992). Theoretically, both of these methods can be applied to reactive distillation with appropriate modifications and additions to reflect the reaction.

The steady state simulation of reactive distillation has been tackled frequently since the 1970 s. Simandl and Svrcek (1991) provided an excellent review of the work in this area and alluded to the range of numerical methods that can be applied to solve the system of equations which describe the reactive column. Their survey indicated that only the equilibrium stage model had been used to that point.

Grosser et al (1987) proposed a simplified unsteady state model of reactive distillation for a nylon 6,6 column but their assumption of constant molar overflow in the vapor phase is considered unrealistic for most systems. More recently, several rigorous dynamic models have been proposed for simulating transient events and investigating process dynamics of reactive distillation.

Abufares and Douglas (1995) proposed a model based on the equilibrium stage approach that included expressions to account for tray hydraulics and reaction kinetics. They successfully demonstrated the validity of this approach by comparing simulation results to previously published experimental results from a laboratory scale column.

Ruiz et al. (1995) , Schrans et al.(1996) and Pilavachi et al.(1997) have subsequently produced dynamic models with a similar structure and have shown that these models can be use effectively for reactive MTBE columns although limited data has been provided for model validation

Alejski and Duprat (1996) provided a comprehensive review on reactive distillation simulation and also proposed a rigorous dynamic model of their own which they subsequently applied to a reactive ethyl acetate column. The most important contribution of this work was to compare dynamic models of varying complexity to determine whether various simplifying assumptions were valid or invalid. They concluded that it was necessary to model the hold-up on each stage (e.g. using the Francis weir formula) where the hold-ups are large, but simpler models are acceptable where the hold-ups are smaller and the dynamics are much faster.

Sneesby et al. (1997) simulated an ETBE reactive distillation column using Pro/II4 and SpeedUp. Using this steady-state simulation model, they studied the effects of design and operating variables on column performance and developed a design method for ETBE reactive distillation column.

Vora and Daoutidis (2001) studied the dynamics of a reactive distillation column for the production of ethyl acetate. They proposed a configuration that involves feeding the two reactants in different trays and allows for the attainment of higher conversion and purity at the steady state than does the conventional configuration, which involves feeding in a single tray. An analysis of the column dynamics was then performed, which reveals the existence of fast and slow dynamics corresponding to the liquid flow rates and the liquid compositions and holdups, respectively. Further analysis of the tray hydraulics of the column showed that this time-scale multiplicity is induced by the relatively instantaneous approach of the volume of liquid on a tray to the weir volume.

Jhon and Lee (2002) developed an algorithm for dynamic simulation for reactive distillation with ETBE synthesis. A structured and simple iterative algorithm was devised for the calculation related to the rigorous energy balance. This algorithm was implanted in the conventional Luyben algorithm, in order to achieve the fast convergence and the robustness during the simulation. The characteristics of the RD were investigated for the total condenser and the partial condenser. The reactive distillation performances for these condensers were observed and compared with each other. They studied the open-loop dynamic responses of the system for total and partial condenser configurations. They concluded that the partial condenser operated as one additional separation stage. It provided a similar conversion and a little higher purity than total condenser.

Olanrewaju and Al-Arfaj (2005) presented a comprehensive formulation of a linearized state space process model for a generic two-reactant-two-product reactive distillation system. The development of the model required the knowledge of the desired steady state design data, including liquid holdups and composition profiles. The linearity of the model is attested by the uniform and symmetric nature of the output responses to different magnitudes of step inputs. The application of a linear state estimator in a model-based control of a reactive distillation using the linearized process model was explored. It is demonstrated that a robust linear estimator can be successfully implemented in a feedback system of a linear reactive distillation provided that a reliable approximate process model could be obtained.

2.3 Control of Reactive Distillation Column

Most of the literature available on a reactive distillation is based on steady-state conditions including process design and the analysis of multiple steady states. Dynamic modeling and simulation have also been studied, but a relatively small amount of work has been reported on the control of reactive distillation columns. Reactive distillation is a challenge for control due to process nonlinearity and complex interactions between vapor–liquid equilibrium and chemical reactions.

Kumar and Daoutidis (1999) studied the control of a reactive distillation column in which ethylene oxide and water react to form ethylene glycol. A consecutive reaction to form diethylene glycol also occurs. Fresh ethylene oxide and fresh water are fed into the column, and the product is removed from the base. There is no distillate product. The variables they chose to control are pressure, base level, and the purity of the bottoms product (the concentration of ethylene glycol).They studied an input/ output linearizing controller that was claimed to be effective at moderate product purities but was unstable at higher purities.

Vora and Daoutidis (1999) extended this work to the ethyl acetate reactive distillation case. Although there were both distillate and bottoms products in this column, they only controlled the distillate purity. They again used a nonlinear input/output linearizing state feedback controller, which was tested by making set point changes in the distillate composition controller. The two fresh feed streams were apparently flow controlled, so the proposed structure suffers from the stoichiometric imbalance problem.

Sneeby et al. (1997) studied the dynamic behavior and control aspects of the ETBE column and tested several control configurations, and among them LV and LB configurations were found to be more effective for single-composition control. In another study, Sneesby et al.(1999) proposed a two-point control scheme for an ethyl tert-butyl ether (ETBE) reactive distillation column in which both bottoms product purity and conversion are controlled. Conventional PI controllers are used to control a temperature in the stripping section by manipulating the reboiler heat input and to control conversion by manipulating the reflux flow rate. Conversion is calculated inferentially from several temperature and flow measurements.

Al-Arfaj and Luyben (2000) studied an ideal, generic two-product reactive distillation column with simple thermodynamic and kinetic behavior. They proposed a variety of control structures for two-point control of product purity in which the concentrations were controlled with direct measurement of compositions in the system. A single-point control structure with the stripping section stage temperature used as an inferred variable to control the bottom product purity was also investigated. In another study, Al-Arfaj and Luyben (2002) applied the control structures developed in the previous work to an optimized double-feed ETBE reactive distillation column. They also proposed a single point control structure controlling either the bottom product composition or a stripping section stage temperature for a single-feed ETBE reactive distillation.

Gruner et al. (2003) developed a general controller for reactive distillation systems based on asymptotically exact input/output linearization and applied their controller to a simulation of an industrial reactive distillation column with improved performance compared to a well-tuned linear controller.

Tian et al. (2003) proposed a pattern-based predictive control (PPC) scheme for single-point control of the bottom product ETBE purity using the reboiler duty as the manipulated variable. They introduced a nonlinear transformation to obtain a pseudolinear input-output relationship and incorporated the PPC with conventional PI control.

Wang, Wong and Lee (2003) investigated the control strategy of a reactive distillation for butyl acetate synthesizes. The main purpose was to investigate whether a simple linear control strategy could be used to maintain high product purity and reactant conversion for a reactive column that was in the kinetic control regime. They showed that, for such columns, the desired steady state temperature and composition profiles change when there was a change in the production rate or when the catalyst deactivates. They were able to locate points in the temperature and composition profiles that are relatively insensitive to throughput rate changes and catalyst deactivation. Reboiler duty is used as the manipulated variable in the temperature control loop. The feed rates of butanol and acetic acid were controlled, with the set point of the acetic acid flow loop ratioed to that of the butanol flow loop. The proposed scheme was able to reject various disturbances and maintain the desired high-purity and high-conversion operation.

Kaymak and Luyben (2005) studied an ideal reaction system and the methyl acetate system. The purpose of their work was to compare two different types of two-temperature control structures for reactive distillation columns. The effectiveness and robustness of these control structures were compared in the face of disturbances in the production rate and fresh feed compositions. One of the main conclusions was that the selection of the manipulated fresh feed stream in the control structure had an important role in the stability of the system. Sequential tuning of the interacting temperature controllers was sometimes necessary.

Zeng, Kuo, Chien (2005) studied the design and control of a reactive distillation column system for the production of butyl acrylate. The proposed design was quite simple including only one reactive distillation column and an overhead decanter. The optimal design was selected based on the minimization of total annual cost for the overall system. The highest purity stable steady state was selected as the base case condition for the control study. The overall control of this system could be achieved with no on-line composition measurements. Simple single-point tray temperature control loop was designed to infer final product purity. From results of dynamic simulation, the proposed control strategy performed very well in rejecting various disturbances while maintaining butyl acrylate product at high purity. One of the important finding in this paper was that it is better to operate this reactive distillation column not at the exact feed stoichiometric balance point for better operability reason.

Khaledi and Young (2005) proposed a model predictive control (MPC) scheme for two-point control (purity and conversion) of an ETBE reactive distillation column. They applied PI control and linear model predictive control to a dynamic simulation of an ETBE reactive distillation column using the control configuration. The linear model predictive controller used first-order plus dead time models for each of its input/output models. The model predictive controller was found to be very efficient for disturbance rejection and setpoint tracking, but it was slightly slower than the PI controller. However, the model predictive controller was able to handle the process interactions perfectly.

Kawathekar and Riggs (2006) proposed the application of nonlinear model predictive control (NLMPC) to a highly nonlinear reactive distillation column. NLMPC was applied as a nonlinear programming problem using orthogonal collocation on finite elements to approximate the ordinary differential equations that constitute the model equations for the reactive distillation column. Diagonal PI controls were used to identify that the control configurations performed best. NLMPC was found to provide significantly better control performance than PI controls, but this work quantifies the advantage as a factor of 2–3 reduction in variability. The advantage of the NLMPC controller came from faster closed-loop dynamic performance compared to the PI controller resulting from using a nonlinear dynamic model of the reactive distillation column considered here. In addition, NLMPC was found not to be particularly sensitive to process/model mismatch. Even though the reactive distillation column considered in this study was shown to be extremely nonlinear, the PI control was able to control the process reasonably well.

CHAPTER III

THEORY

3.1 Butyl acetate synthesis

3.1.1 Reaction kinetics

Butyl acetate is commonly produced by the esterification of acetic acid and butanol as shown in Eq.(3-1). This reversible reaction needs to be catalyzed by strong acids. Sulfuric acid, p-toluenesulfonic acid, or some solid acidic catalysts are usually used for this purpose.

$$\underset{\text{(Acetic acid)}}{CH_3COOH} + \underset{\text{(Butanol)}}{C_4H_9OH} \Leftrightarrow \underset{\text{(Butyl acetate)}}{CH_3COOC_4H_9} + \underset{\text{(Water)}}{H_2O} \tag{3-1}$$

Kinetic data for butyl acetate synthesis was reported by Steinigeweg et al. (2002). They used pseudohomogeneous and adsorption-based model to describe esterification reaction that is catalyzed by ion-exchange resins (Amberlyst 15) as shown in Eq.(3-2) using UNIQUAC model to estimate the activity coefficients.

$$r = \frac{1}{m_{cat}}\frac{1}{\nu_i}\frac{dn_i}{dt} = k_1 a_{acetic\,acid} a_{butanol} - k_{-1} a_{butyl\,acetate} a_{water} \tag{3-2}$$

where r is the reaction rate, k_1 is the forward reaction rate constant, k_{-1} is the backward reaction rate constant, and a_i is the liquid phase activity of i component.

The temperature dependence of the rate constant is expressed by Arrhenius' law:

$$k_i = k_i^0 \exp\left(\frac{-E_{A,i}}{RT}\right) \tag{3-3}$$

where k_i^0 and $E_{A,i}$ represent the preexponential factor and activation energy of forward reaction rate constant, respectively, and R and T are the gas constant and temperature, respectively. The values of kinetic parameters are given in Table 3-1.

Table 3-1 Kinetic parameters for the pseudohomogeneous kinetic model

Reaction	i	k_i^0 (mol g^{-1} s^{-1})	$E_{A,i}$ (kJ mol^{-1})
Esterification	1	6.1084×10^4	56.67
Hydrolysis	-1	9.8420×10^4	67.66

3.1.2 Thermodynamic properties

The components involved with the esterification of acid acid and butanol have distinctly different thermodynamic properties and form very non-ideal mixture. The thermodynamic properties estimation methods are shown in Table 3-2. The vapor equation coefficients, the heat capacity coefficients and the heat of formation are obtained from Chemical Properties Handbook (Yaws, C.L., 1999).

The Raoult law was applied to find the phase equilibrium composition of the gas phase, $y_{i,j}$,using Antoine expression to obtain vapor pressures of pure components, P^0. As the sum of the gas fractions on each stage must be equal to one, the Raoult equation and the consistency equation can be combined into the following single algebraic equation:

$$1-\frac{1}{P_j}\sum_{i=1}^{Nc}\gamma_{i,j}x_{i,j}P_{i,j}^0 = 0 \tag{3-4}$$

The activity coefficients γ are necessary for the liquid phase nonidealities to be taken into account .In this work, the UNIQUAC equation is employed for the calculation of the activity coefficient (γ) values to be used not only for the satisfactory description of the reaction kinetics but also for the determination of phase and chemical equilibriums (see Appendix A for the UNIQUAC parameters involving butyl acetate system).

Table 3-2 Thermodynamic property estimation

Thermodynamic property	Estimation method
Vapor pressure	Antoine equation
Activity coefficients	UNIQUAC method
Phase equilibrium composition	Raoult law

3.2 Reactive distillation

The relatively large amount of new interest in reactive distillation is due to the numerous advantages it has over ordinary distillation. Enhanced reaction rates, increased conversion, enhanced reaction selectivity, heat integration benefits and reduced operating costs are just to name a few. All these factors contribute to the growing commercial importance of reactive distillation. However, since heat transfer, mass transfer, and reactions are all occurring simultaneously, the dynamics which can be exhibited by catalytic distillation columns can be considerably more complex than found in regular columns. This results in an increase in the complexity of process operations and the control structure installed to regulate the process

.

The principle of building a reactive distillation column is quite simple. A distillation column having a catalyst zone strategically placed in the column to carry out the desired reaction is a reactive distillation column. The catalyst may be in the same phase as that of the reacting species or it can be in the solid phase. The feed for the process is fed either above or below the reactive zone depending upon the volatility of the components and to carry out the desired reaction. The reaction occurs mainly in the liquid phase, in the catalyst zone.

A simple example considering a reversible reaction can explain some of the mentioned properties. Consider a reaction $A + B \leftrightarrow C + D$, where the boiling points of the components follow the order A, C, D and B.

The traditional flowsheet can be a series of distillation columns as shown in Figure 3-1 (a) where the mixture of A and B is fed to the reactor, where the reaction takes place in the presence of a catalyst. The mixture of reactant and product is passed through a series of distillation columns and the unreacted components A and B are recycled back to the reactor. The alternative reactive distillation approach consists of a reactive section in the middle of the column. The task of the rectifying section is to recover reactant B from the product stream C. In the stripping section the reactant A is stripped from the product stream D. The relative volatility between the components has been used more effectively in the later case.

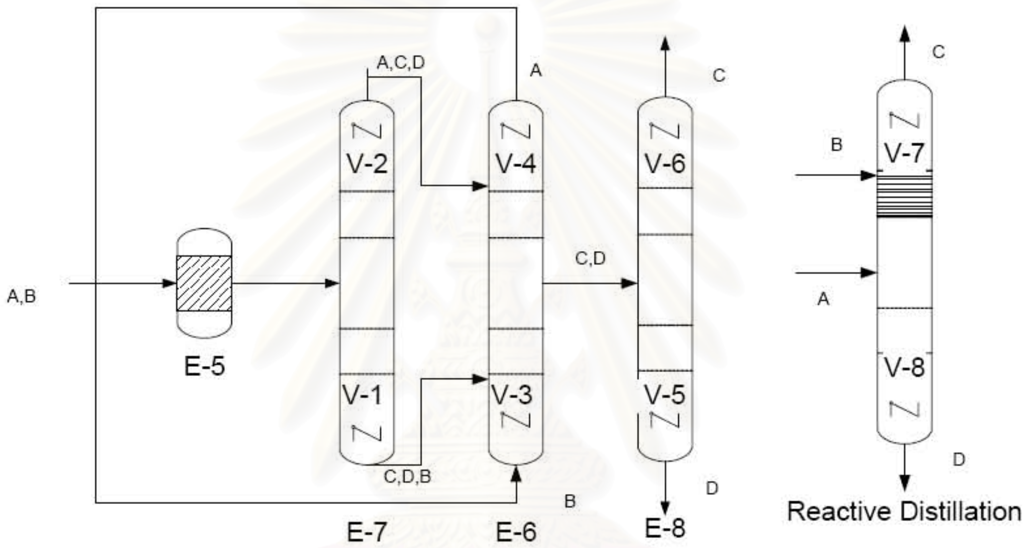


Figure 3-1 Comparison of conventional scheme and reactive distillation for a generic system

3.3 Model Predictive control

3.3.1 Introduction

Model predictive controller (MPC) is traced back to the 1970s. It started to emerge industrially in the 1980s as IDCOM (Richalet et. al.) and DMC (Cutler and Ramaker). The initial IDCOM and MPC algorithms represented the first generation of MPC technology. Generally, MPC is a family of controllers in which there is a direct use of an explicit identifiable model. It is also described as a class of computer control schemes that utilizes a process model for two central tasks:

- Explicit prediction of future plant behavior
- Computation of appropriate corrective control action required to drive the predicted output as close as possible to the desired target value.

Control design methods based on MPC concept have found wide acceptance in industrial applications and have been studied by academia. It is currently the most widely used of all advanced control methodologies in industrial applications. The reason for such popularity is the ability of MPC design to yield high performance control systems capable of operating without expert intervention for long periods of time.

3.3.2 Motivation

The typical goals of process control are:

- Disturbance rejection to decrease variability in the key variable
- Improve the operation of a process, the productivity of the plant, the quality of the product.
- Stable and safe operation.

While achieving the above tasks, consideration should be continuously given to the following issues:

- *dynamic and unpredictable marketplace conditions*: It is generally accepted that the most effective way to generate the most profit out of the plant while responding to marketplace variation with minimal capital investment is provided by the integration of all aspects of automation.
- *safety and environmental regulations*: Some process variables must not violate specified bounds for reasons of personnel or equipment safety or because of environmental regulation. Safety measures and environmental regulations are continuously changing.

To develop better, fast, accurate and robust process control, model-based modern control algorithms and efficient adaptive and learning techniques are required.

The requirement for consistent attainment of high product quality, more efficient use of energy, and an increasing awareness of environmental responsibilities have all combined to impose far restrict demand on control systems than can be met by traditional techniques alone. The industrial response to these challenges led to the development of the successful MPC algorithm. It had an enormous impact on industrial process control and served to define the industrial MPC paradigm.

3.3.3 Benefits of MPC

- Most widely used control algorithm in material and chemical processing industries.
- Increased consistency of discharge quality. Reduced off-specs products during grade changeover.
- Minimizing the operating cost while meeting constraints (optimization, economic).
- Superior for processes with large number of manipulated and controlled variables (Multivariable, strong coupling).
- Allows constraints to be imposed on both MV and CV.
- Cope with processes with time delays, inverse response, inherent nonlinearities (difficult dynamics)

3.3.4 General concept

Block diagram for the MPC implementation is shown in Figure 3-2. As shown in the figure, a process model is used in parallel to the plant. MPC uses a dynamic model of the process in order to predict the controlled variable. The predicted controlled variable is fed back to the controller where it is used in an on-line optimization procedure, which minimizes an appropriate cost function to determine the manipulated variable. The controller output is implemented in real time and then the procedure is repeated every sampling time with actual process data. The difference between the plant measurement, y_p and the model output y_m is also fed to the controller to eliminate steady state offset. Usually the cost function depends on the quadratic error between the future reference variable and the future controlled variable within limited time horizon.

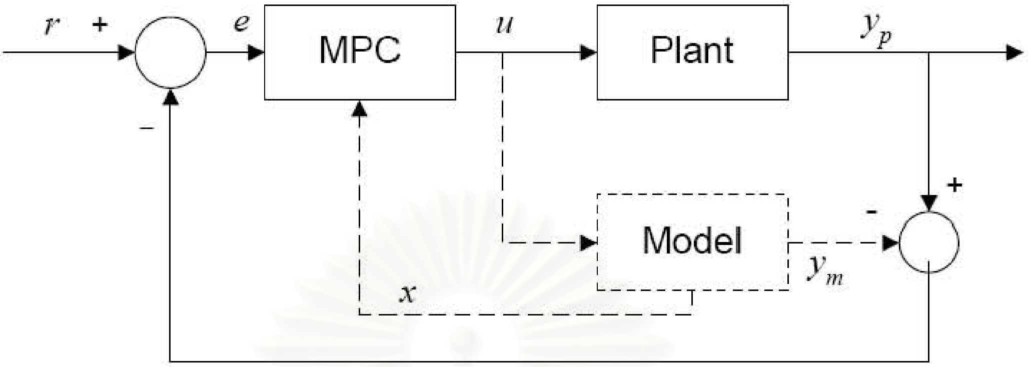


Figure 3-2 Block diagram for MPC implementation

3.3.5 The receding horizon concept

MPC is a digital controller, i.e. a discrete time technique. The control calculation is computed via a digital computer and the result is implemented online each sampling time. As mentioned earlier this procedure is repeated in a moving horizon approach. The concept of moving horizon can be understood from Figure 3-3. Assume we are at certain sampling time k. The past trend for the output (y) up to k and input (u) up to k-1 are known. The objective is then to find the future trend for the input (control actions) that moves the future trend of the output approaches the desired reference trajectory $r(k+1)$. The control actions are found through iteration. In fact, an optimization problem is solved to compute online and in real-time the open loop sequence of present and future control moves [$u(k/k)$, $u(k+1/k)$ … $u(k+M-1/k)$], such that the predicted outputs [$y(k+1/k)$ $y(k+2/k)$ … $y(k+P/k)$] follow the predefined trajectory. The optimization is solved taking into consideration constraints on the outputs and inputs. The first control action $u(k/k)$ is then picked and implemented on the real plant over the interval [k,k+1]. In the method, M is known as the control horizon and P as the prediction horizon.

At the next sampling time k+1, the prediction and control horizon are shifted ahead by one step and a new optimization problem is solved using updated measurements from the process. Thus, by repeatedly solve an open-loop optimization problem with every initial conditions updated at each time step, the model predictive control strategy results in a closed-loop constrained optimal control technique.

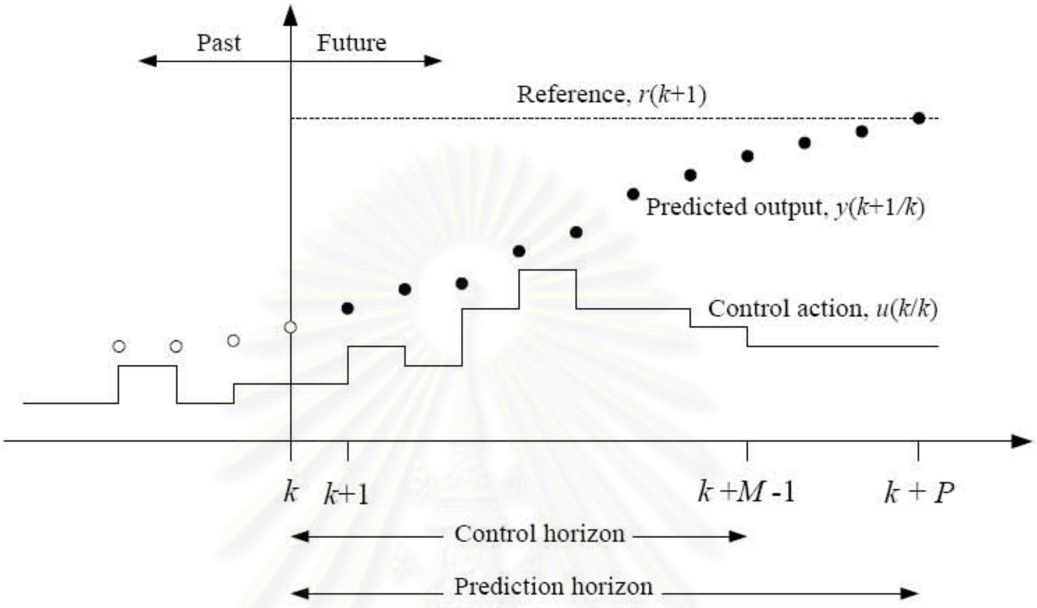


Figure 3-3 Receding horizon control scheme

3.3.6 MPC based on State-Space Models

A state space based model predictive control is described in this section. The controller design is based on a model of the open loop process.

$$\begin{aligned} x(k+1) &= Ax(k) + B_u u(k) + B_d d(k) + w(k) \\ z(k) &= Cx(k) \\ y(k) &= Cx(k) + v(k) \end{aligned} \tag{3-5}$$

where $y(k)$ and $z(k)$ are vectors with measured and noise free process variables, $u(k)$ is the vector with manipulated outputs, and $d(k)$ is the vector with measurable disturbances. The noise vector $w(k)$ and $v(k)$ are assumed to be white noise sequences. It is assumed that the model (3-5) is stabilizable and detectable.

Integrators are introduced by using an extended state space model that uses the differentiated state vector $\Delta x(k) = x(k) - x(k-1)$ and the controlled output $z(k)$ of (3-5). This gives

$$\begin{bmatrix} \Delta x(k+1) \\ z(k+1) \end{bmatrix} = \begin{bmatrix} A & 0 \\ CA & I \end{bmatrix} \begin{bmatrix} \Delta x(k) \\ z(k) \end{bmatrix} + \begin{bmatrix} B_u \\ CB_u \end{bmatrix} \Delta u(k) + \begin{bmatrix} B_d \\ CB_d \end{bmatrix} \Delta d(k) + \begin{bmatrix} I \\ C \end{bmatrix} \Delta w(k)$$

$$z(k) = \begin{bmatrix} 0 & I \end{bmatrix} \begin{bmatrix} \Delta x(k) \\ z(k) \end{bmatrix}$$

$$y(k) = z(k) + v(k)$$

which in short notation can be written as

$$\begin{aligned} \hat{x}(k+1) &= \widetilde{A}\hat{x}(k) + \widetilde{B}_u \Delta u(k) + \widetilde{B}_d \Delta d(k) + \Delta \hat{w}(k) \\ z(k) &= \widetilde{C}\hat{x}(k) \\ y(k) &= z(k) + v(k) \end{aligned} \tag{3-6}$$

The state vector is estimated using a state observer. It is based on the model (3-5). The observer is given by

$$\begin{aligned} \varepsilon(k) &= y(k) - \widetilde{C}\hat{x}(k \mid k-1) \\ \hat{x}(k+1 \mid k) &= \widetilde{A}\hat{x}(k \mid k-1) + \widetilde{B}_u \Delta u(k) + \widetilde{B}_d \Delta d(k) + K\varepsilon(k) \end{aligned} \tag{3-7}$$

The observer (3-7) provides the one step ahead prediction of the extended state vector. Further predictions are obtained by repeated use of (3-6) with the assumption that $\Delta u(k)=0,\ k>M$ (control horizon) , $\Delta d(k) = 0,\ k>1$, and $\varepsilon(k) = 0,\ k>1$. Multiplication with C provides prediction of z, based on estimated state, actual measurements, and future manipulated output moves. The output vector is predicted P samples ahead (prediction horizon) and control actions are considered for M future samples, $M \le P$ (control horizon). To simplify the notation, introduce

$$U(k)=\begin{bmatrix} u(k) \\ : \\ u(k+M-1) \end{bmatrix} \quad , \quad Z(k)=\begin{bmatrix} z(k) \\ : \\ z(k+P-1) \end{bmatrix} \tag{3-8}$$

that collects manipulated outputs over the control horizon and process variables over the prediction horizon. Then the predicted process variables over the prediction horizon are

$$Z(k+1|k)=\begin{bmatrix} \tilde{C}\tilde{A} \\ : \\ \tilde{C}\tilde{A}^{P} \end{bmatrix}\hat{x}(k|k-1)+\begin{bmatrix} \tilde{C}\tilde{B}_u & \ldots & 0 \\ : & & 0 \\ \tilde{C}\tilde{A}^{(P-1)}\tilde{B}_u & \ldots & \tilde{C}\tilde{A}^{(P-M)}\tilde{B}_u \end{bmatrix}\Delta U(k)+\begin{bmatrix} \tilde{C}\tilde{B}_d \\ : \\ \tilde{C}\tilde{A}^{(P-1)}\tilde{B}_d \end{bmatrix}\Delta d(k)+\begin{bmatrix} \tilde{C}K \\ : \\ \tilde{C}\tilde{A}^{(P-1)}K \end{bmatrix}\varepsilon(k)$$

which in short notation can be written as

$$Z(k+1|k)=S^{X}\hat{x}(k|k-1)+S^{U}\Delta U(k)+S^{d}\Delta d(k)+S^{\varepsilon}\varepsilon(k) \tag{3-9}$$

The presence $\varepsilon(k)$ in (3-9) shows that the feedback is based on the most recent measurement of $y(k)$. The control error over the prediction horizon is the difference between predictions and the trajectory of future set-point, i.e.

$$E(k+1)=Z(k+1|k)-R_f(k+1) \tag{3-10}$$

The three degrees of freedom design is obtained by splitting the error function in three different parts,

$$E(k+1|k)=E_{sp}(k+1|k)+E_{ff}(k+1|k)+E_{fb}(k+1|k) \tag{3-11}$$

each with its own state vector definition. The first part is the set-point error, the second part is the feed forward error, and the third part is the remaining error. Three consecutive optimization problems are then solved for the three error functions to provide $\Delta U_{sp}(k)$, $\Delta U_{ff}(k)$, and $\Delta U_{fb}(k)$. These sum up to $\Delta U(k)$, the increments of the manipulated outputs.

Each optimization problem is of the form:

$$\min_{\Delta U(k/k)} [E(k+1)]^T \Gamma [E(k+1)] + [\Delta U(k)^T \Lambda \Delta U(k)] \tag{3-12}$$

with respect to constraints on predicted process variables and to constraints on manipulated outputs.

3.3.7 Tuning

Basically, the MPC has four main tuning parameters namely; the control horizon M, the prediction horizon P, the output weight matrix Γ and the input weight matrix Λ. The last two matrices are diagonal ones. The sampling time has a strong impact on the control performance, however it is not used as a tuning parameter since it is often fixed based on the equipment at installation. Therefore, the total number of tuning parameters is $2+nu+ny$. These parameters have profound and somewhat overlapping effect on the closed-loop performance. The definition and function of each parameter is outlined next:

The control horizon, M is the number of MV moves that MPC computes at each sampling time to eliminate the current prediction error. A large M has the advantage that it allows detection of constraints violation before they are reached, averages the control objective over time, and handle unknown variable time delays.

The prediction horizon, P, represents the number of samples into the future over which MPC computes the predicted process variable profile and minimizes the prediction error. Usually $P>M$ is selected to avoid dead-band effect. Increasing P, result in a significant non-monotonic response ranging from damped to under-damped to damped.

The move suppression factor (weight on the MV move, Λ). They indicate the trade-off between the amount of movement allowed in the manipulated variables and the rate at which the output deviation from set point is reduced over the prediction horizon. They serve dual purpose of suppressing aggressive control action and conditioning the system matrix before inversion.

The weighting matrix Γ, is used primarily for scaling in the multivariable case; it permits the assignment of more or less weight for the objective of reducing the predicted error for the individual output variables. When a single γ is increased, the set point tracking response for the corresponding process variable has a faster rise time.

According to the understanding of the function of the MPC parameters, general guidelines are available (Moreshdi and Garcia, 1986; Garcia and Morari, 1982; Ohno et. al. 1988; Meadows and Rawlings, 1997). However, due to high nonlinearity of the process and/or the presence of modeling errors, MPC are commonly tuned by trail-and-error procedure.

In general, based on general reasoning, one can simplify the tuning problem to a single primary tuning parameter, which is Λ. Note that P can not be made independent of the sampling time. In addition, for stability reasons, P must be selected such that it includes the steady state effect of all past MV moves, i.e., it should equal the open-loop settling time (n) of the process in samples. Note also that the relative value of M to P is more important, therefore, M can be fixed at a small number to reduce computational effort. On the other hand, Γ do not affect the invertibility of the overall system matrix. Thus, they can be specified by the user on the basis of control objective priorities or saved for later fine-tuning.

3.4 Control of Reactive distillation

Control of reactive distillation columns refers to the ability of keeping certain variables at or near their set points whenever there is a disturbance or set point change in the plant. The procedure for determining which process variables should be controlled by manipulating certain values is called is control strategy design. Dynamic simulations can be used to provide a picture of how the plant it will behave when there is a set point change and disturbances.

Controller system design can be broken into the following steps.

- Formulate control objective
- Identify controlled and manipulated variables

- Choose a control strategy and structure
- Specify controller settings

The control objective can generally be formulated based on safety concerns, environmental regulations, and economic objectives. Of the steps involved in designing a control strategy for a distillation column identifying controlled and manipulated variables is one of the first steps. The controlled and the manipulated variables are paired based upon the sensitivity between them. The controller is implemented between these controlled and manipulated variables to get the closed loop plant. The selection of variables depends on the control strategy to be used.

3.4.1 Degrees of freedom analysis

A simple two-product distillation column with a single feed and a total condenser has five degrees of freedom. These correspond to control valves that vary the following quantities:

a) The distillate product draw rate (D),
b) The bottoms product draw rate (B),
c) The reboiler duty (Q_R or V for the internal vapor rate),
d) The reflux rate (L for the internal liquid rate),
e) The condenser duty (Q_C)

The condenser and reboiler duties usually cannot be manipulated directly but the designation, Q_R and Q_C, are used to represent the group of variables which could used to manipulate the duty in each case. For example, the control valve which is designated to regulate the condenser duty might actually adjust the coolant flow rate, the active surface area of the condenser or the rate at with vapor is withdrawn from the column. Similarly, the method of regulating the reboiler duty could be the heating medium flow rate, the reboiler exchanger area or the process flow through the reboiler.

The column pressure, the column base level and the reflux accumulator level (i.e. the column vapor and liquid inventory) must all be stabilized for the column to operate in a steady-state. The column pressure is almost always controlled via the condenser duty and tight control is usually achievable with a simple SISO loop. The liquid inventory can usually also be controlled by two simple SISO controllers provided either the distillate rate or the reflux rate is used to control the reflux accumulator level and either the bottoms rate or the reboiler duty is used to control the column base level.

Therefore, two degrees of freedom remain for the control of the process. If neither of these variables is used within a control loop (i.e. the process operator manipulates the control values directly), the column is said to be operated in open-loop or manual. If only one of these variables is manipulated automatically to control a measured property, a one-point or single composition control scheme is deemed to be in use. In this case, the remaining degree of freedom is usually fixed at a constant value or manipulated only occasionally to reflect capacity constraints (e.g. maximum reboiler duty or flooding). Finally, both available degrees of freedom can be utilized within control loops. This is known as a two-point or dual composition control.

3.4.2 One point control

One point control is relatively easy to implement, is not subject to interactions between opposing composition control loops, and provides a form of effective constraint management.

The ability to implicitly incorporate constraint management into a control scheme is often more important than the composition control of the secondary product in an industrial environment. This is possible with one-point control but is often difficult with two-point control as one less degree of freedom is present in the latter case. Throughput constraints such as column flooding and reboiler and condenser duty limitations are usually nearly proportional to the internal column flows. The manipulated variable that is not used for composition control in a one-point scheme can, therefore, be fixed at a value that corresponds closely to the equipment constraint. This is particularly effective if the unused manipulated variable is the reflux rate or reboiler duty as these variables substantially determine the internal column flows.

The relationship between the unused manipulated variable and the column constraints can also be seen if it is considered that the two degrees of freedom in a distillation process determine the feed-split and fractionation. In a two point control scheme, the feed-split controls the primary product composition and fractionation control the secondary product composition. Fractionation is a function of the internal column flows, as are the likely column constraints.(i.e. flooding, reboiler duty, etc.).

The properties of one point distillation control make it an attractive option in many cases, but the implementation of a one-point control scheme for an reactive distillation column maybe present some difficulties. These aspects relate primarily to the selection of the controlled variable and the organization of the manipulated variable.

The controlled variable should always be selected in order to reflect the process objectives as reliably as possible. Before this can be done, it is necessary to determine the priority of the process objectives since there is only one available degree of freedom in a one point control scheme. It is also necessary to find methods of determining whether the process objectives are bring met.

The most important process objective is likely to be the control of the bottoms product composition as the process will normally be constrained by external quality specifications. This makes the bottoms product composition the most likely control objective. There are essentially three methods for monitoring product composition: (a) directly, with one or more online analyzers; (b) indirectly, using a temperature or pressure corrected temperature to infer composition; and, (c) externally, using process samples taken at regular intervals and appropriate laboratory equipment. Analyzers have many advantages but are costly, require regular maintenance, and usually introduce a significant time delay in the process. Inferential control is cheaper and, often, more reliable but can also be less accurate. The use of external measuring equipment (e.g. laboratory techniques) limits the measurement frequency and is unsuitable for closed loop control.

If some form of inferential control is to be used (either in a closed-loop or open-loop system) to monitor product composition, the temperature sensor must be located carefully to ensure that changes in the composition are accurately reflected and good sensitivity to set point changes is provided. The reboiler sump is commonly used as a sensing location as it minimized process dead time and sensitivity is usually high (except with very high product purities). It might be expected that variations in the reboiler temperature (at constant pressure) would directly relate to changes in the product composition whereas other locations would be susceptible to interference from changes in the stage-to-stage composition profile.

The control configuration designations refer to the two manipulated variables that are not used for inventory control. In one-point control, one of these variables is used to control a product composition and one is fixed or adjusted only intermittently (normally to manage an equipment constraint).The distillate and bottom product draw rates directly affect the feed split (i.e. the column material balance) while the reflux rate and reboiler duty only affect fractionation. The relative magnitudes of these effects differ by approximately an order of magnitude so that the manipulated variable selected for composition control should be one of the product draw rates where a choice exists. With this restriction, it is possible to ascertain the variable pairings that are implied by each control configurations. These are indicated in Table 3-3 for several common control schemes, including some ratio schemes. Figure 3-4 depicts the control connection for the LV configuration. The other configurations can be setup similarly.

The ratio control schemes have been strongly recommended by some authors but are generally more suited to two-point control. These schemes effectively result in one or more non-diagonal 2×2 controllers (i.e. a MIMO controller instead of several SISO controllers). For example, schemes involving L/D or the reflux ratio L/(L+D) lead to controller that manipulates both L and D for reflux drum level control, regardless of how the composition ratio is configured. The advantages of ratio control schemes generally centre on the implicit decouling that is achieved. For example, the reflux ratio and boilup ratio are essentially independent while the reflux rate and the reboiler duty are closely related. This feature of ratio control schemes is highly advantageous for two-point composition control but less important for one point composition control where control loop interactions are less significant. The disadvantages of ratio schemes are that they often make consistent operation at equipment constraints harder to achieve and they increase the complexity of the control problem.

3.4.3 Two point control

There are usually at least two major process objectives in reactive distillation: to control the purity of the desired product; and to maximize the conversion of reactants. Only one of these objectives can be satisfied automatically with a one point control structure although some implicit control of the secondary objective might be realizable if the control system is formulated well. If more direct and exact control of the secondary objective is required, two point control must be implemented.

A combined composition and conversion control scheme which automatically ensures that satisfactory reaction conditions are maintained was implemented on the reactive distillation column using the inferential conversion model and the LV control configuration. The five degrees of freedom that are available are utilized according to the variable pairings given in Table 3-4.

The reboiler duty and reflux rate were assigned for composition and conversion control, respectively. The distillate draw rate and bottoms draw rate were assigned to level control according to dynamic responsiveness considerations and the condenser duty was used for pressure control as is the popular convention. This arrangement is also shown diagrammatically in Figure 3-5, to indicate where the inferential model was applied.

Table 3-3 Distillation control configurations

Configuration	Composition Control		Inventory Control	
	Varied	Fixed	Reflux Accumulator	Reboiler Sump
LV	Reflux rate/ Reboiler duty	Reboiler duty/ Reflux rate	Distillate rate	Bottoms rate
LB	Bottoms rate	Reflux rate	Distillate rate	Reboiler duty
DV	Distillate rate	Reboiler duty	Reflux rate	Bottoms rate
(L/D)V	Reboiler duty	Reflux ratio	Distillate rate	Bottoms rate
(L/D)(V/B)	Reflux ratio/ Boilup ratio	Boilup ratio/ Reflux ratio	Distillate rate	Bottoms rate
(V/B)L	Reflux rate	Boilup ratio	Distilalte rate	Bottoms rate
(D/F)V	Distillate yield	Reboiler duty	Reflux rate	Bottoms rate

Table 3-4 Variable pairings for the two point reactive distillation control

Controlled variable	Manipulated variable
Pressure (overhead)	Condenser duty
Reflux accumulator level	Distillate draw rate
Reboiler sump level	Bottoms draw rate
Bottoms composition	Reboiler duty
Reactant conversion	Reflux rate

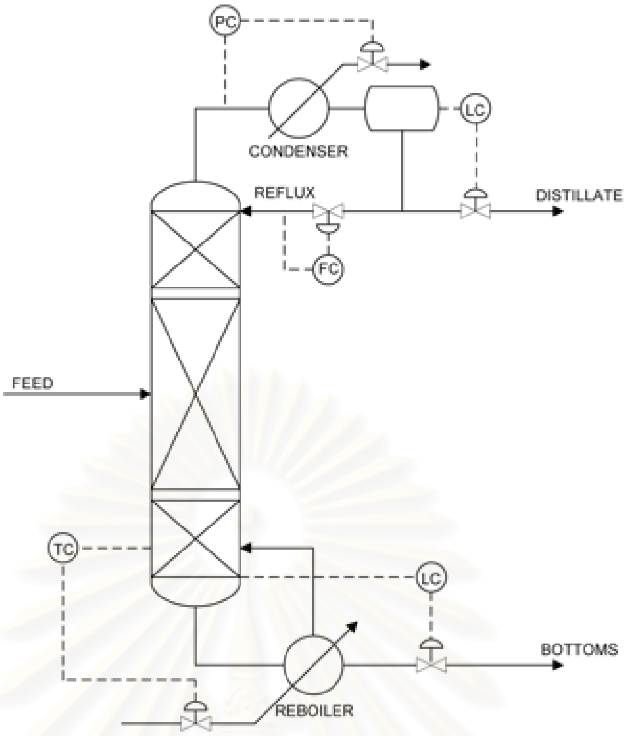


Figure 3-4 One point control configuration for reactive distillation columns.

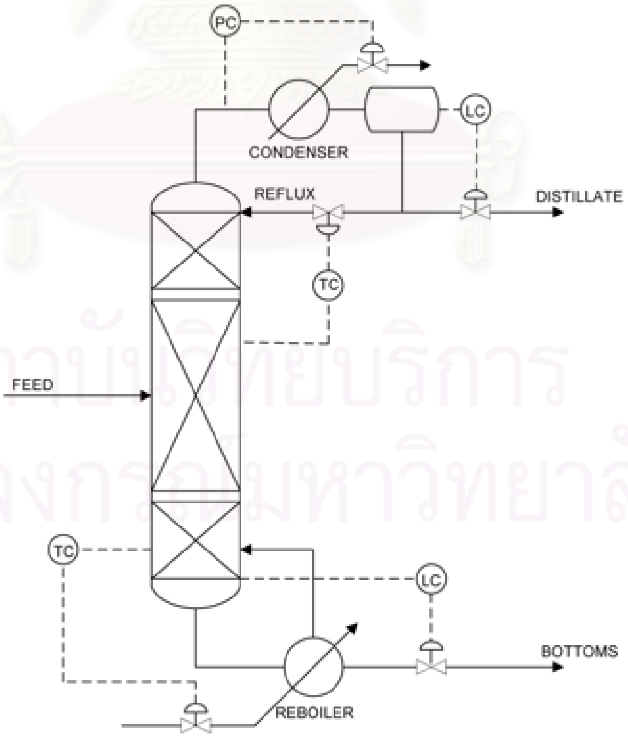


Figure 3-5 Two point control configuration for reactive distillation columns.

CHAPTER IV

DYNAMIC MODELING
OF REACTIVE DISTILLATION COLUMN

4.1 Modeling

Figure 4-1 shows the reactive distillation for butyl acetate production. There are three zones in the column. The rectification zone and stripping zone operate exactly as a nonreactive distillation column, purifying top and bottom products. Butyl acetate is formed in the reaction zone. The overhead vapor, with a composition close to that of the heterogeneous ternary azeotrope between water, butanol, and butyl acetate, is condensed and then separated into aqueous and organic phases in the decanter. The aqueous phase is completely withdrawn, whereas the organic phase is completely refluxed to the column. Pure butyl acetate is withdrawn from the column bottom.

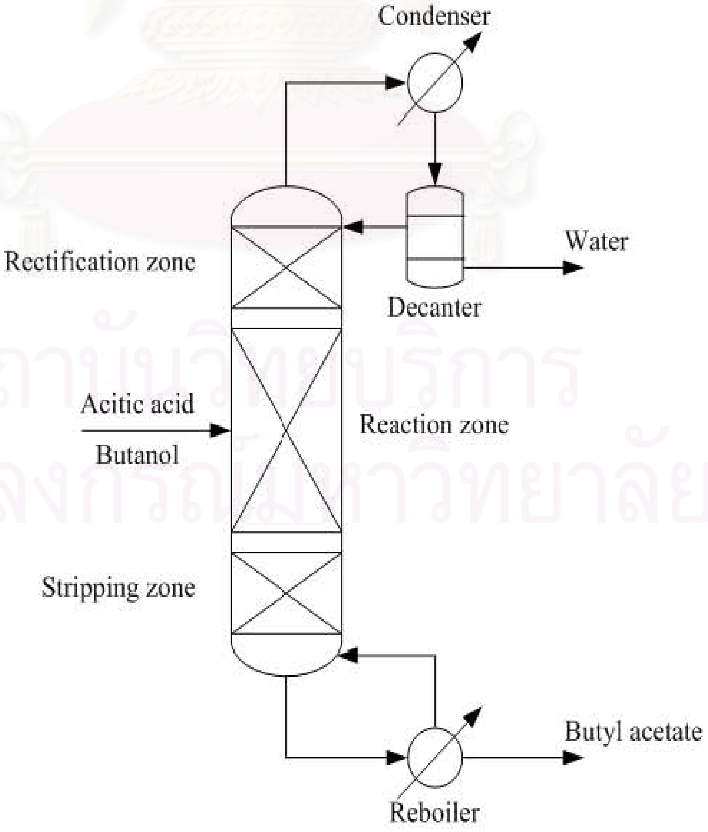


Figure 4-1 Reactive distillation column for butyl acetate synthesis

The mathematical model of reactive distillation column for butyl acetate production is developed based on the MESH (material balance, phase equilibrium, mole fraction summations and heat balance) equations. The assumptions used are:

1. Vapor and liquid on each stage are in vapor-liquid equilibrium and no liquid-liquid phase splitting is assumed inside the column.
2. Two liquid phases in decanter are in liquid-liquid equilibrium.
3. Reaction proceeds only in the liquid phase in reactive zone and their course can be described by kinetic model.
4. Constant pressure drop through the column is assumed and heat losses across the column wall are neglected.
5. Enthalpy per stage equals to enthalpy of liquid on the stage (i.e. vapor phase enthalpy neglected).
6. A quasistatic energy energy balance was considered, to calculate the liquid and vapor flow rate.
7. Liquid holdup in the condenser, decanter and reboiler are held perfectly constant changing the flow rates of distillate, reflux and bottom.
8. Reflux R and heat input to the reboiler Q_R are held constant for dynamic open-loop simulation.

The above assumptions lead to the model that consists of a set of ordinary differential and algebraic equations. Differential equations are obtained from mass and energy balances around each stage of the reactive distillation column. The algebraic equations describe the particular variables, physical properties and reaction kinetics. The stages are numbered from top to bottom. Stage 1 represents the decanter, and stage N represents the reboiler. Thus, the model equations are written as follows:

Stage model

Overall material balance on stage *j*:

$$\frac{dU_j}{dt} = V_{j+1} + L_{j-1} + F_j - V_j - L_j + \left(\sum_i v_i\right) r_j \tag{4-1}$$

Component *i* material balance on stage *j*:

$$\frac{dU_j x_{i,j}}{dt} = y_{i,j+1}V_{j+1} + x_{i,j-1}L_{j-1} + z_{i,j}F_j - y_{i,j}V_j - x_{i,j}L_j + v_i r_j \tag{4-2}$$

Enthalpy balance on stage *j*:

$$\frac{dU_j H_j}{dt} = V_{j+1}H_{j+1}^V + L_{j-1}H_{j-1}^L + F_j H_j^F - V_j H_j^V - L_j H_j^L - Q_j \tag{4-3}$$

It is noted that enthalpy is calculated such a way that the heat of reaction is calculated implicitly and need not be included in the equation.

Condenser model (total condenser)

Overall material balance:

$$\frac{dU_1}{dt} = V_2 - L_1 \tag{4-4}$$

Component material balance:

$$\frac{d(U_1 x_{i,1})}{dt} = y_{i,2}V_2 - x_{i,1}L_1 \tag{4-5}$$

Energy balance:

$$\frac{d(U_1 H)}{dt} = V_2 H_2^V - L_1 H_1^L - Q_c \tag{4-6}$$

Reboiler model (partial reboiler)

Overall material balance:

$$\frac{dU_N}{dt} = L_{N-1} - L_N - V_N \tag{4-7}$$

Component material balance:

$$\frac{d(U_N x_{i,N})}{dt} = x_{i,N-1}L_{N-1} - x_{i,N}L_N - y_{i,N}V_N \tag{4-8}$$

Energy balance:

$$\frac{d(U_N H)}{dt} = L_{N-1}H_{N-1}^{L} - L_N H_N^{L} - V_N H_N^{V} + Q_r \tag{4-9}$$

Decanter model

Overall material balance

$$L_1 = R + D \tag{4-10}$$

Component material balance

$$x_{i,1}L_1 = x_{i,1}^{O}R + x_{i,1}^{A}D \tag{4-11}$$

Definition equations

Vapor-liquid equilibrium relations (modified Raoult's law):

$$y_{i,j}P = \gamma_{i,j}x_{i,j}P_j^0 \tag{4-12}$$

Liquid-liquid equilibrium relations:

$$\gamma_{i,j}^{a}x_{i,j}^{a} = \gamma_{i,j}^{o}x_{i,j}^{o} \tag{4-13}$$

Mole fraction summations:

For decanter, $\sum x_{i,j}^{O} = 1 \qquad \sum x_{i,j}^{A} = 1$ (4-14)

For equilibrium stage, $\sum x_{i,j} = 1 \qquad \sum y_{i,j} = 1$ (4-15)

Reaction rate formula:

$$r = r(x, T, m_{cat}) \tag{4-16}$$

Thermodynamic properties:

$$\rho = \rho(x, T) \tag{4-17}$$

$$H^{L} = H^{L}(x, T) \tag{4-18}$$

$$H^{V} = H^{V}(y, T) \tag{4-19}$$

4.2 Solution method

The model equations described above (Eqns. (4-1)-(4-19)) corresponds to differential algebraic equations system. Due to its flexibility and computational power, the MATLAB simulation tool is used to implement the full dynamic simulation of reactive distillation for butyl acetate production. Solution method involves solving total material balance and energy balance equations separately as algebraic equations and component material balance are treated as differential equation. The solution strategy is as follows:

1. Determine operating conditions, specifications of the column, and initial conditions (i.e., liquid compositions, liquid holdup and pressure on all stages).
2. Calculate temperatures, activities coefficients and vapor compositions from UNIQUAC equations and phase equilibrium relation, using the bubble point method.
3. Calculate liquid and vapor phase enthalpies.
4. Calculate reaction rate from kinetic model.
5. Solve total material and energy balance equations for vapor and liquid flows (non-linear algebraic equations).
6. Calculate the new liquid compositions from integration all ordinary differential equations using Euler method.
7. Go back to step 2 and repeat for the next step in time.

4.3 Simulation results

The feed conditions and column specifications for butyl acetate reactive distillation under study in this work are shown in Table 4-1. The column consists of 39 stages, including a total condenser, a partial reboiler and 37 column stages. There are three zones in the column. Rectification zone (stage 2-11) and stripping zone (stage 25-38) operate exactly as in a nonreactive distillation column, purifying top and bottom products. Feed, comprised of 50 % mol acetic acid and 50% mol butanol, is fed to the column on stage 17, numbering from top to bottom. Butyl acetate and water are formed in the reaction zone between stage 12 and stage 24. The column is operated at 1 atm. The pressure drop in the column is assumed to be 0.134 atm. The column is sized as a tray column. A liquid holdup of about 850 L was obtained. We assume that a reactor volume of 800 L is available on each tray in the reactive section. Once the dynamic model had been implemented, it has been validated in dynamic simulation and compared against available data. The accuracy of the dynamic simulation results of reactive distillation obtained under this work is verified by comparing with the literature. The computer simulation parameters were the same in literature. The steady state simulation has been carried out to get the initial values for dynamic simulations. Because of the difference in the time scale of composition and holdup, the component material balance equations have been modeled as ordinary differential equations and the remaining equations as algebraic equations. Solution method described in section 4.2 was made to solve algebraic and differential equations separately in sequence. Solution method could achieve the fast convergence and the robustness during the simulation. Dynamic simulations resulted in convergence to the new steady state value. However, the high purity of butyl acetate in bottom product can't be achieved in our simulation when using the same parameters with the literature. To increase the performance of product separation in reactive distillation, heat supplied to the reboiler is adjusted to increase butyl acetate purity in bottom product. The reboiler duty is varied from 2.0 to 2.5 Mcal/h while

Table 4-1 Feed conditions and column specifications

Rectification section	Stage 1-11
Reaction section	Stage 12-24
Stripping section	Stage 25-39
Reflux rate (kmol/h)	100.76
Overhead pressure (atm)	1
Pressure drop (atm)	0.134
Feed stage	17
Feed flowrate (kmol/h)	200
Feedcomposition (mol%)	50% HOAc, 50% BuOH
Feed temperature (°C)	25
Feed pressure (atm)	1

reflux rate, feed flowrate, feed composition are kept constant at design specification value. From simulation results, high butyl acetate purity in bottom product close to 99.96 % mol can be achieved when the reboiler duty is more than 2.4 Mcal/h. Therefore, reboiler duty at 2.4 Mcal/h has been used as nominal operating condition. Figure 4-3 shows the composition and temperature profiles in the column at nominal operating conditions. As can be seen from the simulation results in Figure 4-2, the liquid phase becomes almost-pure butyl acetate in the stripping section of the column immediately after the reactive zone. The stripping section of the column accomplishes good separation efficiency for separating acetic acid from butyl acetate. Table 4-2 shows the results of the simulation run in comparison to the literature. Accuracy of simulation is good with regard to condenser temperature, rectifying zone temperature, reaction zone temperature, stripping zone temperature, reboiler temperature, internal flow, butanol conversion and bottom butyl acetate purity. The mismatch in some data, e.g., reboiler duty, can possibly be due to simplifications of mathematical model, differences of the solution method, the kinetic and vapor-liquid equilibrium description for highly nonideal and multicomponent system.

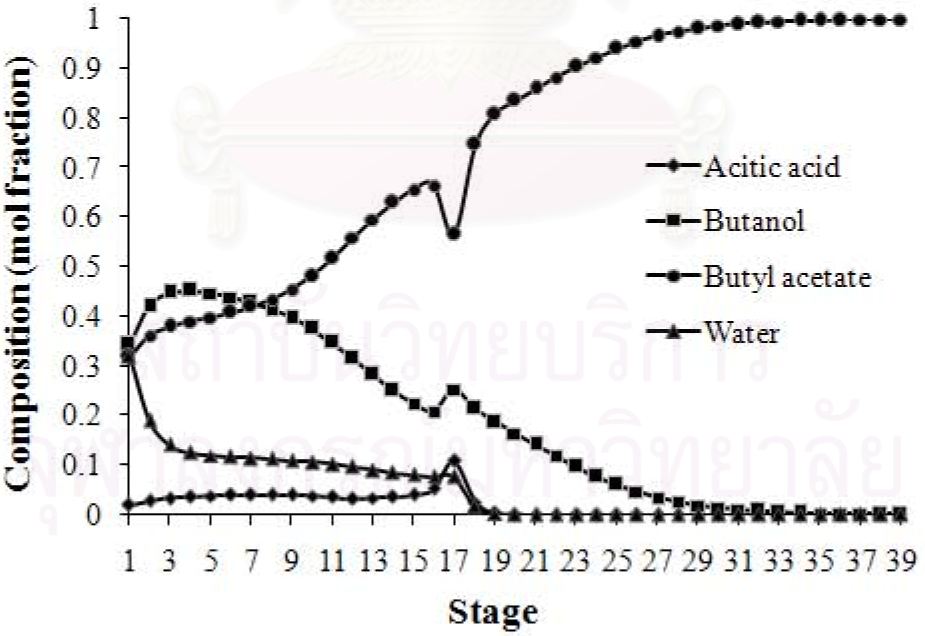


Figure 4-2 Liquid compositions profiles at nominal operating condition

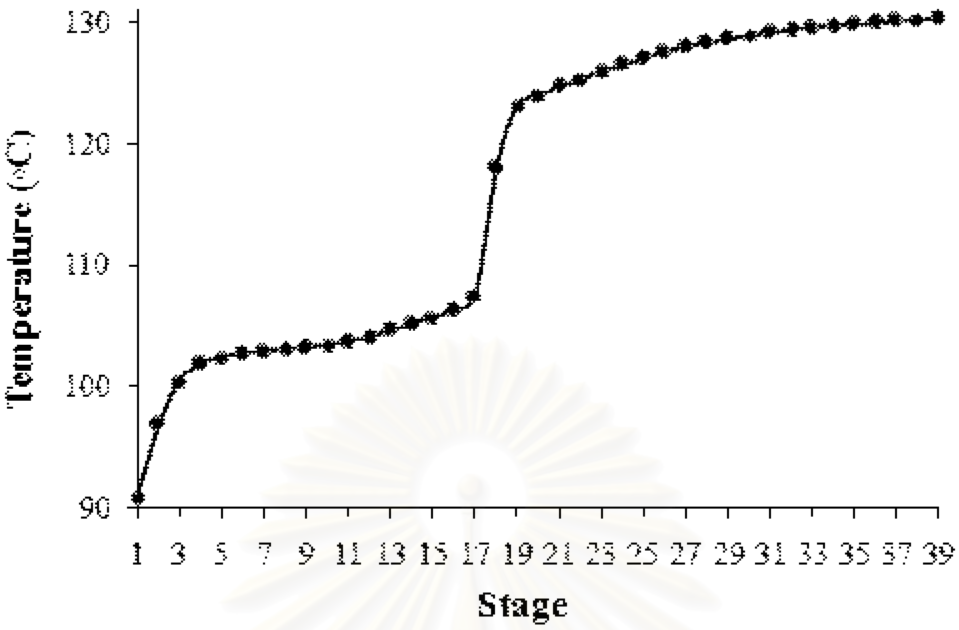


Figure 4-3 Temperature profiles at nominal operating condition

Table 4-2 Reactive distillation column – comparison of simulation data and literature

Parameter	Simulation data	Literature data
Feed flow (kmol/h)	200.00	200.00
Organic reflux rate (kmol/h)	100.76	100.76
Aqueous distillate rate (kmol/h)	101.84	100.75
Bottom rate (kmol/h)	98.16	99.20
Reboiler duty (Mcal/h)	2.40	1.98
Butanol conversion (mol basis)	0.990	0.993
Bottom butyl acetate purity (mol %)	99.96	99.9
Condenser Temperature (°C)	90.86	91.00
Rectifying zone Temperature (°C)	96.85-103.67	96.41- 103.17
Reaction Zone Temperature (°C)	104.02-126.48	103.94-124.76
Stripping zone temperature (°C)	127.04-130.21	125.65-130.0
Reboiler Temperature (°C)	130.33	130.2

Effect of column inputs on output variables

The input-ouput relationships can be complex for reactive distillation due to the high non-linearity introduced by simultaneous reaction and separation. Input variables are manipulated variables such as the reboiler duty, reflux rate or feed flowrate while output variables are controlled variables such as stage temperature/compositions. The potential inputs that can be manipulated for column regulation are the reflux rate and the reboiler duty. In the present system, the reflux rate is decided by the relative proportion of organic phase and aqueous phase in the top product and it is not a variable. The only operating variable that can make significant impact on the separation and reaction is the boil-up rate. Hence, it was decided to study the effect of boil-up rate on output variables of system. Manipulating the reboiler heat duty causes an immediate change in the boil-up rate throughout the column so the reboiler duty can be used for regulating the separation and the reaction in the column.

Influence of reboiler duty

The reboiler duty is usually the main manipulated variable, it is important to understand the effects of changes on the performance of the reactive distillation column during operation. In reactive distillation, the reboiler duty must be set to ensure sufficient recycle of unreacted, heavy reactant to the reaction zone without excluding the light reactant from the reaction zone. If the reboiler duty is too high or too low, conversion, and subsequently product purity, may be reduced. Figure 4-4 indicates the effect of reboiler duty on conversion and butyl acetate purity in the bottom product. It was observed that, at reboiler duty < 2.4 Mcal/h, both butanol conversion and butyl acetate purity increase as reboiler duty increases. After 2.4 Mcal/h of reboiler duty, no significant change was observed in purity while conversion decreases and approaches to a constant after 2.7 Mcal/h of reboiler duty. Therefore, a heat supply of 2.4 Mcal/h is sufficient to obtain higher purity and conversion for the synthesis of a butyl acetate system at a feed flowrate of 200 kmol/h, if heat loss is negligible. Furthermore, the dynamic simulation results show that the responses of butyl acetate purity take less time to reach to the new steady state at higher reboiler duty. Figure 4-5 and 4-6 show the effect of manipulating reboiler

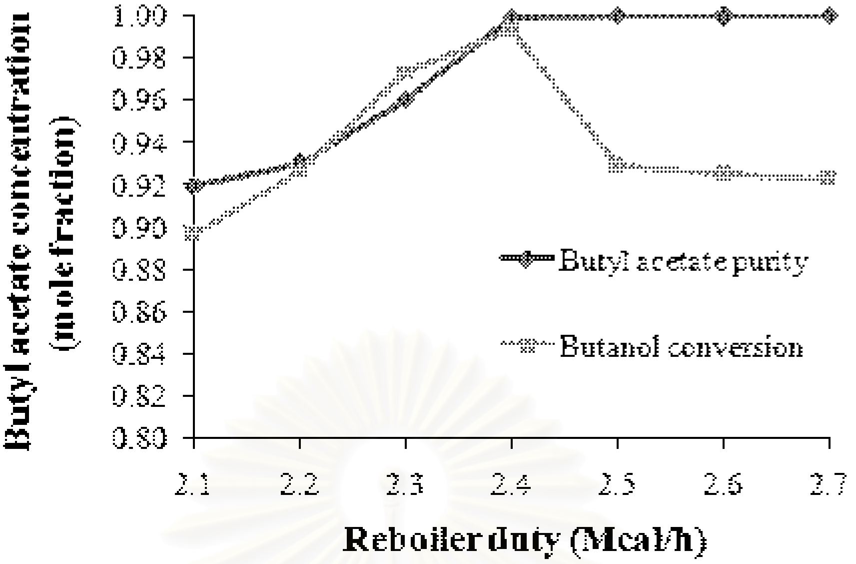


Figure 4-4 Influence of reboiler duty on butyl acetate purity and acetic acid conversion

heat duty at different levels (2.1, 2.2, 2.3 and 2.4 Mcal/h) on the temperature profile and liquid composition of butyl acetate along the column. It can be seen that, depending on the value of the heat duty, the temperature profile and butyl acetate mole fraction in the column switches to different paths. By fixing the pressure in column, the change in stage temperature is due to the change in composition only. According to Figure 4-5 and 4-6, increasing the reboiler heat duty can increase both the butyl acetate concentration in stripping section and temperature profile in column. The temperature profile and butyl acetate mole fraction in stripping section changes were not significant when increasing reboiler duty from 2.1 to 2.3 Mcal/h. On the other hand, the changes in reboiler duty from 2.3 to 2.4 Mcal/h produce the significant changes in stage temperature and butyl acetate mole fraction along the column.

Open loop transients

Reactive distillation for butyl acetate production was simulated under open-loop operation. The effect of disturbances is studied in order to investigate the dynamic performance of the system. Also it gives an idea how much time it takes to reach to the new steady state for the given disturbance. The dynamic simulation of reactive distillation for butyl acetate production was utilized to analyze open-loop dynamic responses for some disturbances to the system: (1) Effect of feed rate, (2) Effect of feed composition, and (3) Effect of reflux rate. The magnitudes of the step changes were sufficient to have a significant affect on the column operation but were not so large that the column stability was compromised.

Effects of a Step Change in Feed Rate. The open loop transient response to a 10 % step increase in the total feed rate with pressure, reboiler duty and reflux rate held constant was employed and the changes in reactive stage temperatures as well as the corresponding changes in the liquid composition for feed stage are shown in Figure 4-7 and 4-8, respectively. Figure 4-9 shows the changes in butyl acetate purity and bottom temperature resulting from the same disturbance. Without an increase in heat input, system has no ability to reject the disturbance as indicated by the changes in the reactive stage temperatures and liquid composition for feed stage from their nominal operating value. The butyl acetate purity drops around 4.6 mol % and the bottom temperature decreases approximately 1.5 °C. The new butanol conversions decrease less than 4 mol %.

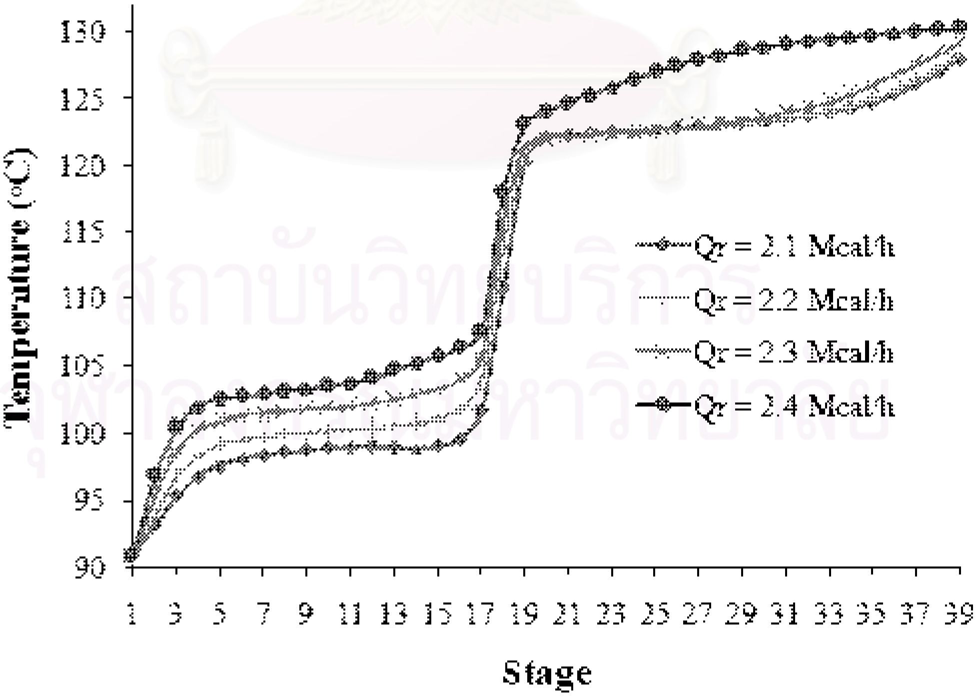


Figure 4-5 Temperature profile in the column

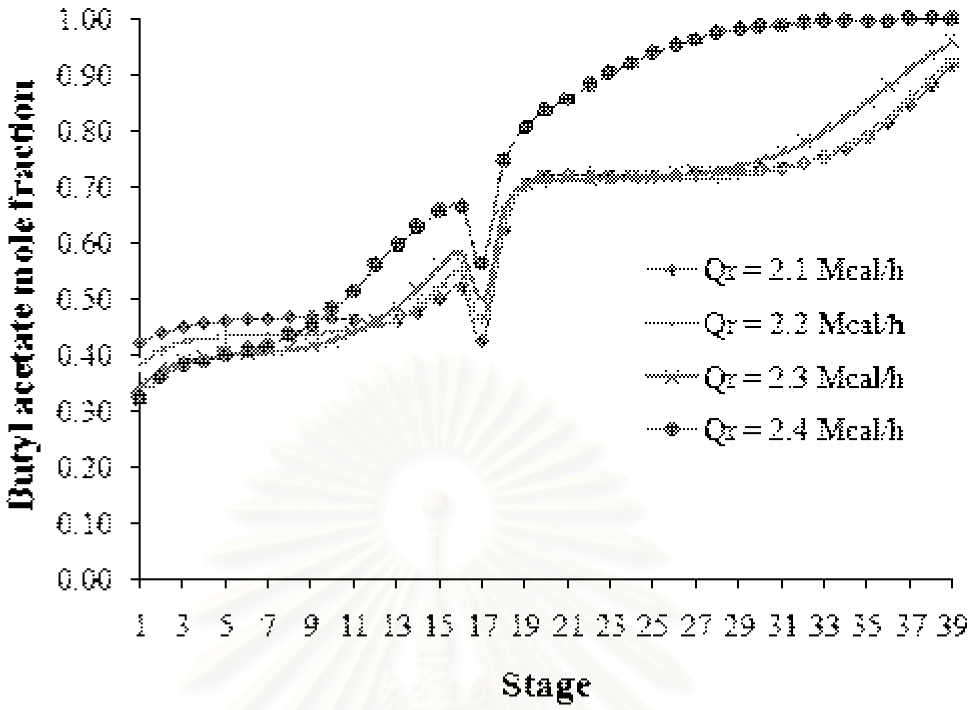


Figure 4-6 Mole fraction of butyl acetate along the column.

The step increase in the feed rate supplied more reactants in the reactive section so that the butanol conversion should increase. However, the additional reactants decreased the column temperature and therefore decreased the reaction rate due to the constant reboiler duty. As a result, the overall butanol conversion was lower. Besides, the butyl acetate purity also decreased due to the additional feed that was withdrawn from the bottom of the column. The composition of the bottom product shifted to include the additional light material and the bottom temperature decreased accordingly as the reboiler remained in phase equilibrium.

Effects of a Step change in Feed Composition. The effect of perturbation in the feed composition was investigated. Starting with stable operation at nominal operating condition, increasing butanol concentration in feed flowrate was considered. The total molar feed rate was fixed as were the pressure, reboiler duty, and reflux rate. Figure 4-10 and 4-11 show the change in the reactive stage temperature and the corresponding changes in the liquid composition for the stage 17 resulting from a 10 % step increase in the butanol concentration in the total feed. Figrue 4-12 shows the changes in the butyl acetate purity and bottom temperature resulting from the same disturbance. The changes in butanol concentration for feed stage significantly

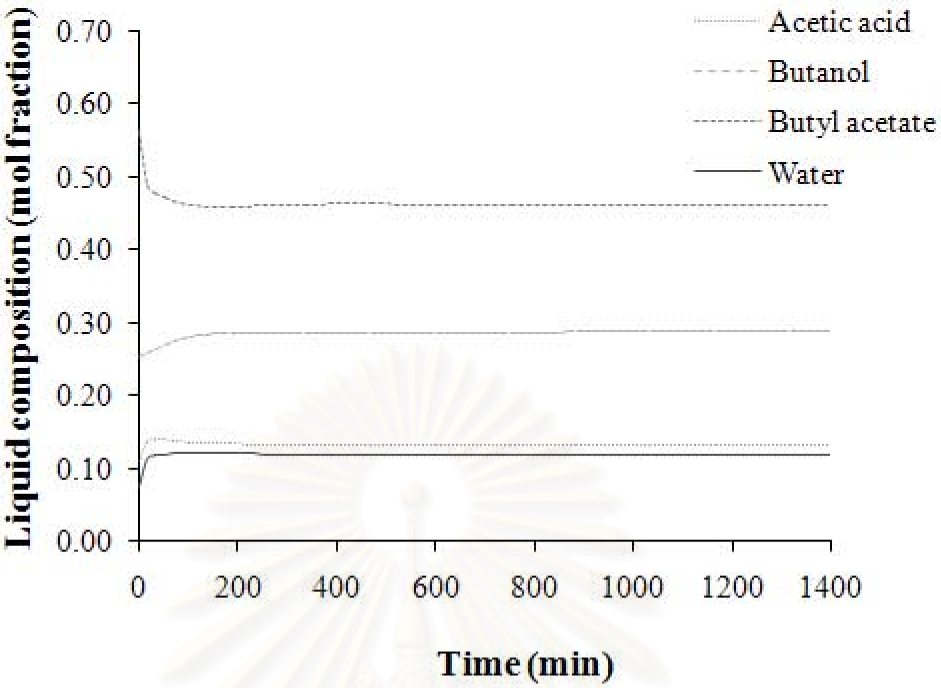


Figure 4-7 Changes in the liquid composition for the stage 17 (feed stage) to +10 % step increase in the feed rate for open-loop operation

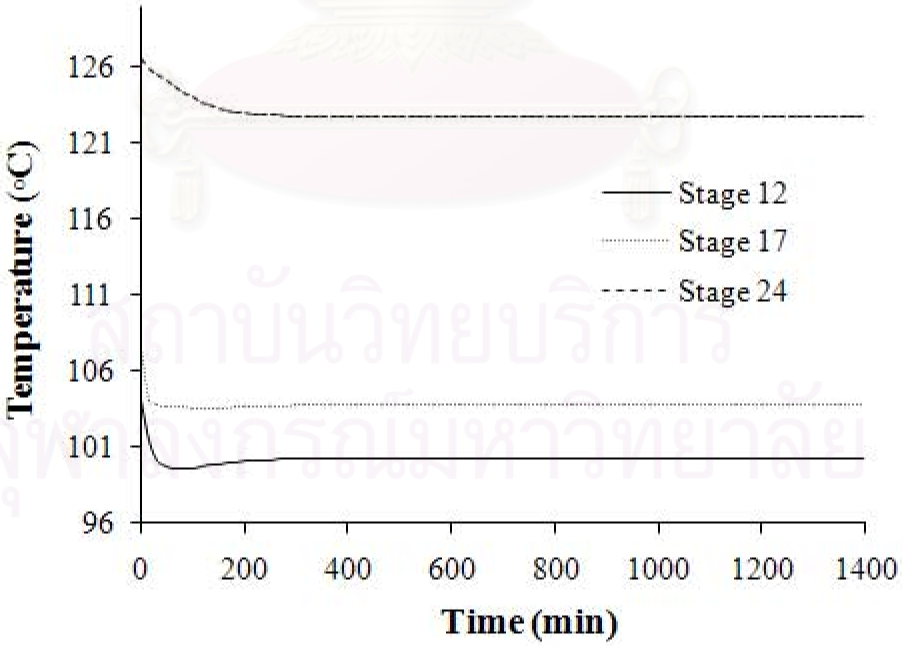


Figure 4-8 Changes in the reactive stage temperatures to +10 % step increase in the feed rate for open-loop operation

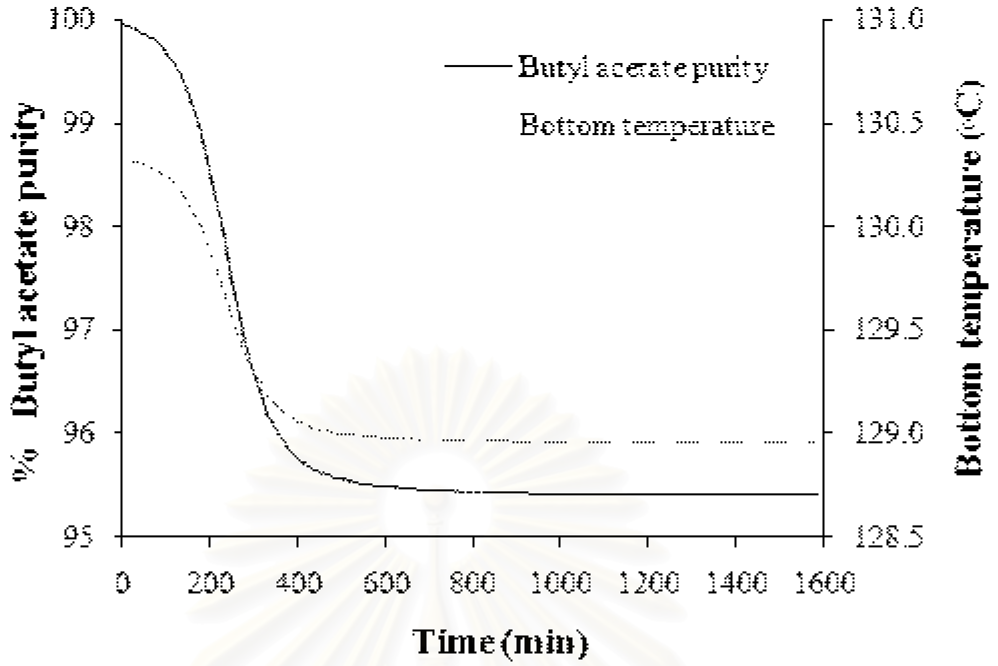


Figure 4-9 Changes in the butyl acetate purity and bottom temperature to +10 % step increase in the feed rate for open-loop operation

increases from 25 to 35 mol % while feed stage temperature slightly decreases, which are less than 1 °C. The butyl acetate purity and the bottom temperature decrease approximately 8 mol % and 2.4 °C, respectively. Butanol conversion slightly decreases as the butanol content of the bottoms product increased without additional heat input.

The step increase in the butanol concentration in the feed promoted further reaction so that the butanol conversion increased due to the additional driving force for the reaction. If the reboiler duty was kept constant, the column temperature was affected and the butyl acetate purity decreased due to the unreacted butanol that was withdrawn in the bottom product. The bottoms temperature followed the composition changes as phase equilibrium was maintained.

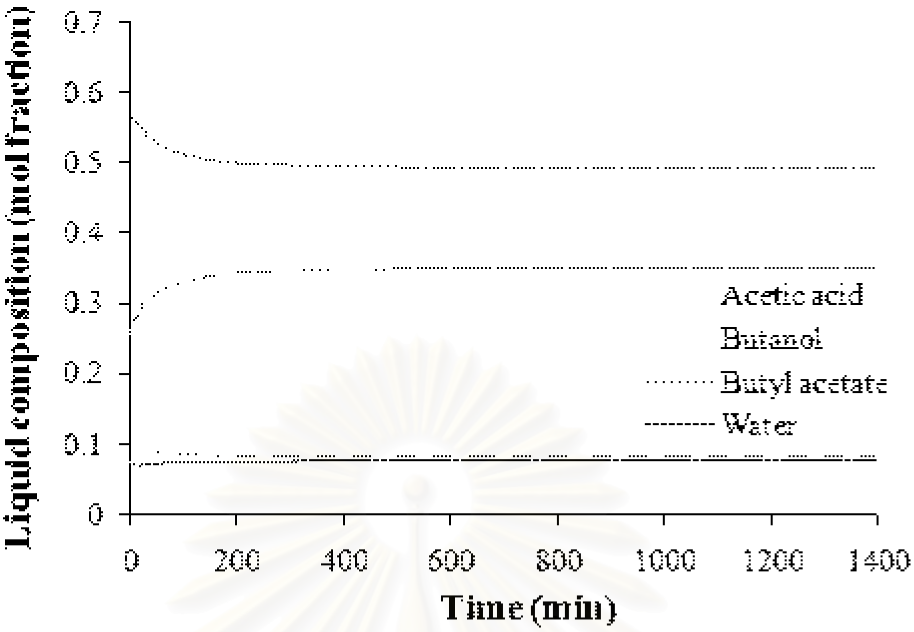


Figure 4-10 Changes in the liquid composition for the stage 17 (feed stage) to 10 % step increase in the butanol feed concentration for open-loop operation

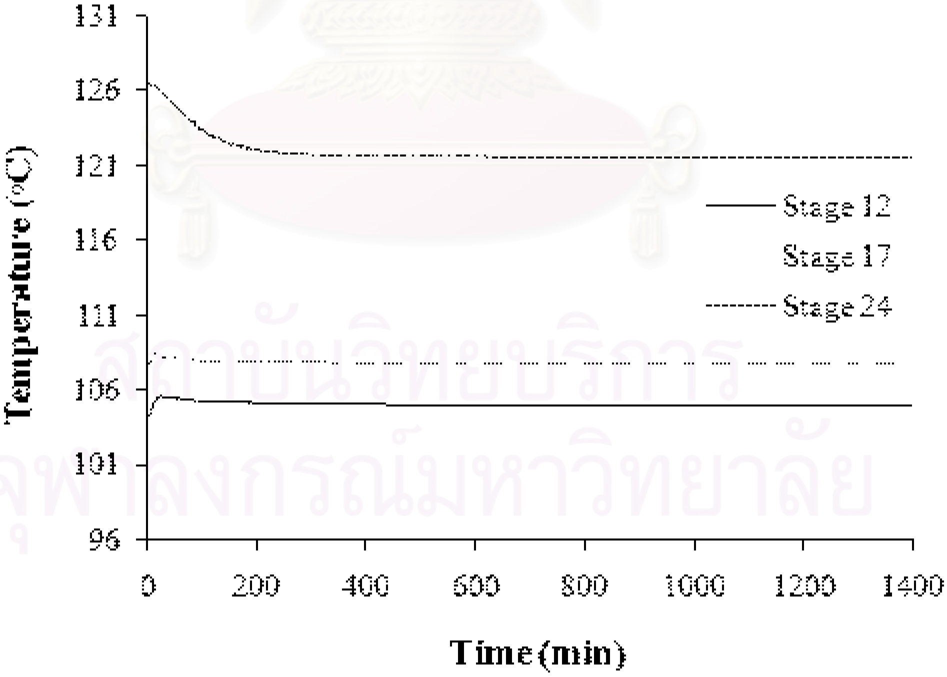


Figure 4-11 Changes in the reactive stage temperatures to + 10 % step increase in the butanol feed concentration for open-loop operation

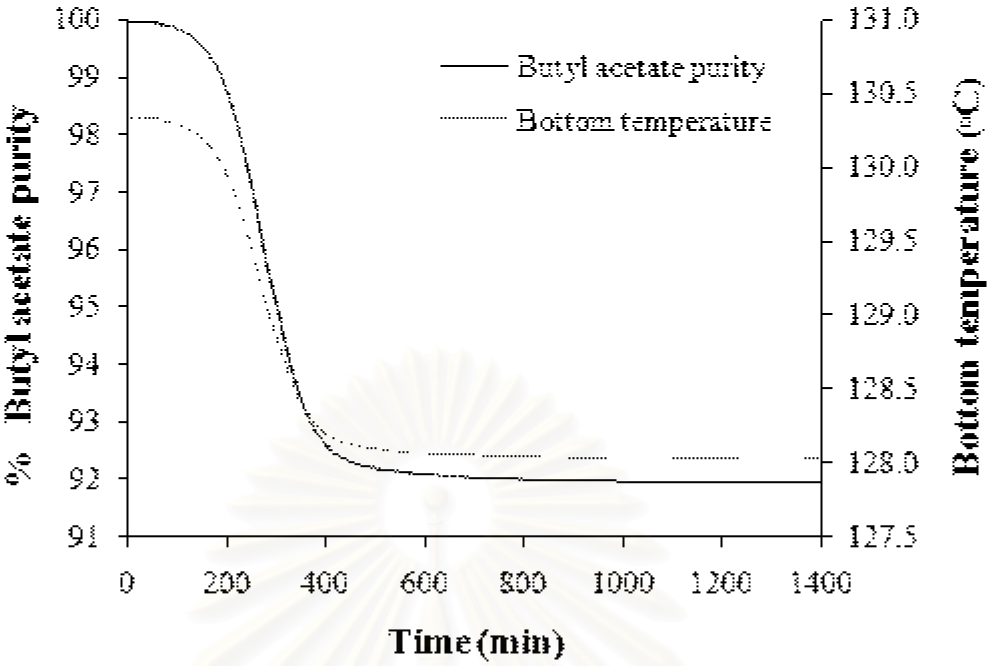


Figure 4-12 Changes in the butyl acetate purity and bottom temperature to +10 % step increase in the butanol feed concentration for open-loop operation

Effect of step change in reflux rate. The transient response to a 10 % increase in reflux rate is performed. The same column was used and pressure and reboiler duty were again held constant. The changes in the reactive stage temperatures and the liquid composition for feed stage are shown in Figure 4-13 and 4-14, respectively. Figure 4-15 shows the changes in butyl acetate purity and bottom temperature resulting from the same disturbance. Increasing reflux rate enhances recycles unreacted butanol to the reaction zone. As a result, the concentration of butanol in reaction zone increase approximately 5 % mol while butyl acetate concentration and temperature in the reaction zone decrease around 6 mol % and 1 °C, respectively.

Without an increase in reboiler duty, the column was effectively quenched by the extra reflux and butanol was shifted to the reboiler. The composition of the bottom product shifted to include the additional light material, so the purity of butyl acetate and bottom temperature moved away from nominal operating values.

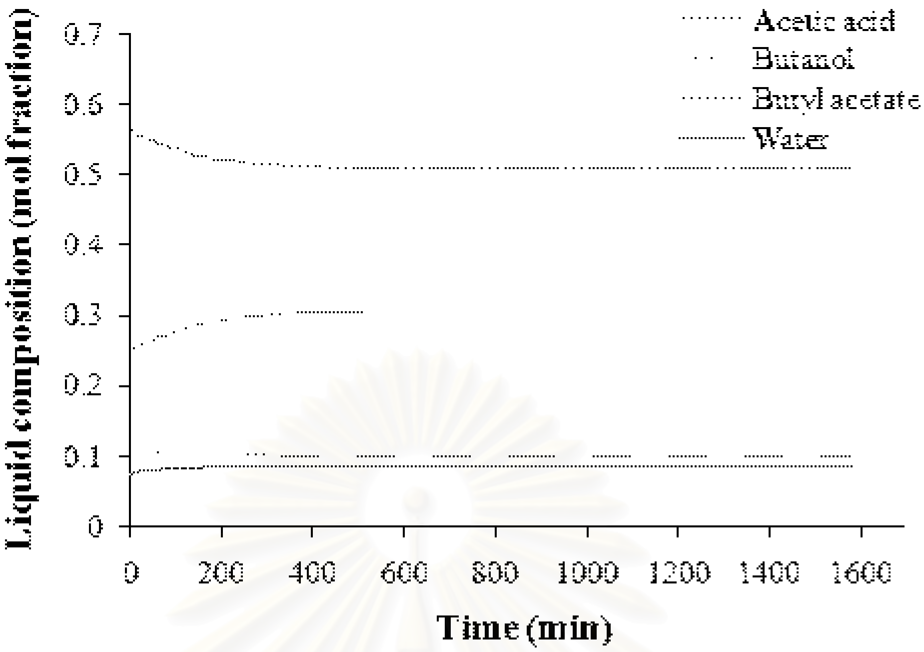


Figure 4-13 Changes in the liquid composition for the stage 17 (feed stage) to +10 % step increase in the reflux rate for open-loop operation

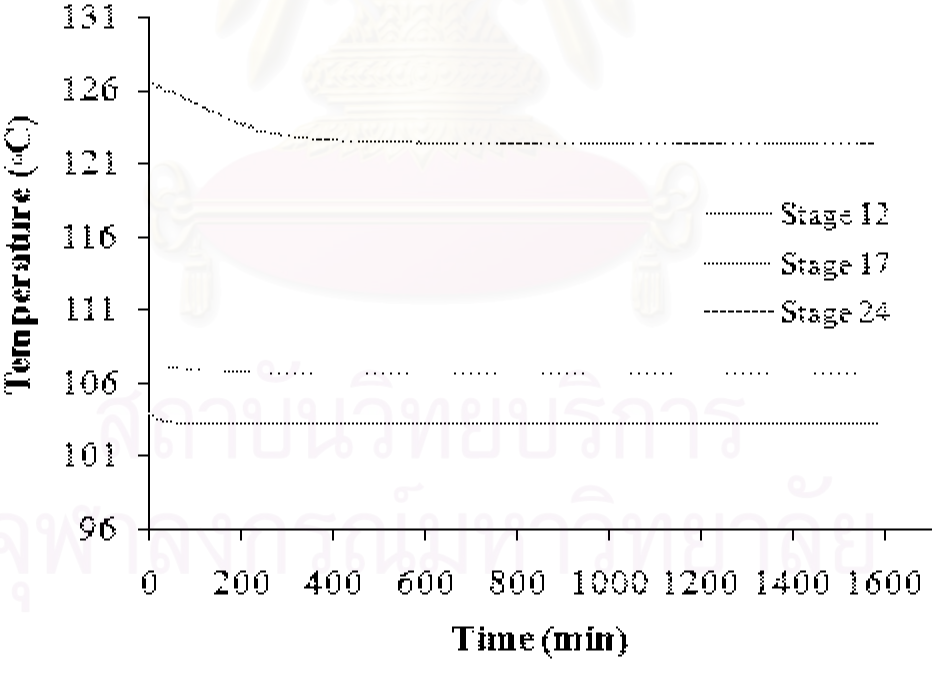


Figure 4-14 Changes in the reactive stage temperatures to +10 % step increase in the reflux rate for open-loop operation

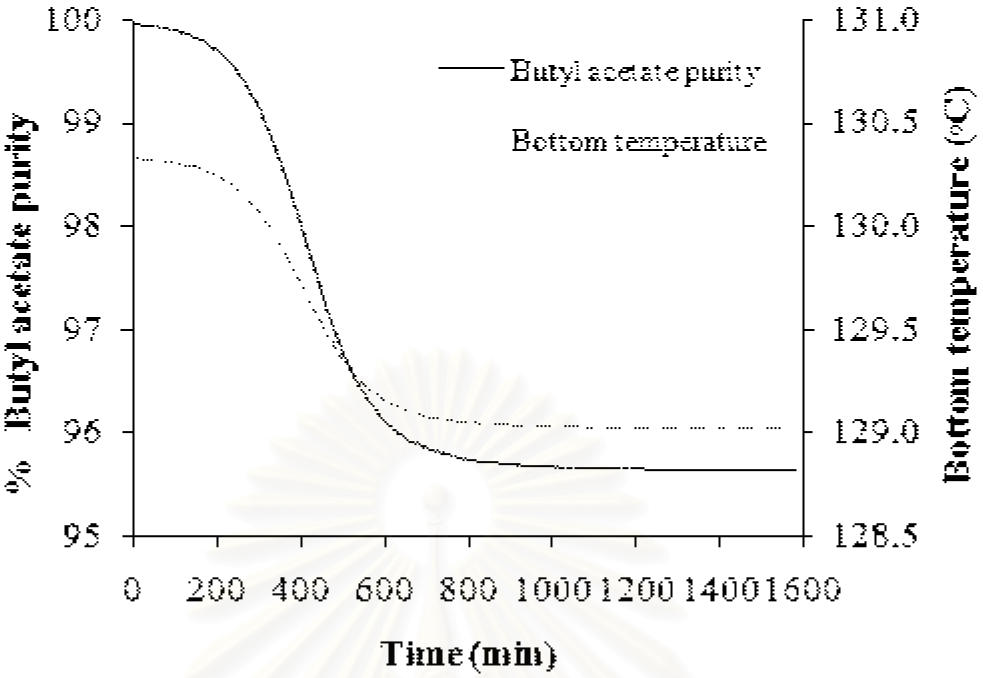


Figure 4-15 Changes in the butyl acetate purity and bottom temperature to +10 % step increase in the reflux rate for open-loop operation

CHAPTER V

MODEL PREDICTIVE CONTROL OF A REACTIVE DISTILLATION COLUMN

5.1 Selection of controlled and manipulated variables for product quality

The primary control objective of the column is to produce n-butyl acetate at bottom product with purity at its designed value. However, to reduce the expensive cost of using process analyzer for online composition measurement and the problem of its large measurement lag, we use some stage temperature measurement to infer the bottom product. The stage temperature control point is determined through open-loop sensitivity analysis to choose one stage temperature at maximum sensitivity while still exhibits near linear dynamic behavior. The relations between stage temperatures and reboiler duty are shown in Figure 5-1. It can be seen that the stage temperatures near the bottom of column (stage 37-39) can be good candidates for temperature control in our interest range. These temperatures have strong correlation with reboiler duty. However, when considering sensitivity between stage temperature and reboiler duty in Figure 5-2, the stage 37 temperature is the most sensitive to reboiler duty changes among the other stages. Therefore, this temperature is chosen as inferred variable for bottom product butyl acetate purity. Figure 5-3 shows the bottom product butyl acetate purity which has a linear relationship with stage 37 temperature.

However, in order to prevent accumulation of unreacted reactants in reactive stage and maintain correct temperature set point to ensure bottom product quality, the feed must be kept in a correct stoichiometric ratio when operating a reactive distillation column. The simplest way to maintain this balance is to use feed ratio control. Al-Arfaj and Luyben (2000) pointed out that a feed ratio control scheme is a feedforward scheme, which has no guarantee of the final product quality. When there is a measurement error or impurity in feed flow rate, the feed ratio control scheme will not be able to maintain stoichiometric balance. To overcome the shortcomings of a feed ratio control, Al-Arfaj and Luyben (2000) suggested that the composition of

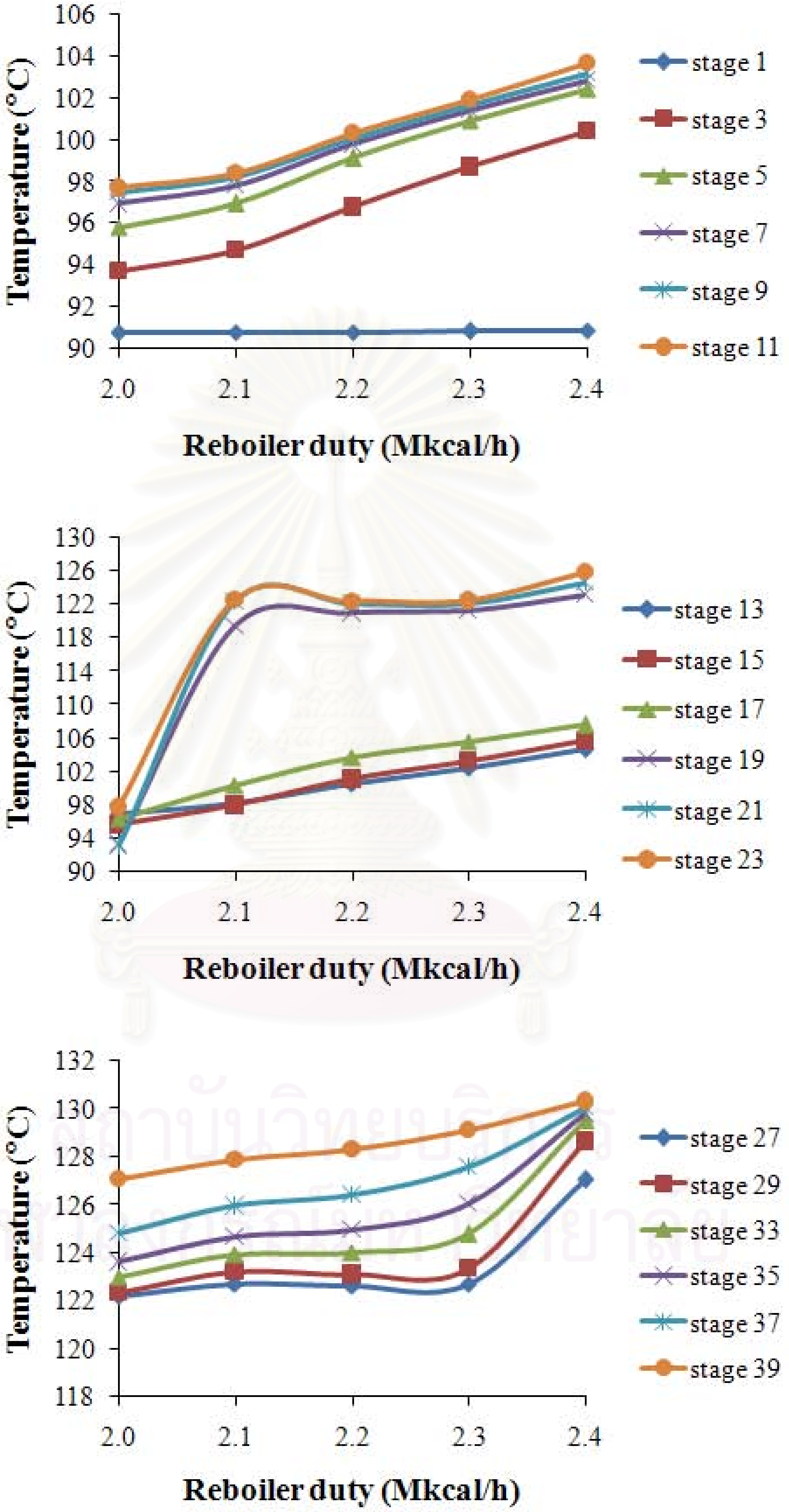


Figure 5-1 Relationship between stage temperature and reboiler duty

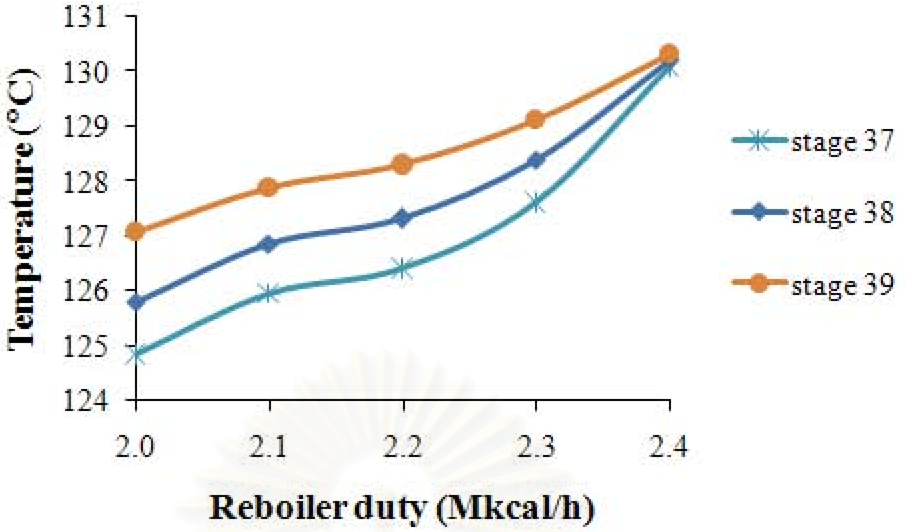


Figure 5- 2 Relationship between stage temperature near the bottom of column and reboiler duty

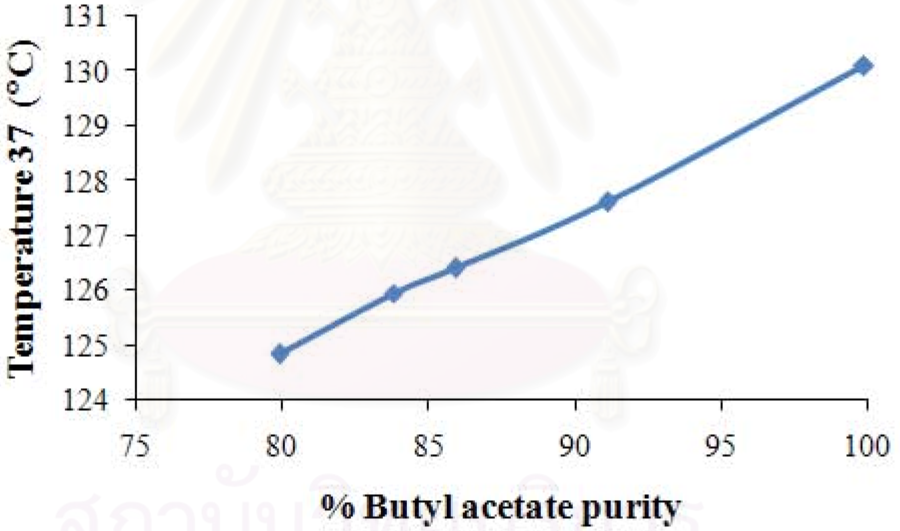


Figure 5-3 Relationship between stage 37 temperature and butyl acetate purity

some column stage be controlled by the feed ratio. However, the direct control of composition is necessary to use online composition measurement which is more expensive and has a longer dead time. Thus, in this work, we use some reactive stage temperature to infer the reactant composition in reactive zone as another alternative control strategy. For composition control in reactive zone, we should be careful to select the proper candidates for manipulated variable and controlled variable. Figure 5-4 and 5-5 show the effects of changes in the feed ratio on the acetic acid

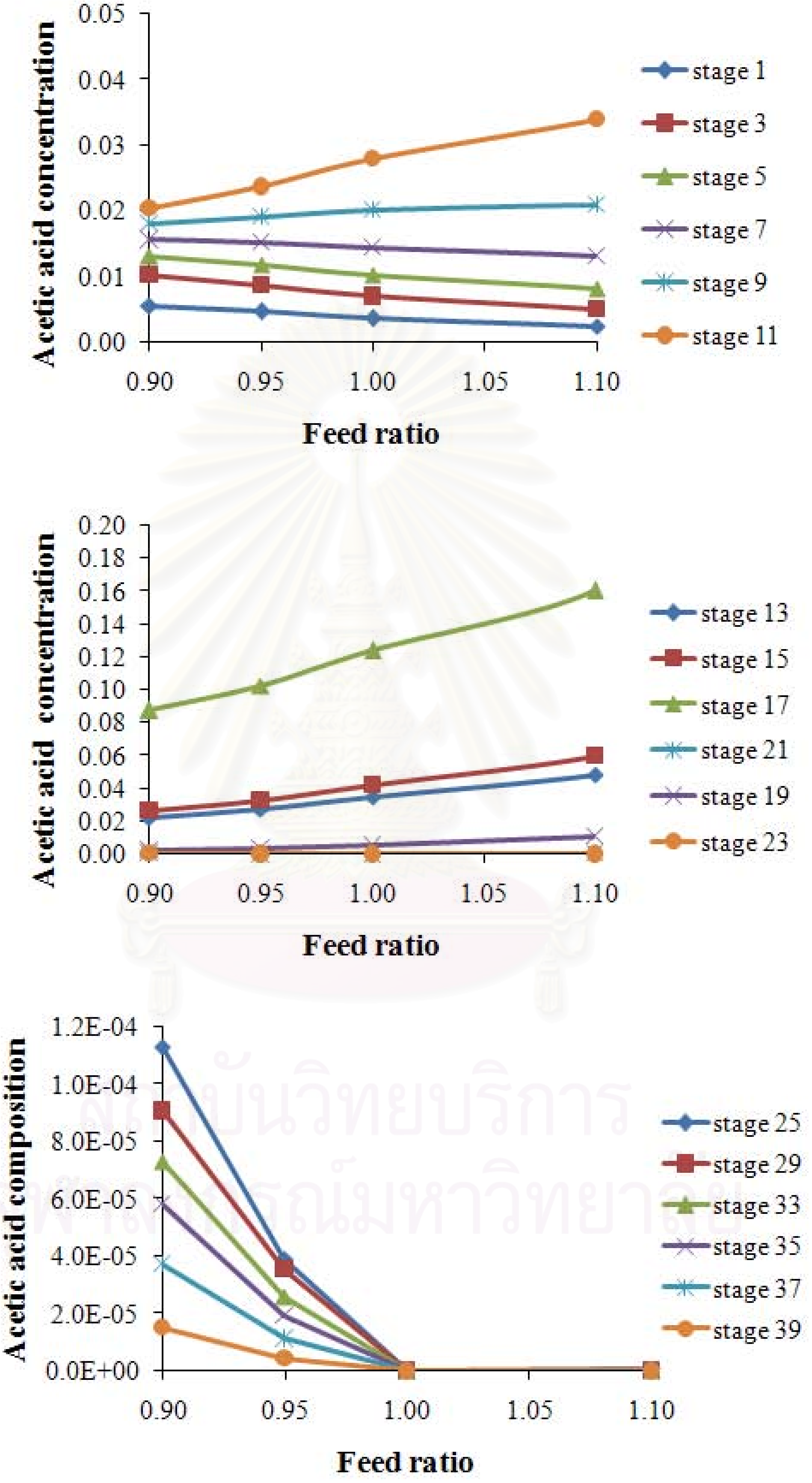


Figure 5-4 Relationship between acetic acid concentration and feed ratio

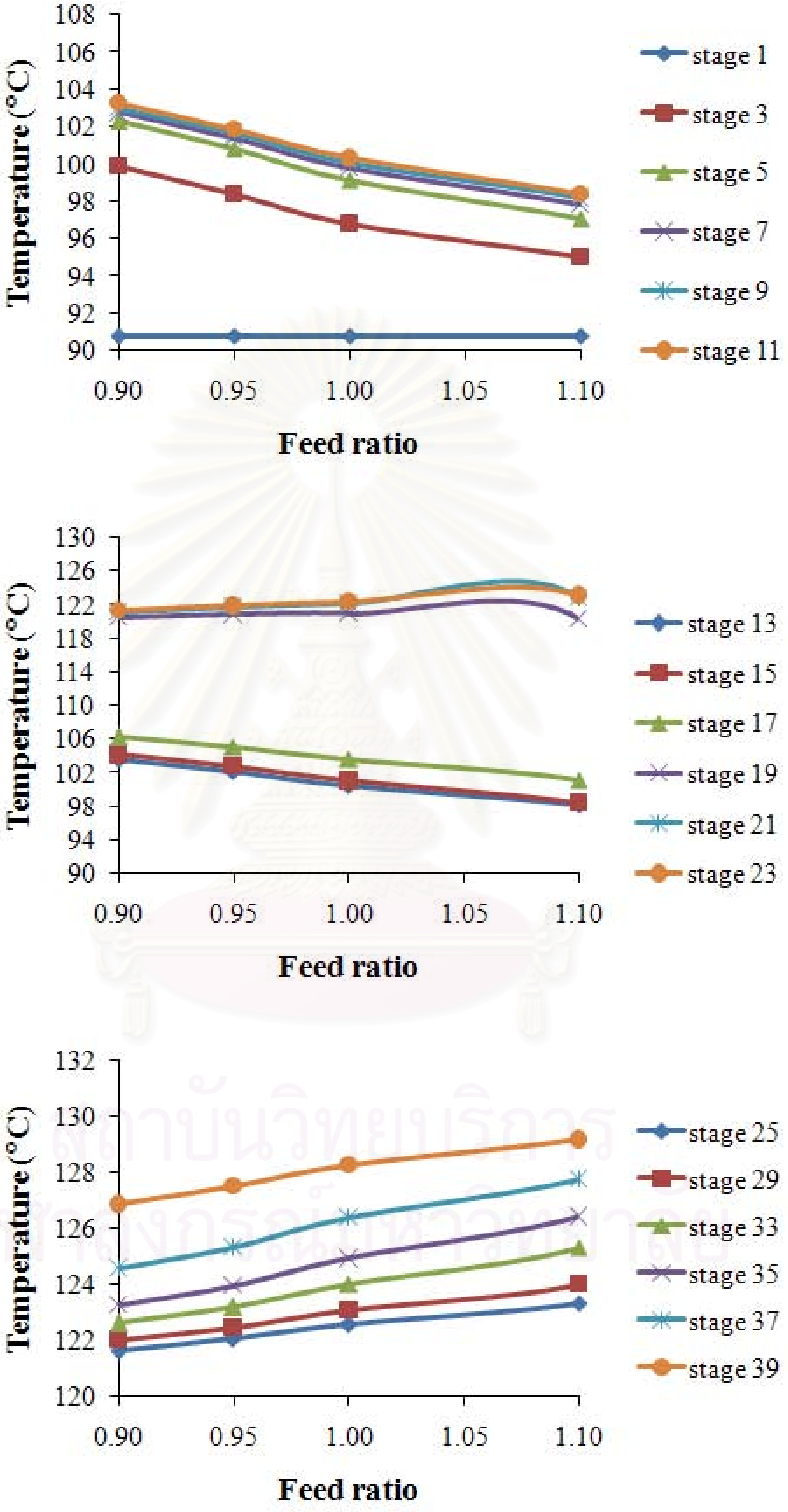


Figure 5-5 Relationship between stage temperature and feed ratio

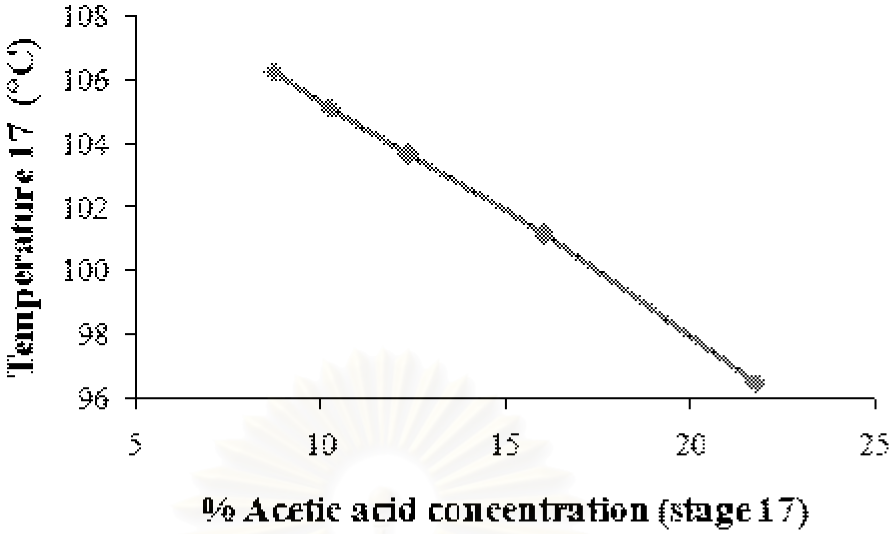


Figure 5-6 Relationship between stage 37 temperature and acetic acid composition

compositions and temperatures at different stages when fixing reboiler heat duty. According to these figures, the acetic acid composition and temperature at stage 17 have large sensitivity and linear behavior in our interest operating condition range. Therefore, temperature and acetic acid composition at 17th stage were chosen as the controlled variables of temperature and composition control loop, respectively.

5.2 Process model identification

A nonlinear dynamic model for a reactive distillation for butyl acetate production has been developed in the previous chapter. The dynamic model based on material balance, energy balance, phase equilibrium and reaction kinetics, contains hundreds of equations for the 39 stages RD process under consideration. It is too complicated to be directly used for control system design. Therefore, simplified input-output process models discussed in this section are helpful in understanding the complex process dynamics. They are identified under our interest operating condition range. Following that, the full dynamic model of a reactive distillation for butyl acetate production derived from previous chapter is utilized in performing step testing to generate input-output dynamic data from step changes in reboiler duty and feed ratio at the nominal condition. A simplified input-output dynamic model is generated from input-output dynamic data of the nonlinear reactive distillation process model

Table 5-1 Fitted Transfer Function

	Acetic acid composition at stage 17	Temperature 17	Temperature 37
Reboiler duty	$\frac{-0.335}{34.548s+1}$	$\frac{33.413}{21.682s+1}$	$\frac{16.737}{102.878s+1}$
Feed ratio	$\frac{0.940}{57.42s+1}$	$\frac{-71.491}{57.642s+1}$	$\frac{18.228}{98.0267s+1}$

using System Identification Toolbox in MATLAB software with sampling time $T_s = 1$ min and will be applied in the MPC controller algorithm. For close-loop dynamic simulation, the full dynamic model of nonlinear reactive distillation process model will be used to simulate the plant from which the output measurement will be obtained. This formulation generates model-plant mismatch in MPC controller algorithm, which makes the control problem more challenging.

A simplified input-output dynamic model, which is in transfer function form, is an expression which dynamically relates the input and the output of a process model. If a transfer function is known between one input and output, the change in the output can be computed for a change in the input. For particular control structure studied in this work, there are two manipulated variables which are the reboiler duty and feed ratio and two controlled variables which are temperature or acetic acid composition at stage 17th and temperature at stage 37th. As a result of this a 2×2 control structure there will have 4 transfer functions since each combination of input-output variables results in one transfer function. Figure 5-7 and Figure 5-8 show the dynamic response of the butyl acetate reactive distillation column for a 10 % increase in reboiler duty and a 10 % decrease in feed ratio, respectively. The shape of process variable responses to step changes in manipulated variables reveals that the process exhibits first-order behavior. Therefore, a transfer function between different pairs of input and output can be approximated by first-order equation. Table 5-1 shows the fitted transfer function between different pairs of input and output.

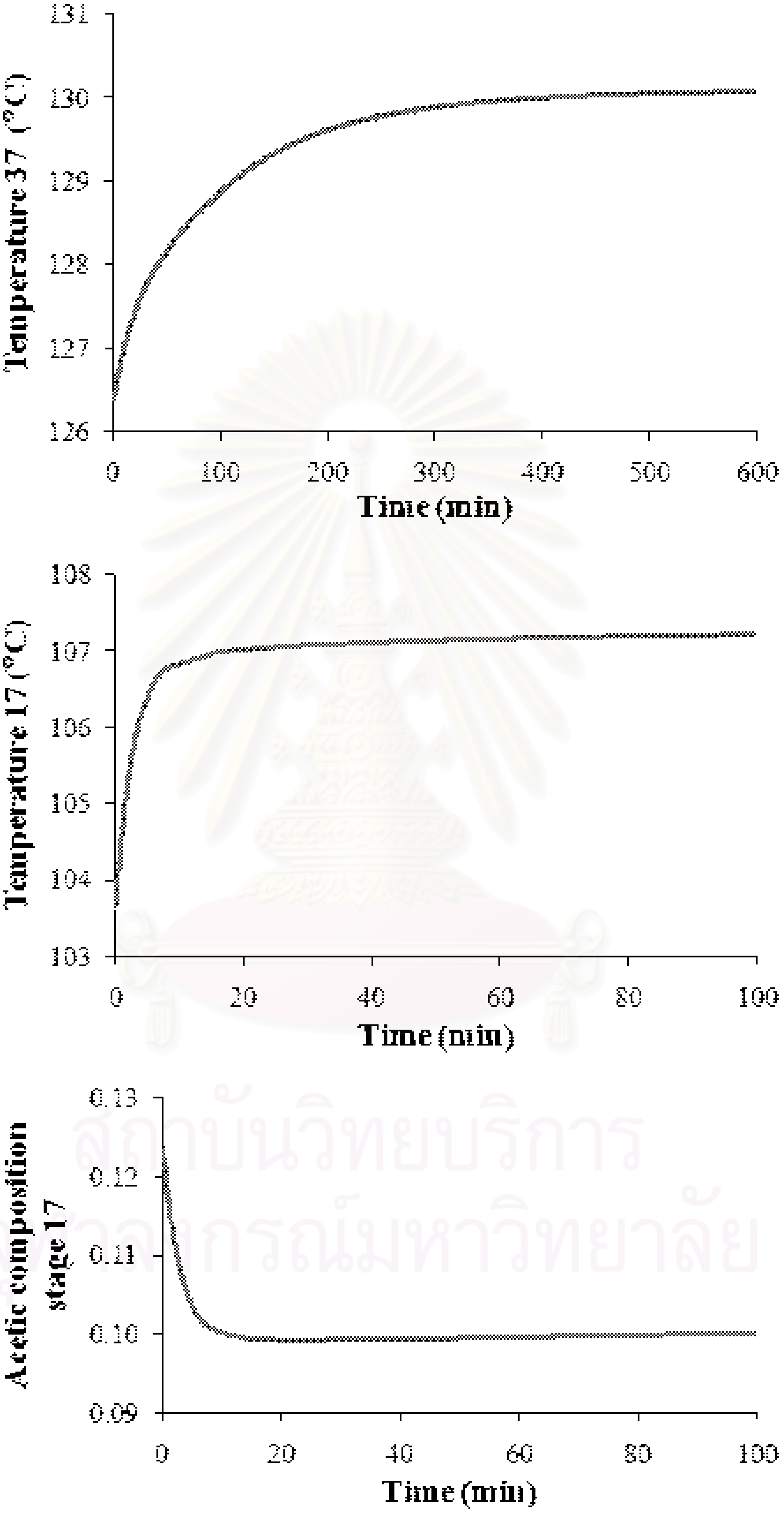


Figure 5-7 Open-loop response to + 10 % step change in reboiler duty.

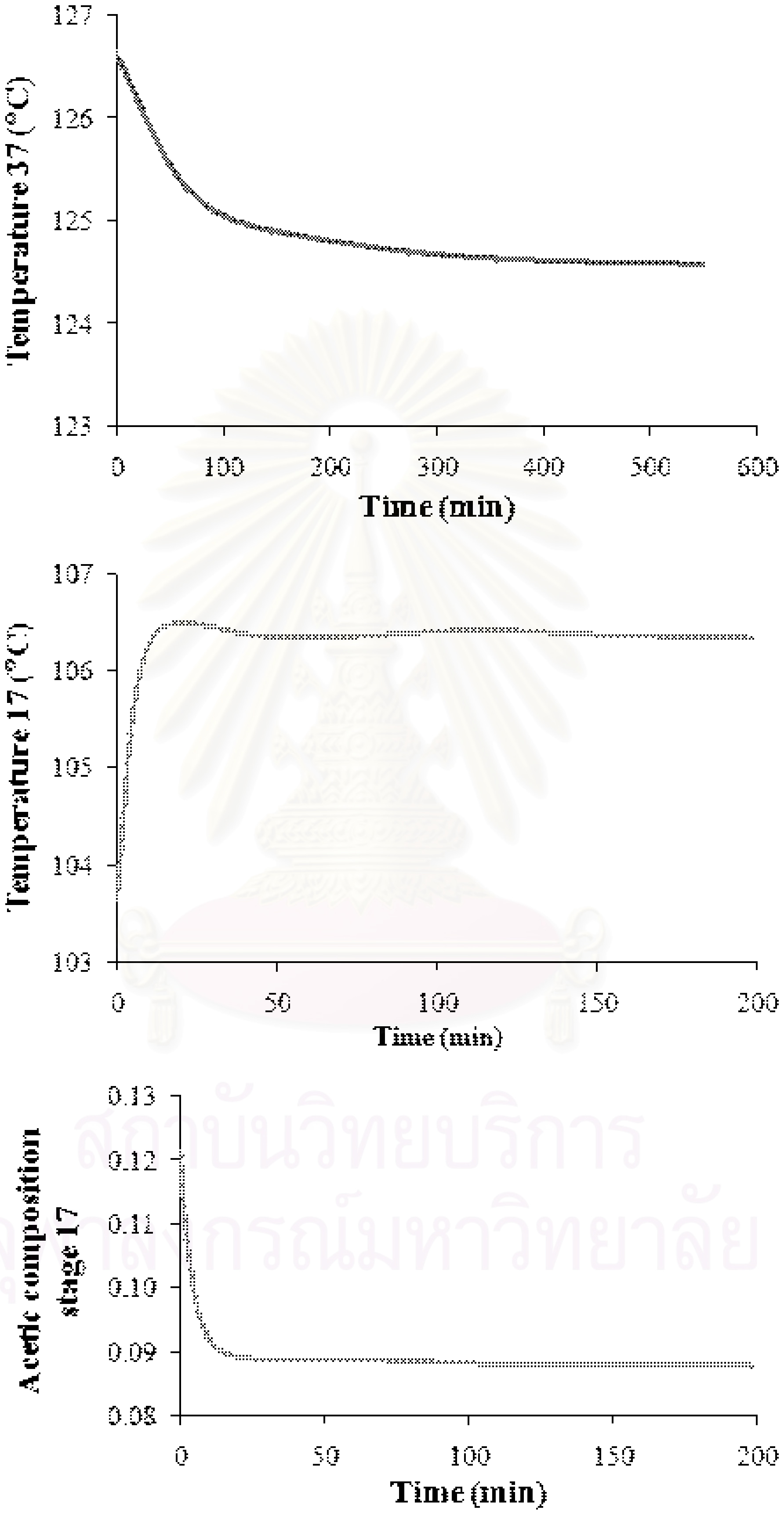


Figure 5-8 Open-loop response to -10 % step change in feed ratio.

5.3 Control configuration of reactive distillation for butyl acetate production

In this work, we considered two alternative control structures of reactive distillation for butyl acetate production shown in Figure 5-9 and 5-10, respectively. Our objective is to compare the close-loop performance of both control structures using MPC controller. However, to avoid the problem of process nonlinearity such as input multiplicity which occurs when reboiler duty > 2.4 Mkcal/h, we change our nominal reboiler duty from 2.4 to 2.2 Mkcal/h. The butyl acetate product purity corresponded with the reboiler duty at 2.2 Mkcal/h is about 93 % mol and this condition will be used as new standard condition to investigate the performance of both control structures.

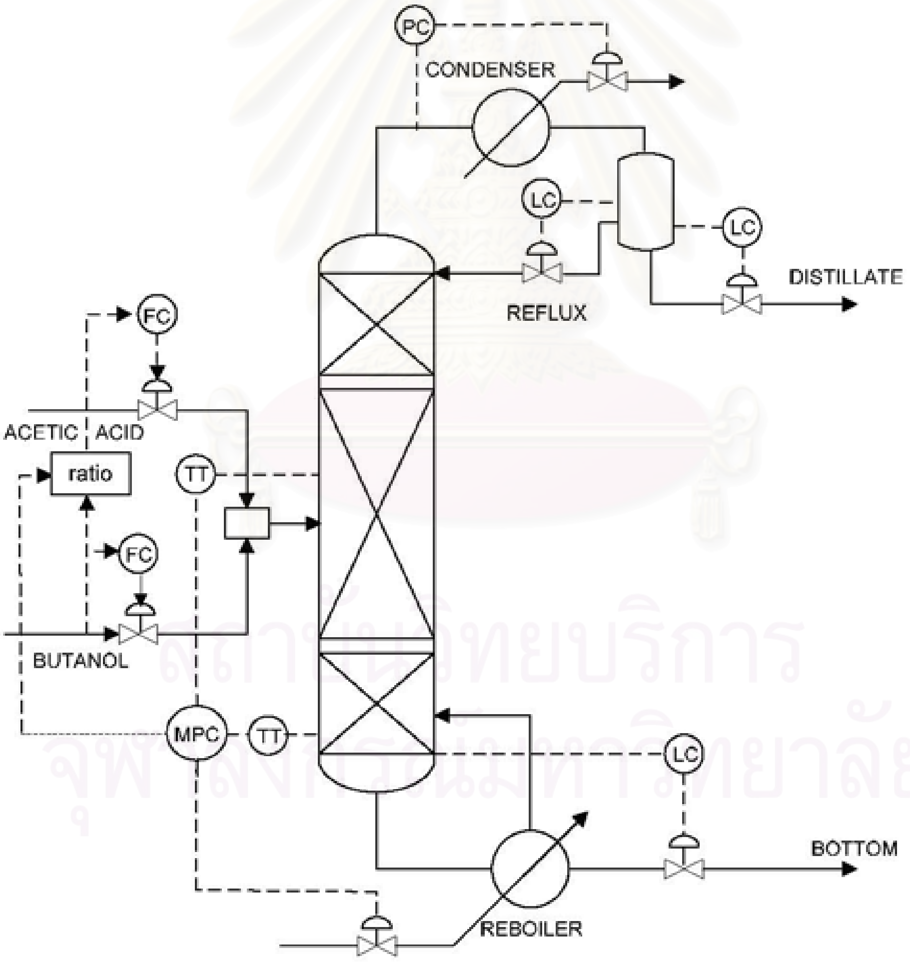


Figure 5-9 Control structure 1 (CS1) of reactive distillation for butyl acetate production (Temperature 17+ Temperature 37 control)

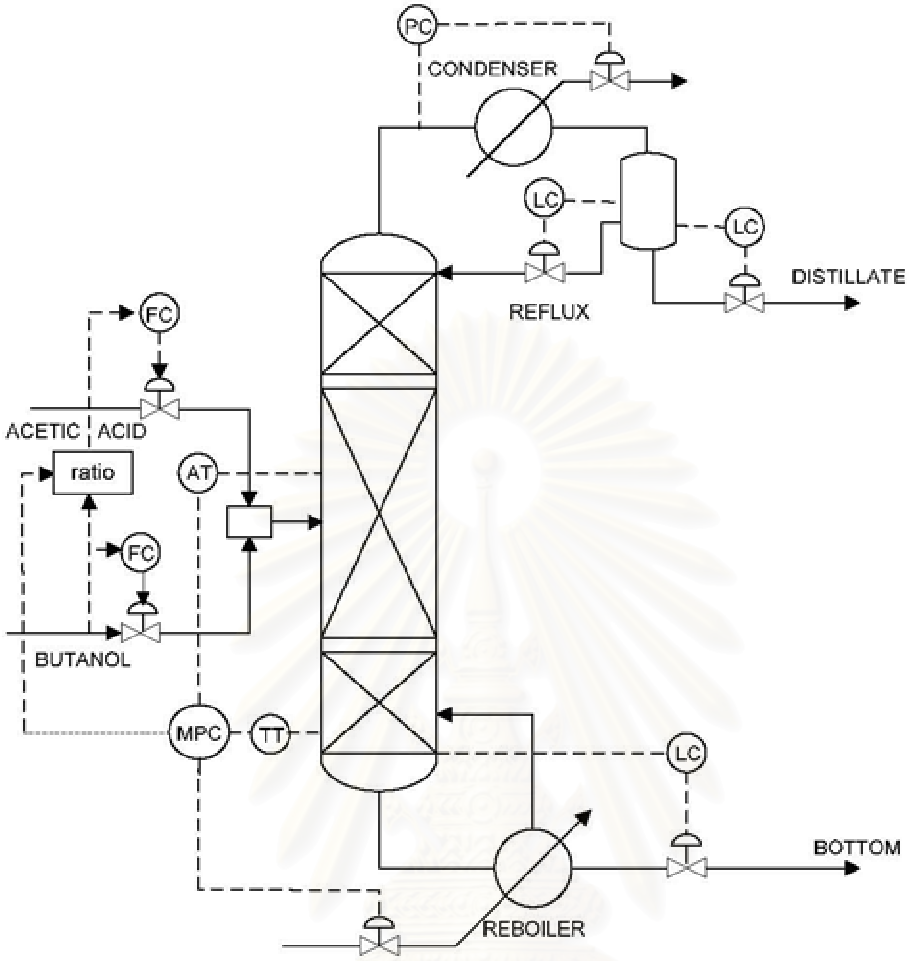


Figure 5-10 Control structure 2 (CS2) of reactive distillation for butyl acetate production (Acetic acid composition at stage 17+ Temperature 37 control)

Therefore, the control objective is to maintain bottom product purity at desired value (≈ 93 % mol of butyl acetate in bottom product) in the face of disturbance changes through proposed control structures. The disturbances considered in this study include changes in butanol feed flow rate and changes in acetic acid feed composition with some water impurity. The first control structure CS1 is to control tray temperature at 17th stage by manipulating the feed ratio of acetic acid to butanol and to control tray temperature at 37th stage by manipulating the reboiler duty. The second control structure CS2 is to control the acetic acid composition at stage 17 by manipulating the feed ratio of acetic acid to butanol and to control tray temperature at 37 th stage by manipulating the reboiler duty. In the control strategy, we assume that the liquid level control loops are perfect controls. Decanter aqueous phase level is held perfectly constant by manipulating the aqueous outlet flow. Organic phase level is held perfectly constant by adjusting the organic reflux flow. The column bottom level is held perfectly constant by changing the bottom product flow.

5.4 MPC controller setup

In this work, we use the model predictive control toolbox box in Matlab for the control of reactive distillation for butyl acetate production. The MPC toolbox requires the model to be linear, time invariant (LTI). Therefore, a simplified process model derived from system identification in the section 5.2 must be converted to linear time-invariant system using methods provided by the Control System Toolbox in Matlab. Once the plant model has been defined, we must specify the characterization of each plant model input and output signal (e.g., manipulated variable and controlled variable) and the characterization of unmeasured and additive disturbances. In the case study we initially accept the defaults of negligible measurement noise and additive integrated white noise on each measured output. Then, we assign the horizons and weights. A value of 1.0 min is used as control interval (sampling time). The prediction horizon is set to 100 samples and the control horizon to 5 samples. There are two manipulated variables which are reboiler duty and feed ratio, and two outputs which are acetic acid composition or temperature at stage 17 and temperature 37 to be controlled at their setpoints. The outputs are equally weighted, while the input weights are left at their defaults, i.e., a small penalty on rate-of-change. Finally, we define input and output constraints. The reboiler duty is bounded between 1.8 and 2.6 Mkcal/h and feed ratio is bounded between 0.9 and 1.1. All outputs are unconstrained.

5.5 Controller performance test

In this section, the performances of the MPC controller for disturbance rejection and set-point tracking of CS1 and CS2 structures are investigated. An efficient control structure should be able to reject unexpected disturbances introduced to the system. Feed composition and feed rate changes are the most probable unpredicted disturbances that affect the reactive distillation column. To examine the MPC controller performance for each control structure, the following disturbances and set-point changes were applied to the reactive distillation column for butyl acetate production:

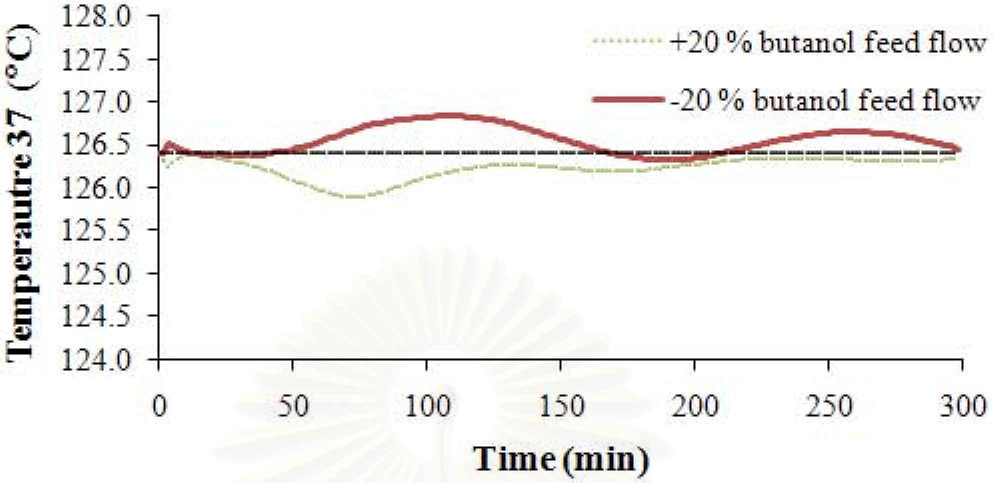
a) ± 20 % step changes in butanol feed flow rate.
b) ± 2 °C step changes in T_{37} set-point.
c) +5 % and +10 % water impurity in the acetic acid feed flow rate

5.51 Performance of CS1 structure

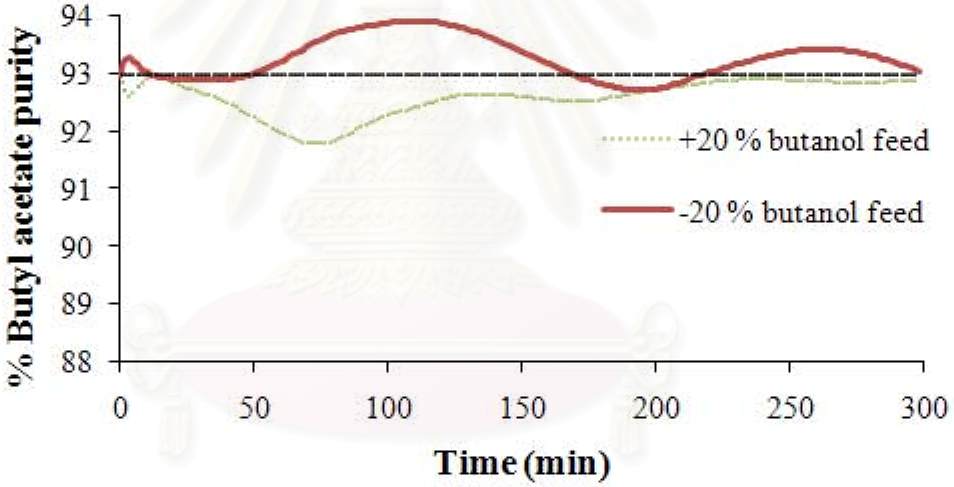
Figure 5-11 shows the responses of the CS1 structure to ± 20 % step changes in butanol feed flow rate. For + 20 % step change in butanol feed flow rate, the temperature 37 and temperature 17 can go back to their setpoints and the purity of butyl acetate product is maintained close to the desired setpoint. The initial responses of temperature 17 and temperature 37 are the same because increasing in butanol feed flow rate produces decreases in the equilibrium temperature on both control trays. The final effects of increasing butanol feed flow rate are increase in reboiler duty and decrease in feed ratio to maintain the temperatures on both control trays at their setpoints. For -20 % step change in butanol feed flow rate, the performance of CS1 structure is not as good as its performance to the step increase. When a negative step change is applied to butanol feed, the tray 17 temperature and the tray 37 temperature start to increase and show an oscillatory response around desired setpoints after time 50 min. Although the response of process is oscillate, butyl acetate product purity remain within 1% of the desired 93 % specification for a 20 % decrease in butanol feed flow rate. The final effects of decreasing butanol feed flow rate are decrease in reboiler duty and increase in feed ratio to maintain the temperatures on both control trays at their setpoints.

Figure 5-12 shows the closed-loop response of the process variables for ± 2 ° C step changes in temperature 37 set-point. It can be seen from Figure 5-12a that the controller is able to track the new set-point for temperature 37 within time 600 min for both an increase and decrease in set-point value. Although there is a small

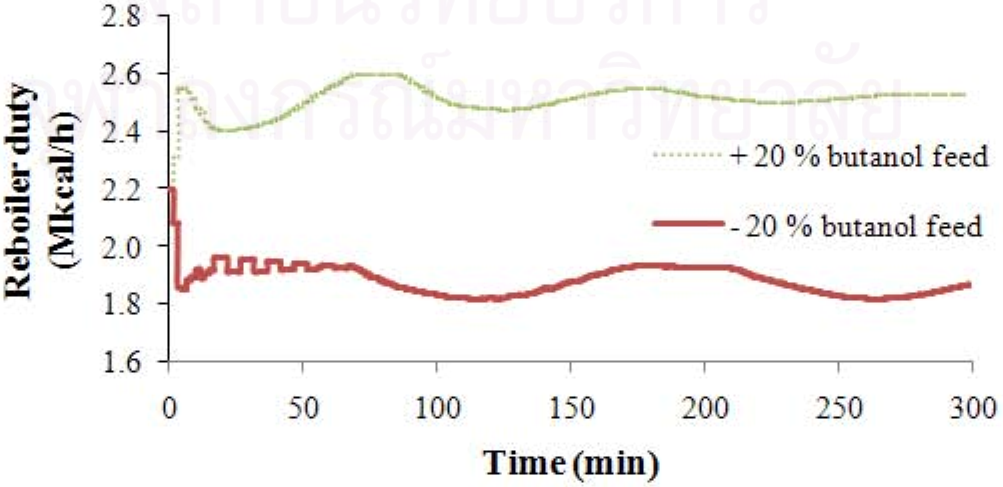
a)


128.0
127.5
127.0
126.5
126.0
125.5
125.0
124.5
124.0
Temperautre 37 (°C)
+20 % butanol feed flow
-20 % butanol feed flow
0
50
100
150
200
250
300
Time (min)

b)


94
93
92
91
90
89
88
% Butyl acetate purity
+20 % butanol feed
-20 % butanol feed
0
50
100
150
200
250
300
Time (min)

c)


2.8
2.6
2.4
2.2
2.0
1.8
1.6
Reboiler duty
(Mkcal/h)
+ 20 % butanol feed
- 20 % butanol feed
0
50
100
150
200
250
300
Time (min)

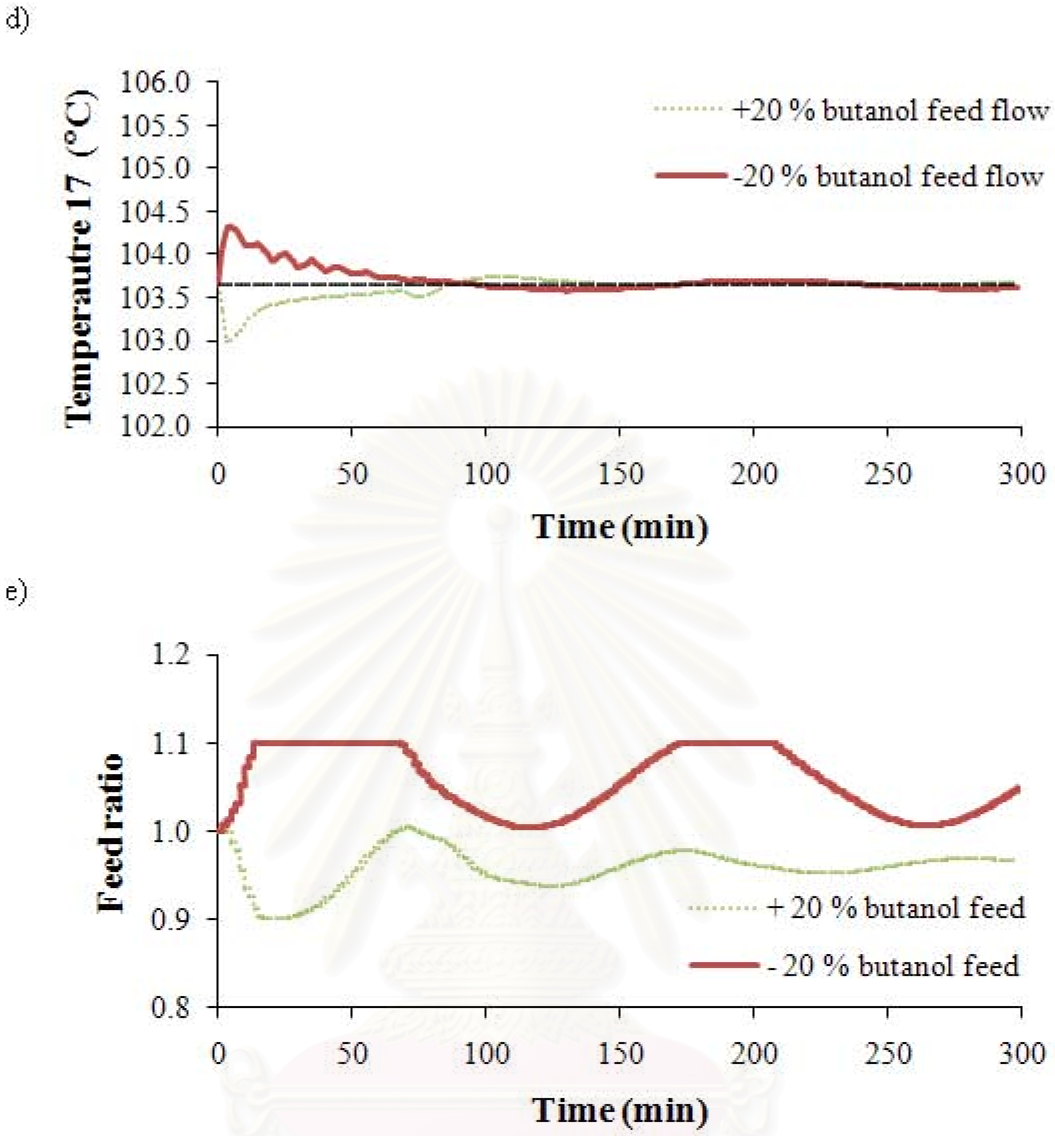
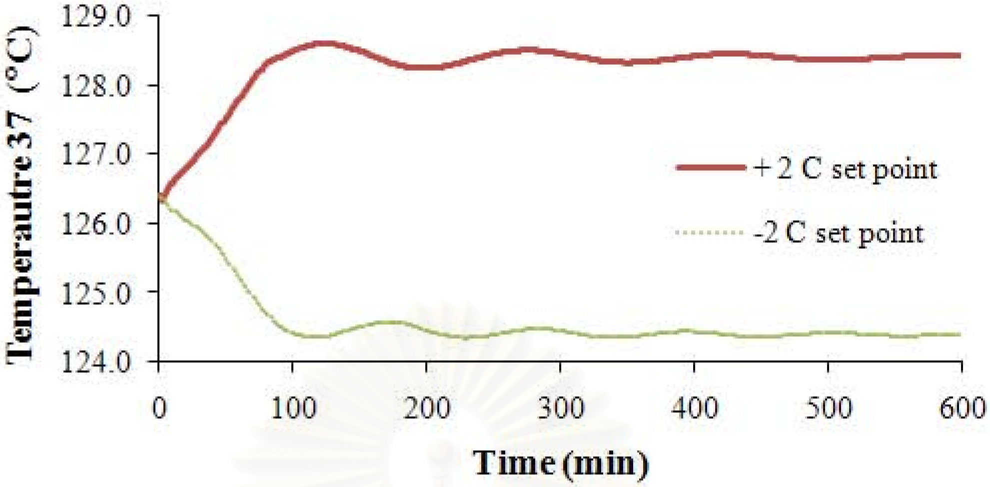


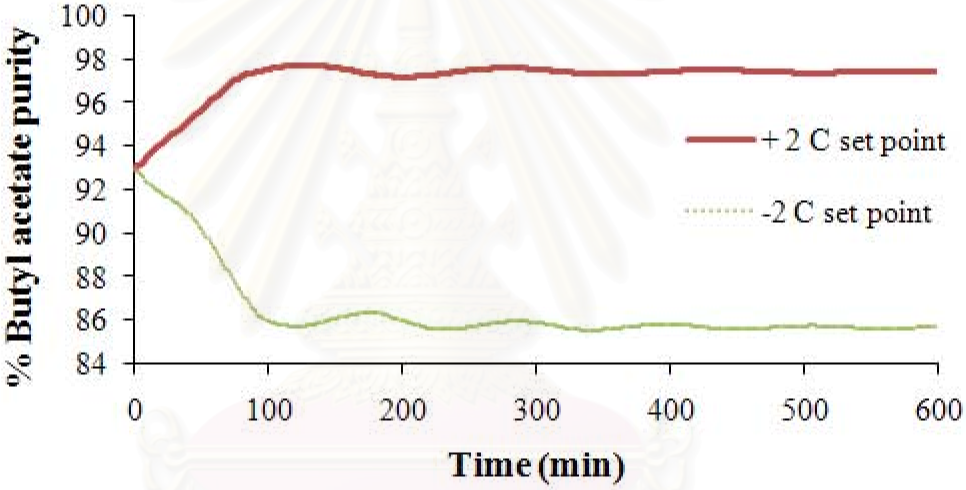
Figure 5-11 Closed-loop responses of MPC controller for ± 20 % step changes in butanol feed flow rate (a) stage 37 temperature (b) % butyl acetate purity (c) reboiler duty (d) stage 17 temperature (e) feed ratio

oscillation in controlled variable, the temperature 37 can reach the new set-point value without any erratic deviation in controlled variable. From Figure 5-12b, it can be seen that bottom product butyl acetate purity increase and decrease with an increase and decrease in temperature 37 set-point. Therefore, the temperature 37 can be used as a single adjusting process variable for slight increase or decrease in bottom product butyl acetate purity. Figure 5-12c,e shows that the controller action adjust reboiler duty and feed ratio for increase and decrease in the temperature 37 set-point,

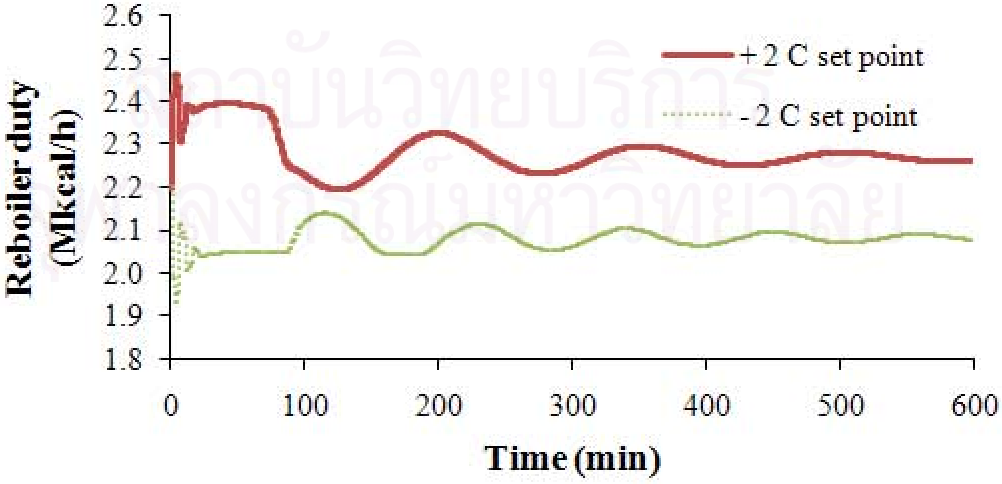
a)


Temperautre 37 (°C)
129.0
128.0
127.0
126.0
125.0
124.0
0
100
200
300
400
500
600
Time (min)
+ 2 C set point
-2 C set point

b)


% Butyl acetate purity
100
98
96
94
92
90
88
86
84
0
100
200
300
400
500
600
Time (min)
+ 2 C set point
-2 C set point

c)


Reboiler duty (Mkcal/h)
2.6
2.5
2.4
2.3
2.2
2.1
2.0
1.9
1.8
0
100
200
300
400
500
600
Time (min)
+ 2 C set point
- 2 C set point

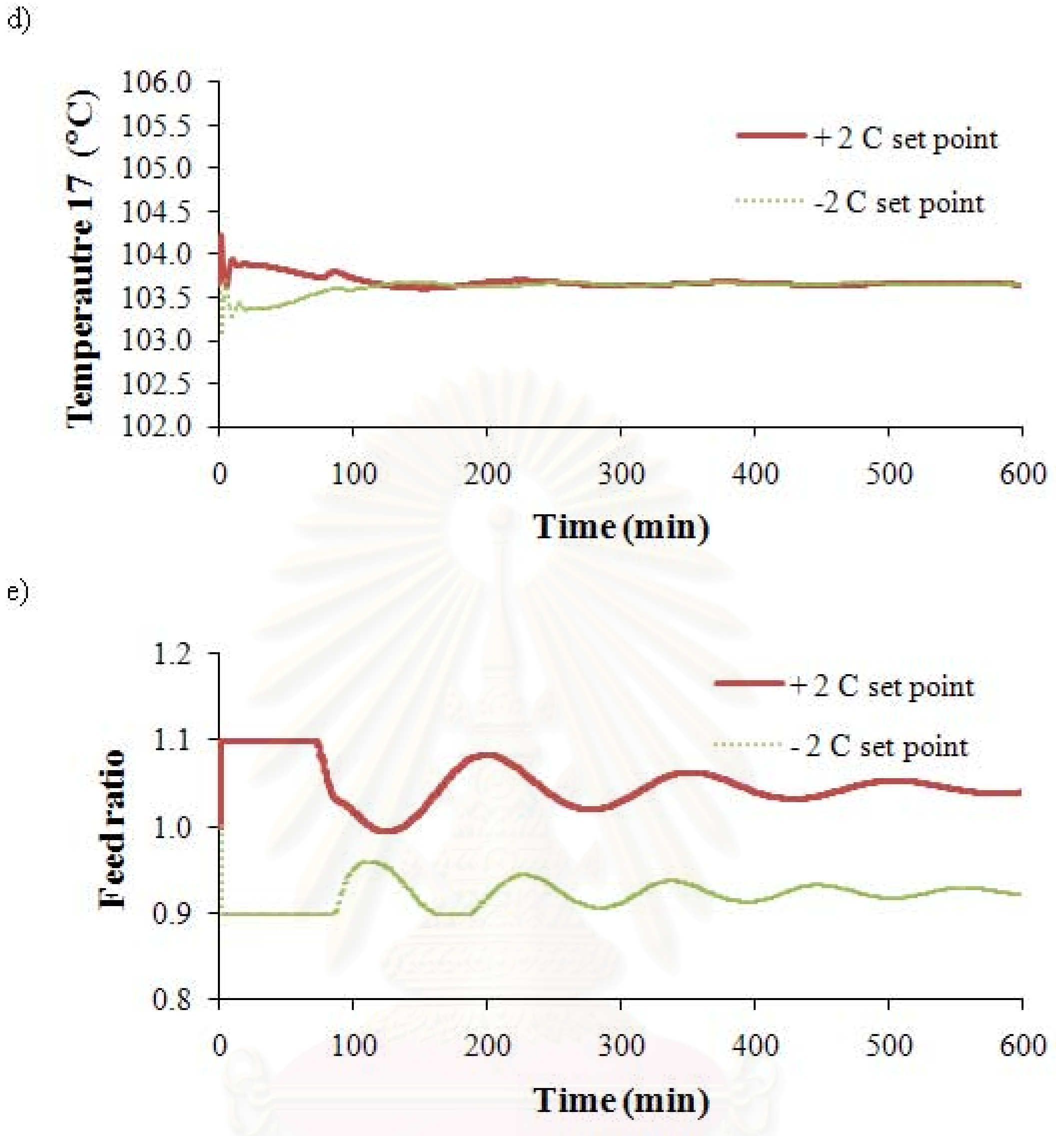


Figure 5-12 Closed-loop responses of MPC controller for ± 2 °C step changes in temperature 37 (a) stage 37 temperature (b) % butyl acetate purity (c) reboiler duty (d) stage 17 temperature (e) feed ratio

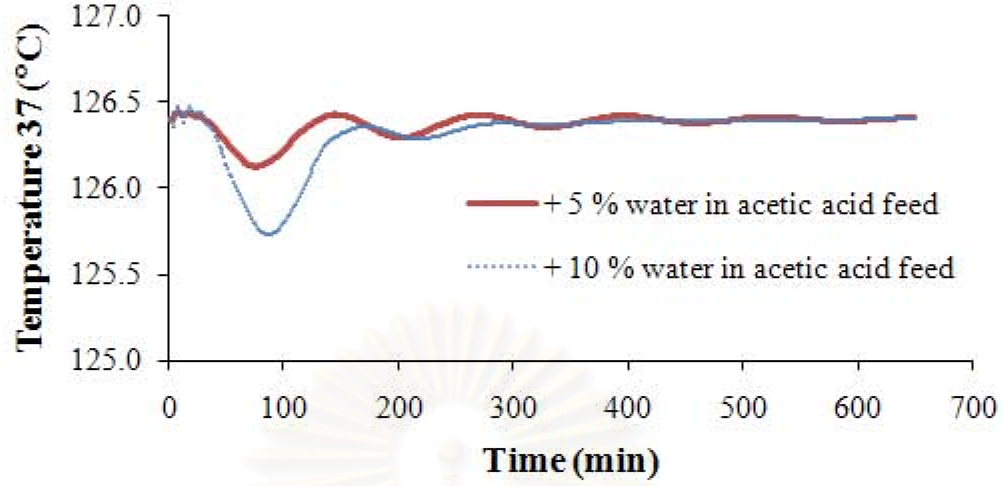
respectively. Figure 5-12d shows the effect of temperature 37 set-point change on the temperature 17. The controller is able to reject the disturbance caused by temperature 37 set-point change and prevent a large deviation in temperature 17 from its set-point value and return it to its set-point within 500 min.

The closed-loop control performances with disturbances of the acetic acid feed composition changes are shown in Figure 5-13. The acetic acid feed composition is changed from pure acetic acid to 90 and 95 mol %. The impurity of the acetic acid feed stream is water. From the Figure 5-13a and Figure 5-13d, the temperature 37 and temperature 17 can be controlled back to their setpoint values within 600 min. Because the impurity of acetic acid feed is increased, this means that the actual key reactant into the system is decreased. Therefore, MPC controller will increase of the acetic acid to butanol feed ratio cope with this disturbance change shown in Figure 5-13e. Similarly, when water in the system is increased, MPC controller will increase of the reboiler heat duty to vaporize additional water to the top product or recycle unreacted reactant to the reaction zone shown in Figure5-13c. Since there is no direct bottom product composition control loop, the bottom butyl acetate purity is deviated from its setpoint shown in Figure5-13b. However, this butyl acetate purity is still pure enough to meet our original control objective.

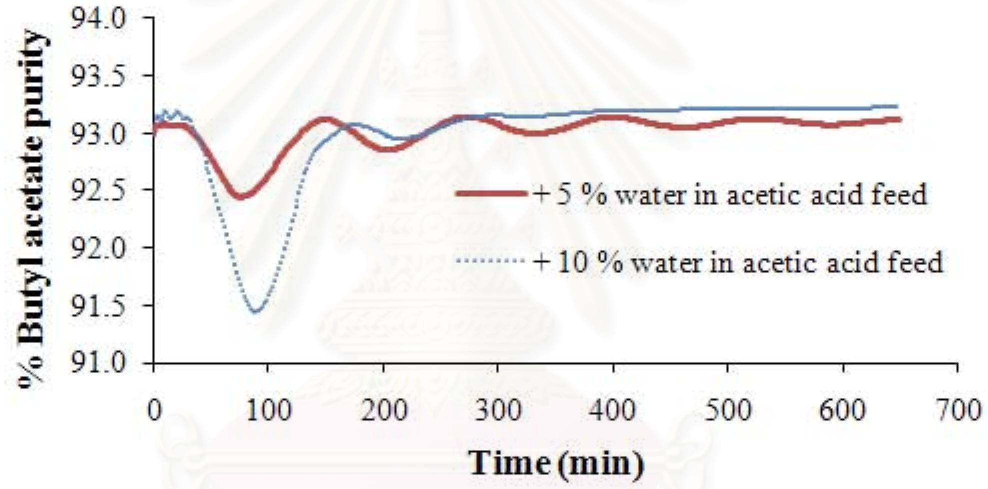
5.5.2 Performance of CS2 structure

The close-loop dynamic responses of the CS2 structure to ± 20 % step changes in butanol feed flow rate are given in Figure 5-14. Results show that a 20 % increase in butanol feed flow rate can be handled without any oscillation, and MPC controller is able to maintain the temperature 37 and acetic acid composition at stage 17 close to their set-points and reject the introduced disturbances within 300 min. Because there is no direct bottom product composition control loop, the butyl acetate product purity is deviated from their original steady-state values before the disturbance was applied, but this small deviation of the bottom product purity is considered to be acceptable. To eliminate this difference, the temperature 37 set-point can be manually adjusted to the desired product purity. For both step increase and decrease in butanol feed flow rate, it should be noted that the CS2 structure has smoother and faster response to this disturbance than CS1 structure has. Similarly, CS2 structure has smoother adjusting of acetic acid to butanol feed ratio and reboiler heat duty than CS1 structure. It should be noted that the direct control of acetic acid composition at stage 17 of CS2 structure instead of the inferential control of temperature 17 of CS1 structure can give better performance in case of ± 20 % step changes in butanol feed flow rate. When there are changes in the butanol feed, the acetic acid compoisiton at stage 17 will be affected directly with changes in the butanol feed, this make controller can see the changes of

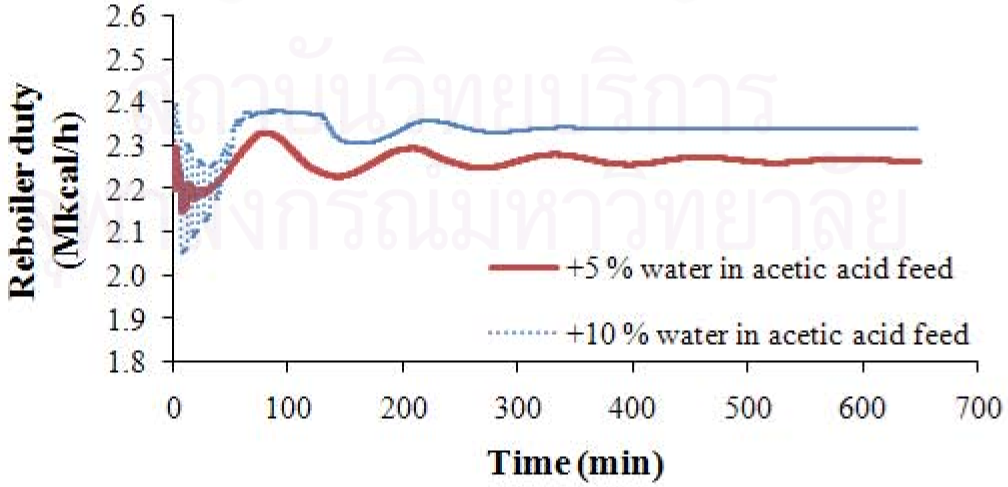
a)


Temperature 37 (°C)
127.0
126.5
126.0
125.5
125.0
0
100
200
300
400
500
600
700
Time (min)
+ 5 % water in acetic acid feed
+ 10 % water in acetic acid feed

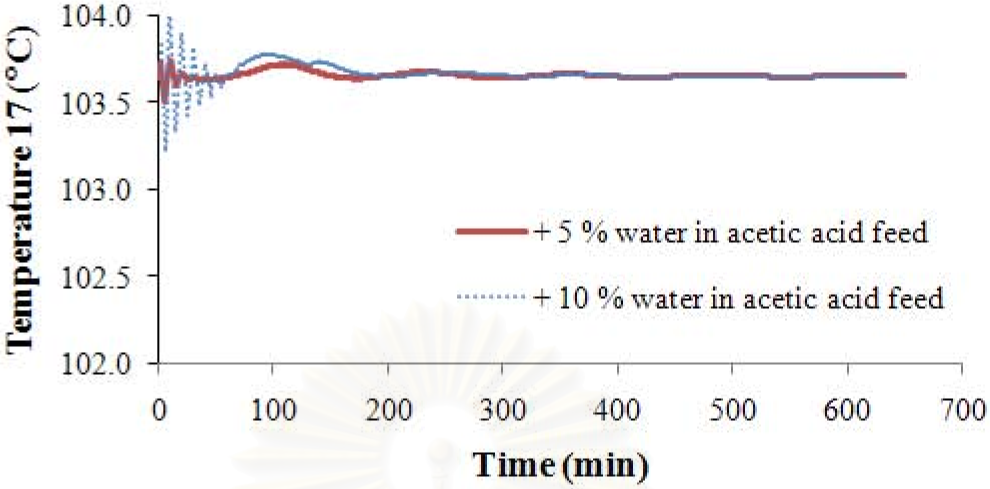
b)


% Butyl acetate purity
94.0
93.5
93.0
92.5
92.0
91.5
91.0
0
100
200
300
400
500
600
700
Time (min)
+ 5 % water in acetic acid feed
+ 10 % water in acetic acid feed

c)


Reboiler duty (Mkcal/h)
2.6
2.5
2.4
2.3
2.2
2.1
2.0
1.9
1.8
0
100
200
300
400
500
600
700
Time (min)
+5 % water in acetic acid feed
+10 % water in acetic acid feed

d)



e)

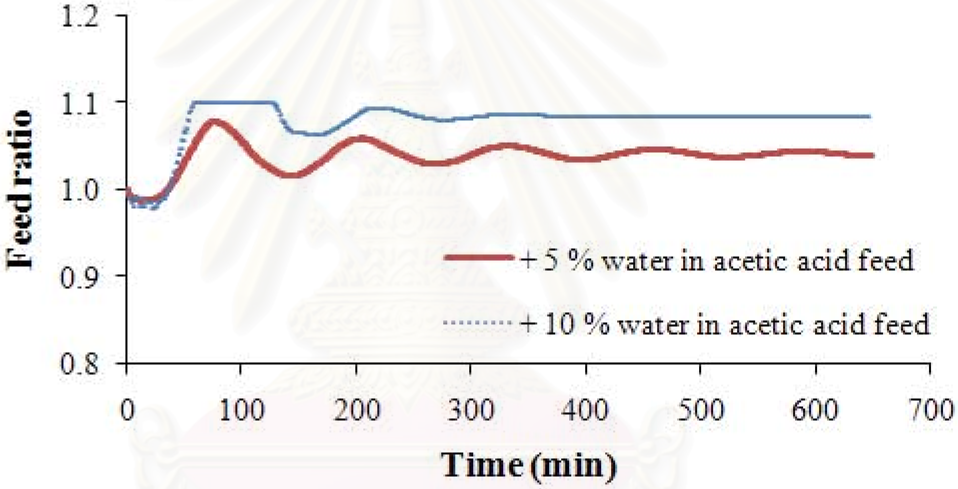
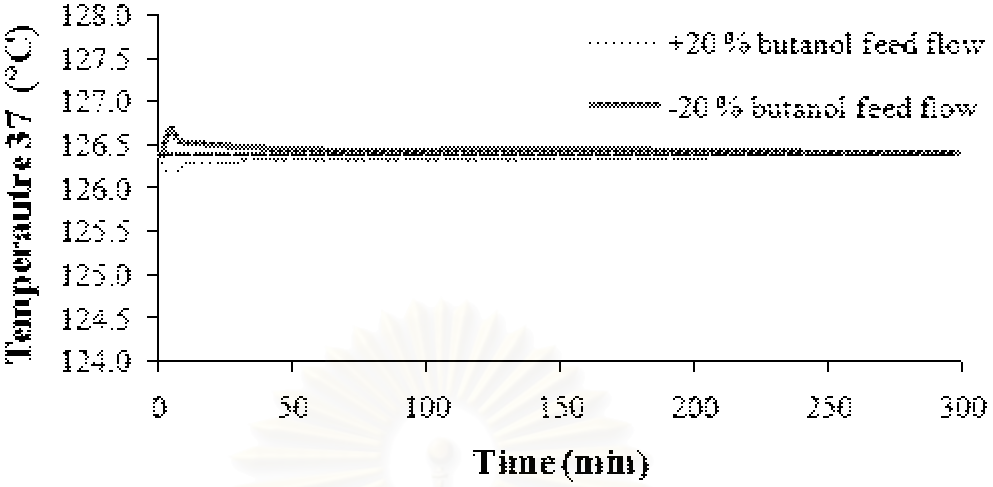


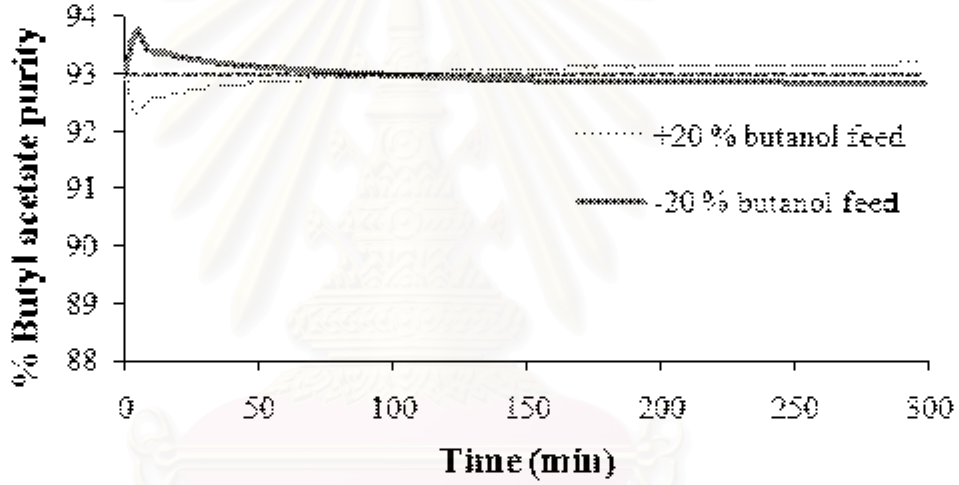
Figure 5-13 Closed-loop responses of MPC controller for 5 % and 10 % water impurity step changes in acetic acid feed (a) stage 37 temperature (b) % butyl acetate purity (c) reboiler duty (d) stage 17 temperature (e) feed ratio

acetic acid composition at stage 17 and adjust feed ratio to maintain acetic acid composition at stage 17 accurately. For inferential control of temperature 17 of CS1 structure, the temperature 17 is not depend on the changes in acetic acid compoisiton at stage 17 only, but it depends on the changes of compositions of all components in the stage 17.

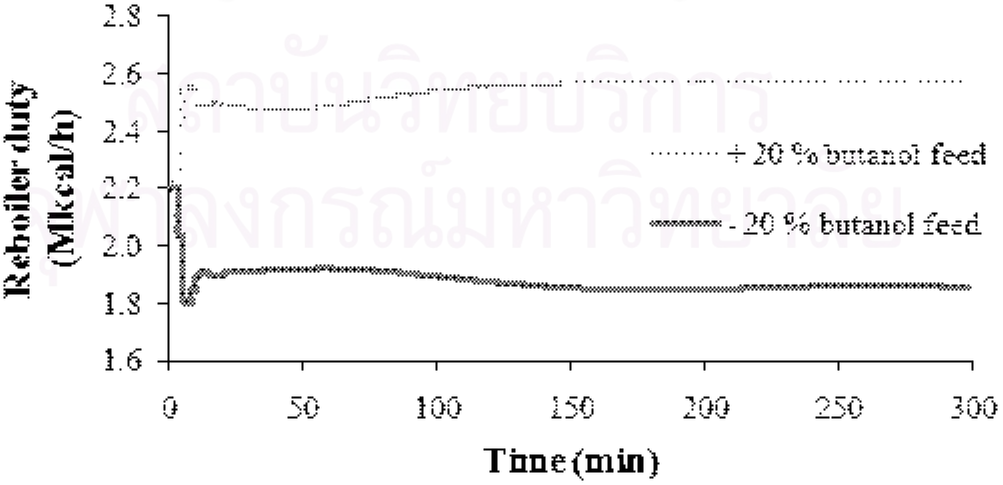
a)


Temperautre 37 (°C)
128.0
127.5
127.0
126.5
126.0
125.5
125.0
124.5
124.0
0
50
100
150
200
250
300
Time (min)
+20 % butanol feed flow
-20 % butanol feed flow

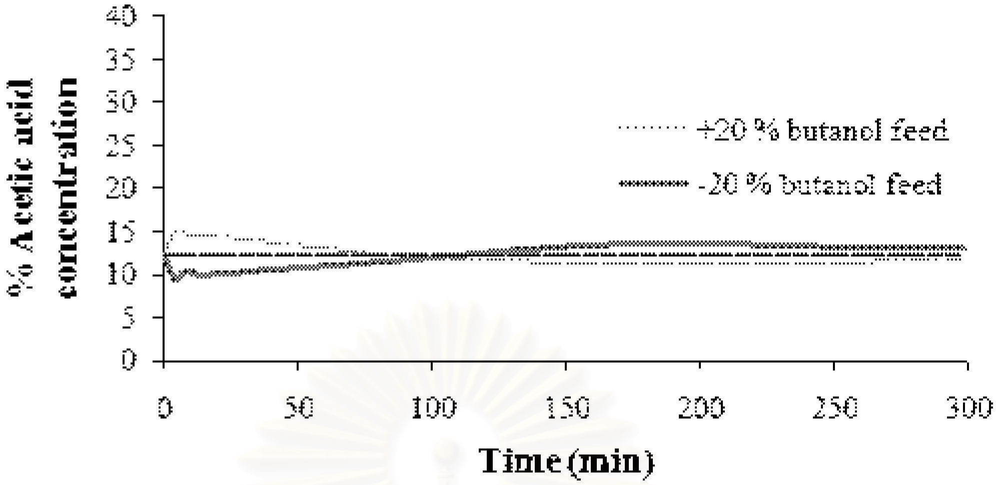
b)


% Butyl acetate purity
94
93
92
91
90
89
88
0
50
100
150
200
250
300
Time (min)
+20 % butanol feed
-20 % butanol feed

c)


Reboiler duty (Mkcal/h)
2.8
2.6
2.4
2.2
2.0
1.8
1.6
0
50
100
150
200
250
300
Time (min)
+ 20 % butanol feed
- 20 % butanol feed

d)



e)

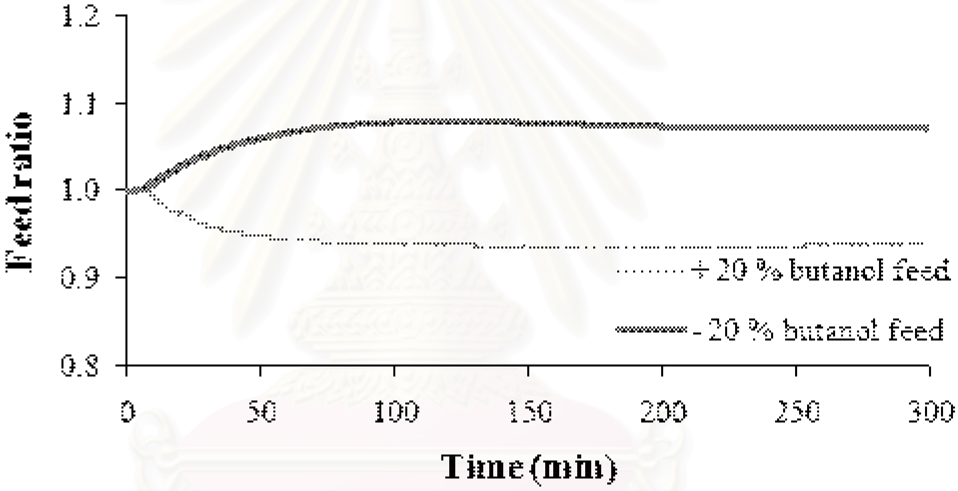
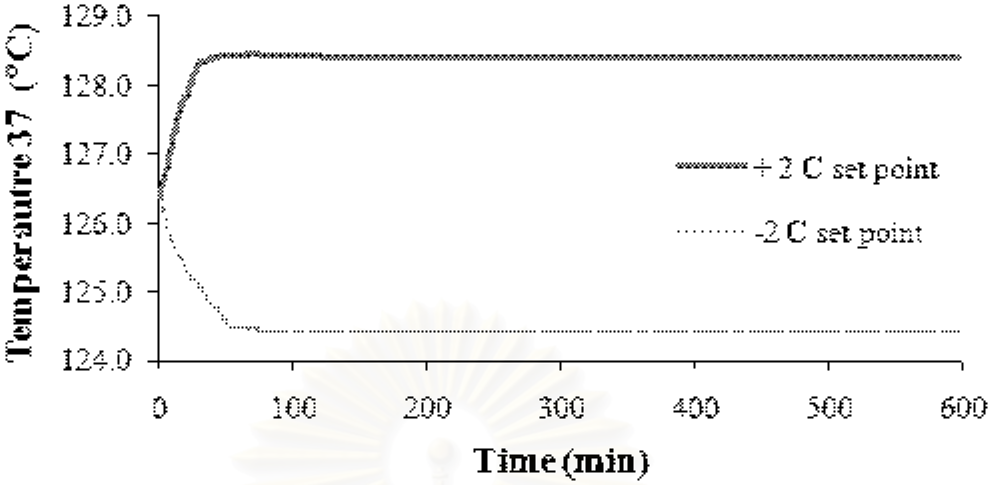


Figure 5-14 Closed-loop responses of MPC controller for ± 20 % step changes in butanol feed flow rate (a) stage 37 temperature (b) % butyl acetate purity (c) reboiler duty (d) stage 17 temperature (e) feed ratio

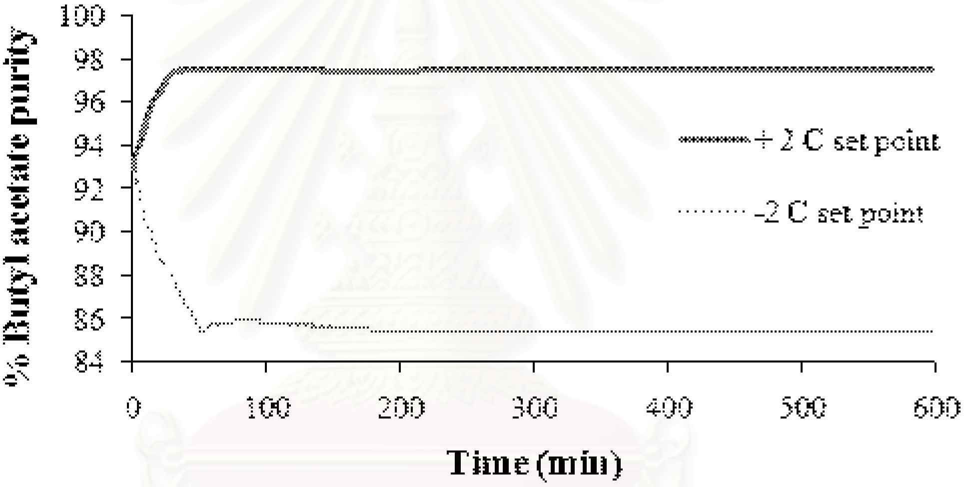
Although the controller can adjust feed ratio to maintain temperature 17 at desired setpoint, it doesn't guarantee keeping in correct stoichiometric ratio of acetic acid and butanol durng operation of a reactive distillation. Therefore, the responses of CS1 structure are not as good as CS2 structure.

Figure 5-15 shows the closed-loop response of CS2 structure for ± 2 ° C step changes in temperature 37 set-point. It can be seen from Figure 5-15a that MPC

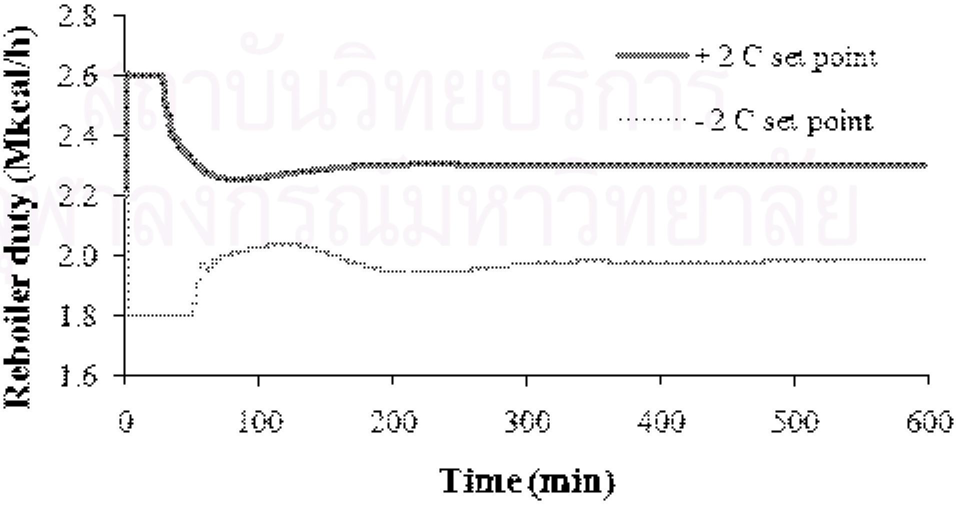
a)


Temperautre 37 (°C)
129.0
128.0
127.0
126.0
125.0
124.0
0
100
200
300
400
500
600
Time (min)
+ 2 C set point
-2 C set point

b)


% Butyl acetate purity
100
98
96
94
92
90
88
86
84
0
100
200
300
400
500
600
Time (min)
+ 2 C set point
-2 C set point

c)


Reboiler duty (Mkcal/h)
2.8
2.6
2.4
2.2
2.0
1.8
1.6
0
100
200
300
400
500
600
Time (min)
+ 2 C set point
- 2 C set point

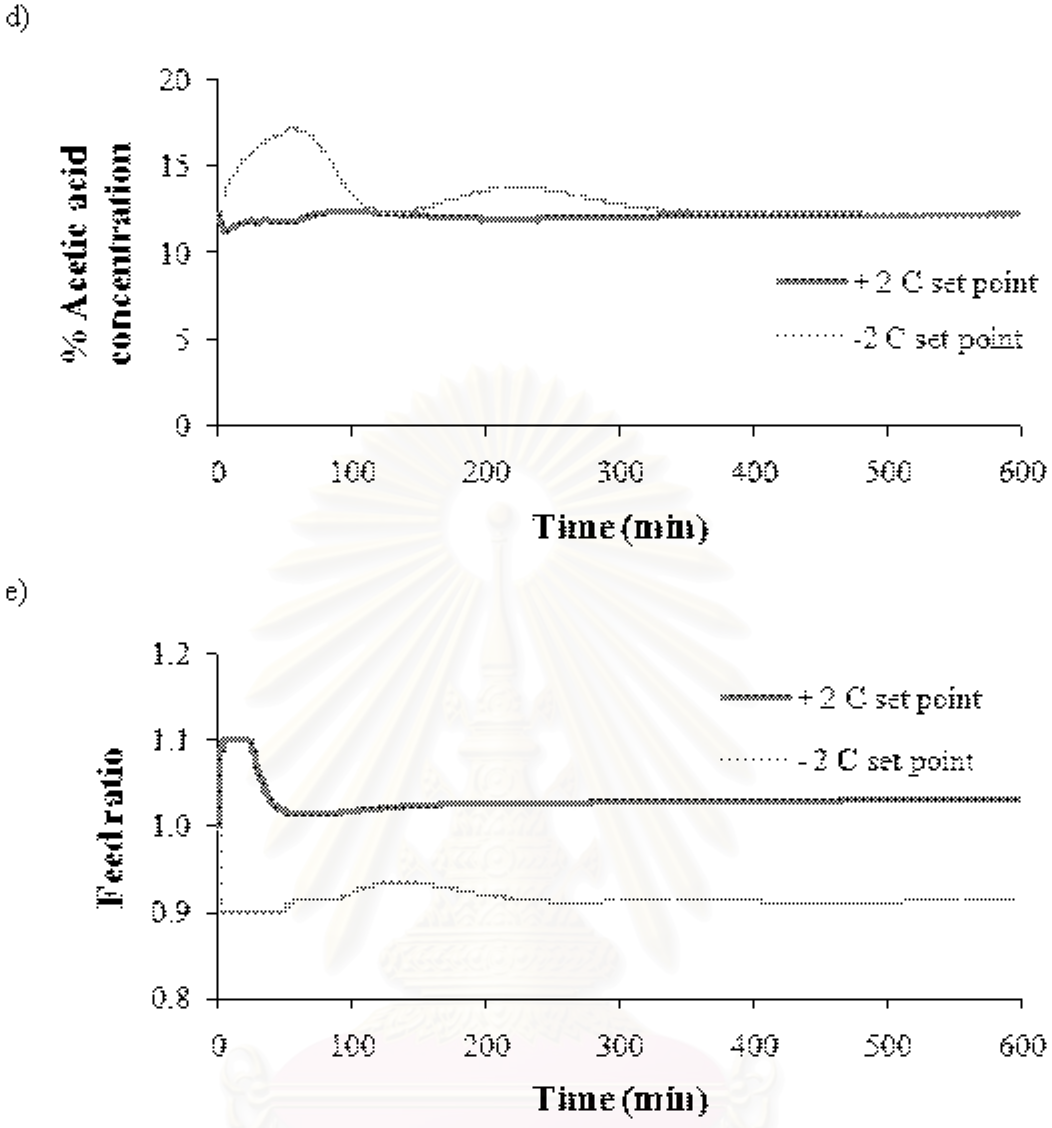
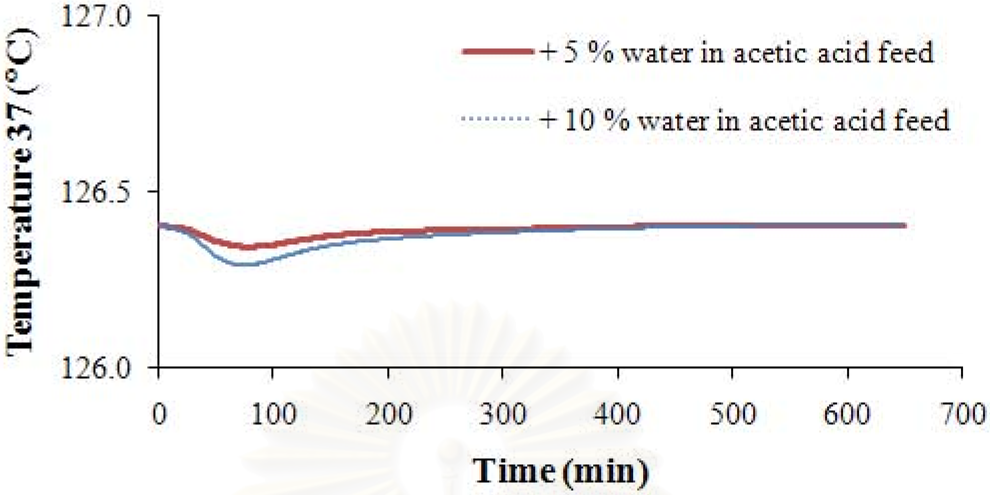


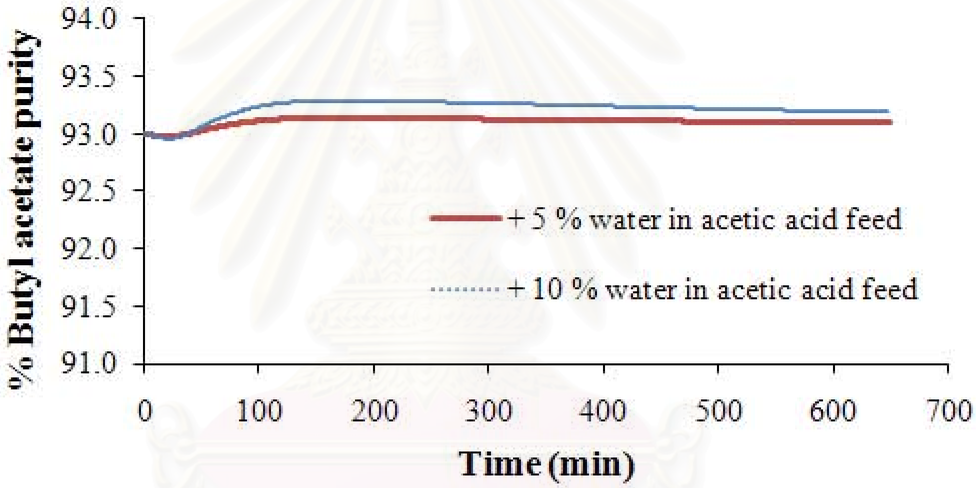
Figure 5-15 Closed-loop responses of MPC controller for ± 2 °C step changes in temperature 37 (a) stage 37 temperature (b) % butyl acetate purity (c) reboiler duty (d) stage 17 temperature (e) feed ratio

controller is able to track the new set-points of temperature 37 quickly within time 300 min for both an increase and decrease in set-point value. Figure 5-15b reveals that the butyl acetate bottom product purity increases with a increase in temperature 37 set-point and vice versa. Figure 5-15d shows the controller reacts quickly to the effect of temperature 37 set-point change on acetic acid composition at stage 17. For set-point changes, it should be also noted that the CS2 structure has smoother and

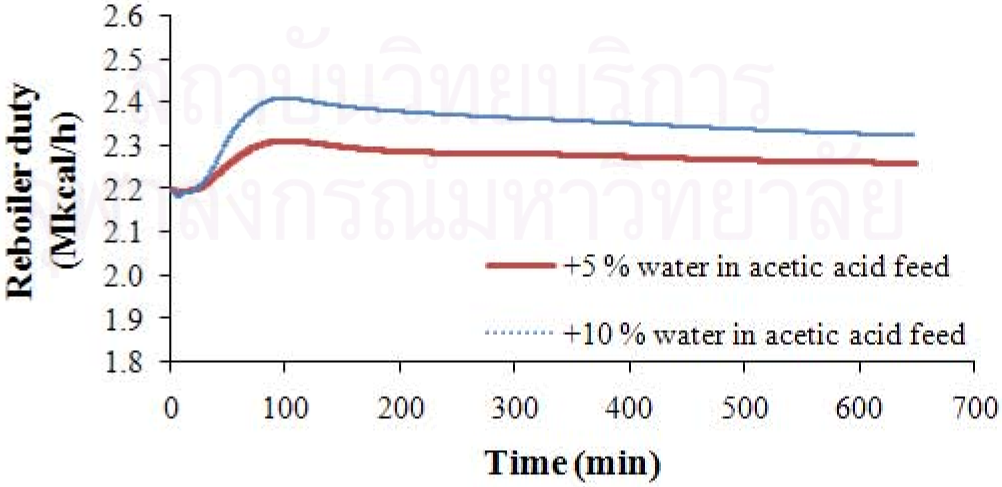
a)


Temperature 37 (°C)
127.0
126.5
126.0
0
100
200
300
400
500
600
700
Time (min)
+ 5 % water in acetic acid feed
+ 10 % water in acetic acid feed

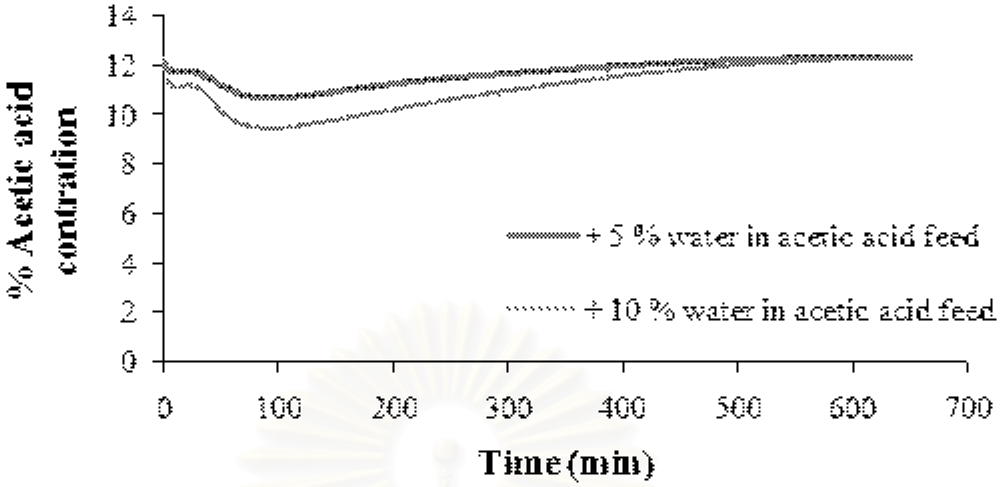
b)


% Butyl acetate purity
94.0
93.5
93.0
92.5
92.0
91.5
91.0
0
100
200
300
400
500
600
700
Time (min)
+ 5 % water in acetic acid feed
+ 10 % water in acetic acid feed

c)


Reboiler duty (Mkcal/h)
2.6
2.5
2.4
2.3
2.2
2.1
2.0
1.9
1.8
0
100
200
300
400
500
600
700
Time (min)
+5 % water in acetic acid feed
+10 % water in acetic acid feed

d)



e)

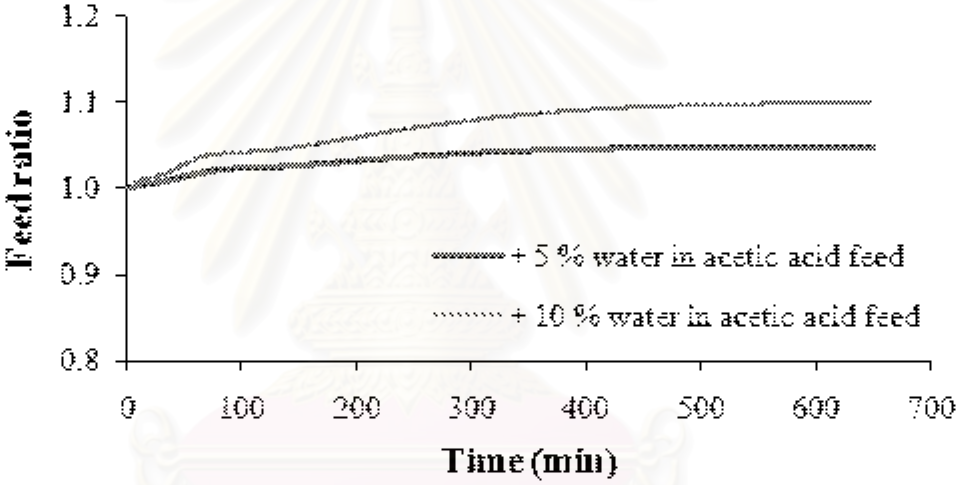


Figure 5-16 Closed-loop responses of MPC controller for 5 % and 10 % water impurity step changes in acetic acid feed (a) stage 37 temperature (b) % butyl acetate purity (c) reboiler duty (d) stage 17 temperature (e) feed ratio

faster response to set-point change than CS1 structure. Similarly, CS2 structure has smoother adjusting of acetic acid to butanol feed ratio and reboiler heat duty than CS1 structure.

The same disturbances for the acetic acid feed composition changes used in CS1 structure are also tested for CS2 structure. From Figure 5-16, one observes that with the acetic acid feed compoisition drops to 90 and 95 mol %, the CS2 structure is able

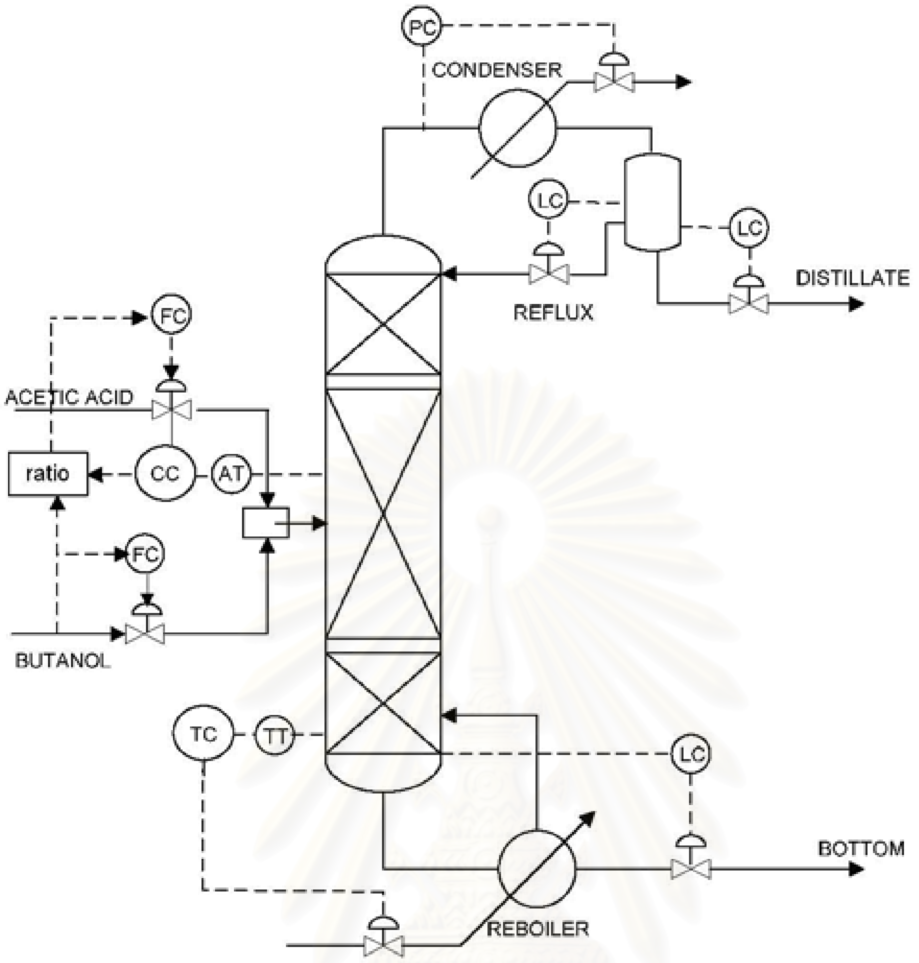


Figure 5-17 Two-point PI control configuration of reactive distillation for butyl acetate production

Table 5-2 Comparison of IAE indice

	MPC		PI	
Magnitude	X17	T37	X17	T37
+ 2 C step in T37	0.36	32.09	0.27	47.64
-2 C step in T37	3.43	48.37	3.70	61.21
+20 % step in Fb	1.18	8.80	0.25	11.32
-20 % step in Fb	1.44	6.93	0.36	13.31
+10 % water in Fa	1.97	6.81	0.31	5.25

to handle to disturbances effectively. Temperature 37 and acetic acid composition at stage 17 can return to their setpoints quickly within 600 min. Figure 5-16b shows that the butyl acetate product purity can reach the steady-state value quickly within 600 min;however, the butyl acetate product purity at the new steady-state value is different from its initially desired value. Although there is deviation of butyl acetate product purity from its desired setpoint, it is still in acceptable range. From the MPC controller performance test for CS1 and CS2 structure, it can be seen clearly that CS2 structure can make the column more quickly operate at a correct stoichiometric ratio between feeds when there are disturbances in the system. Furthermore, the CS2 structure has faster and smoother response when compared with the response in CS1 structure. Thus, we can conclude that the CS2 structure can give better performance than the CS1 structure.

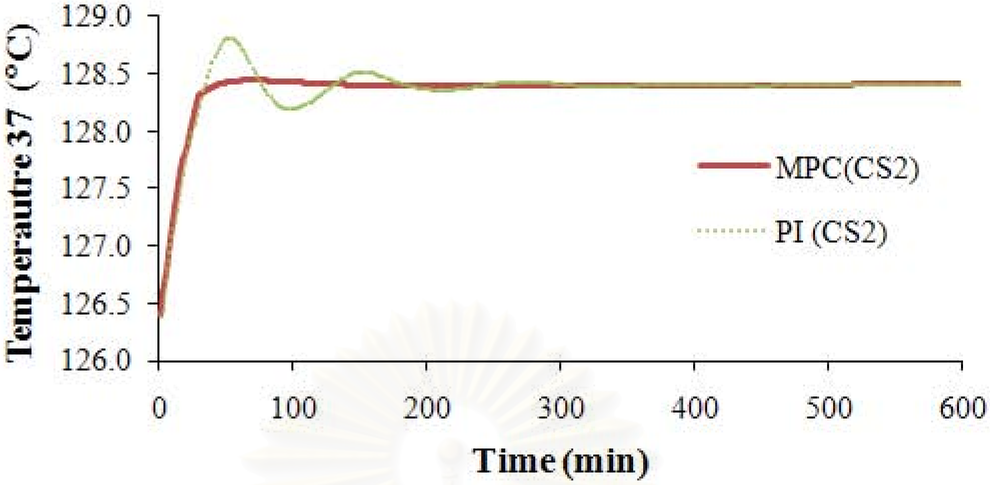
5.6 Comparison of PI and MPC controllers.

The previous control results of MPC controller for two alternative control structures indicated that CS2 structure provided good control performance for setpoint tracking as well as the rejection of various disturbances. Therefore, in this section, the performance of MPC controller is evaluated and compared to that of a conventional PI controller using the CS2 structure. The control structure for 2x2 PI controllers is shown in Figure 5-17. In this 2x2 PI control system, the acetic acid compoisition at stage 17 is controlled using the feed ratio, and the stage 37 temperature is controlled using the reboiler heat duty. The performances of both controllers are evaluated for changes in the set-point (tracking problem) as well as in the feed rate and feed composition (regulation problem).The two PI controllers, which are used to control the temperature 37 by manipulating the reboiler duty and used to control acitic acid composition at stage 17 by manipulating feed ratio for reactive distillation process, are written as

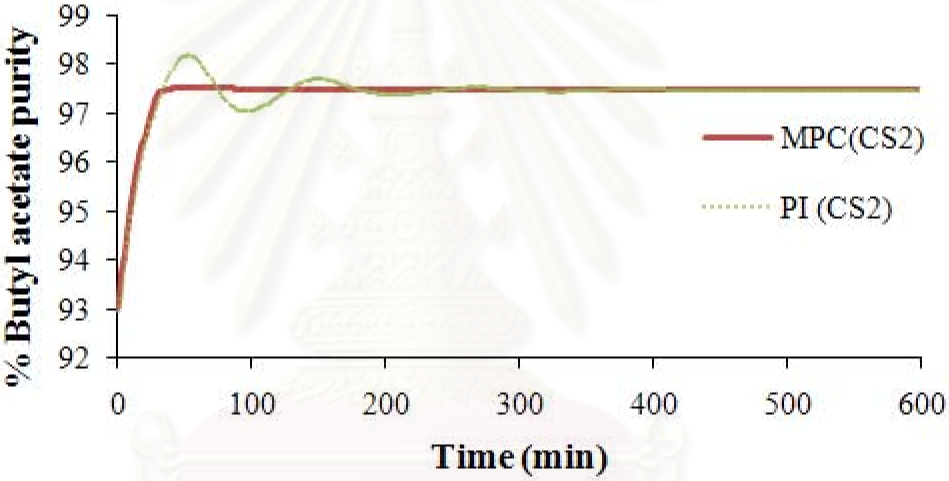
$$Q(t) = Q(t - T_s) + K_{P1}(e_1(t) - e_1(t - T_s) + \frac{e_1(t)T_s}{\tau_{I1}}) \text{ and}$$

$$Fr(t) = Fr(t - T_s) + K_{P2}(e_2(t) - e_2(t - T_s) + \frac{e_2(t)T_s}{\tau_{I2}})$$

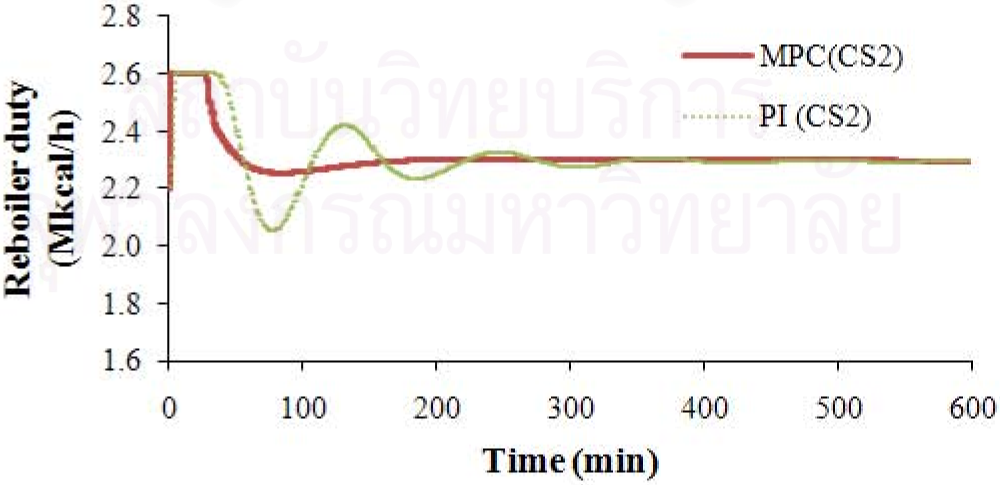
a)


Temperautre 37 (°C)
129.0
128.5
128.0
127.5
127.0
126.5
126.0
0
100
200
300
400
500
600
Time (min)
MPC(CS2)
PI (CS2)

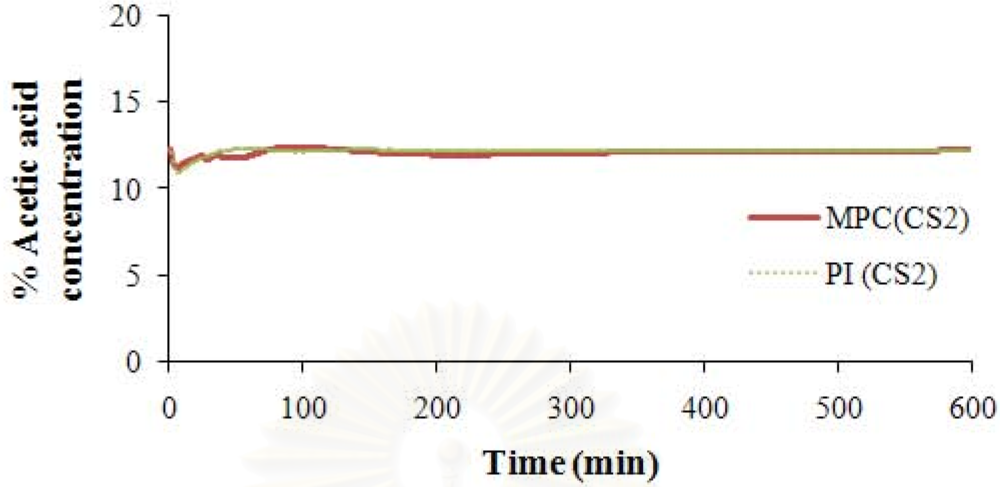
b)


% Butyl acetate purity
99
98
97
96
95
94
93
92
0
100
200
300
400
500
600
Time (min)
MPC(CS2)
PI (CS2)

c)


Reboiler duty (Mkcal/h)
2.8
2.6
2.4
2.2
2.0
1.8
1.6
0
100
200
300
400
500
600
Time (min)
MPC(CS2)
PI (CS2)

d)



e)

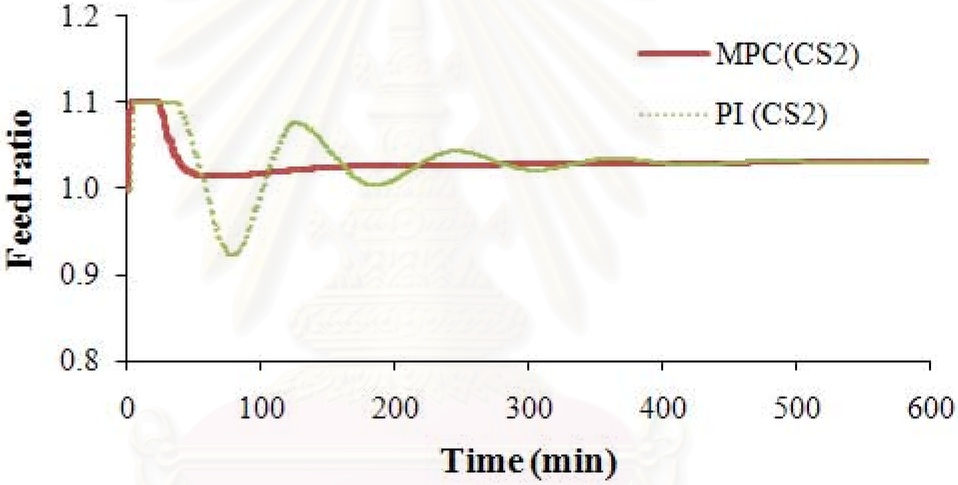
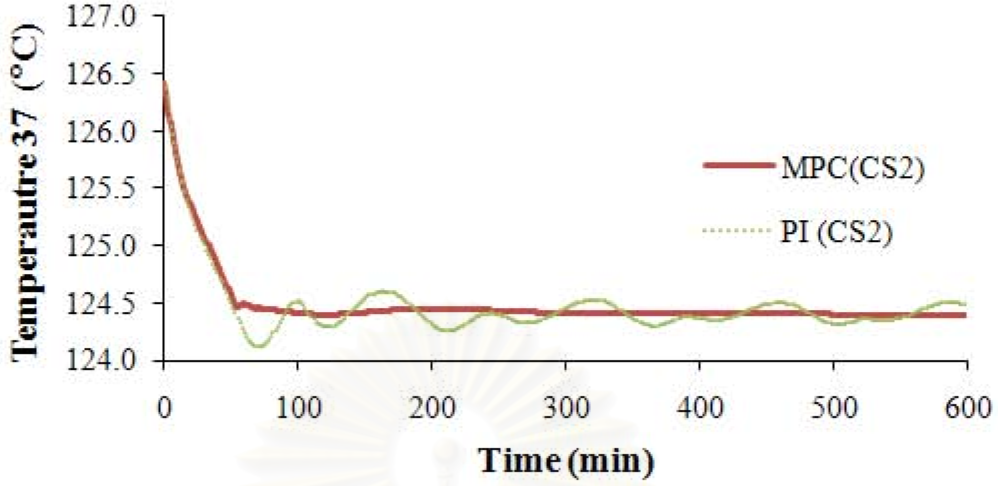


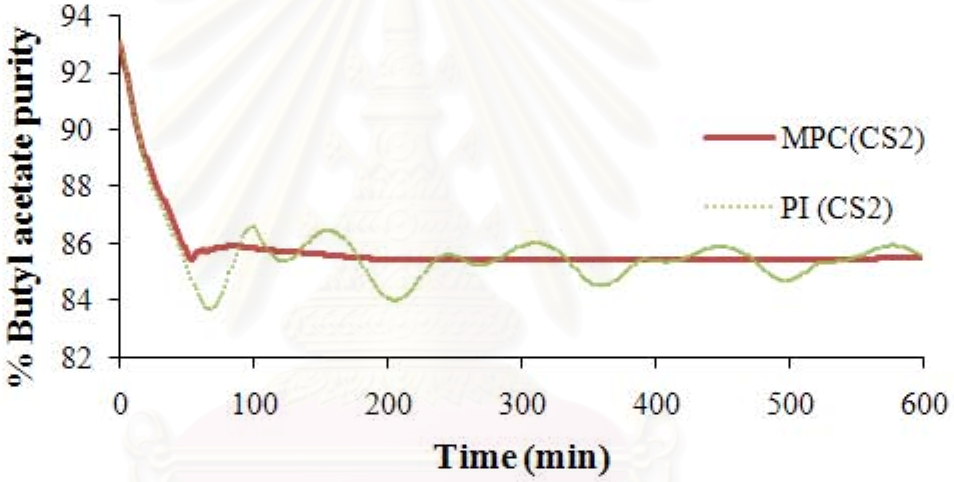
Figure 5-18 Comparison of MPC and PI control, closed-loop responses for +2 °C set point in temperature 37 (a) stage 37 temperature (b) % butyl acetate purity (c) reboiler duty (d) % acetic acid concentration at stage 17 (e) feed ratio

where $e_1 = T_{37sp} - T_{37}(t)$ and $e_2 = x_{17sp} - x_{17}(t)$. The tuning of the two PI controllers parameters has been performed using integral absolute error IAE performance criteria and the optimal values of these parameters are: $K_{P1} = 1.162$ and $\tau_{I1} = 0.12$ min and $K_{P2} = 0.046$ and $\tau_{I2} = 0.51$ min. For both the MPC controller and two PI controllers, the reboiler duty and feed ratio which are manipulated variables are bounded between $1.8 \le Q \le 2.6$ Mkcal/h and $0.9 \le Fr \le 1.1$, respectively.

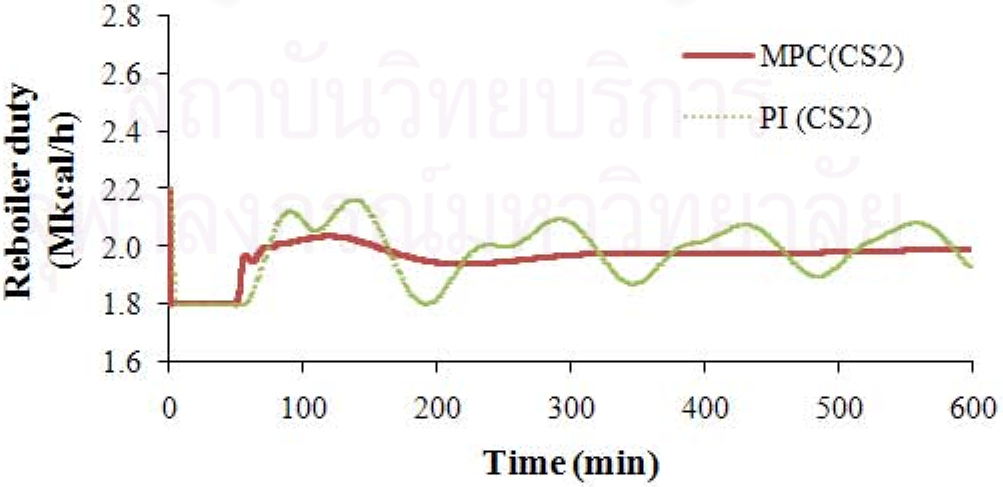
a)


Temperautre 37 (°C)
127.0
126.5
126.0
125.5
125.0
124.5
124.0
0
100
200
300
400
500
600
Time (min)
MPC(CS2)
PI (CS2)

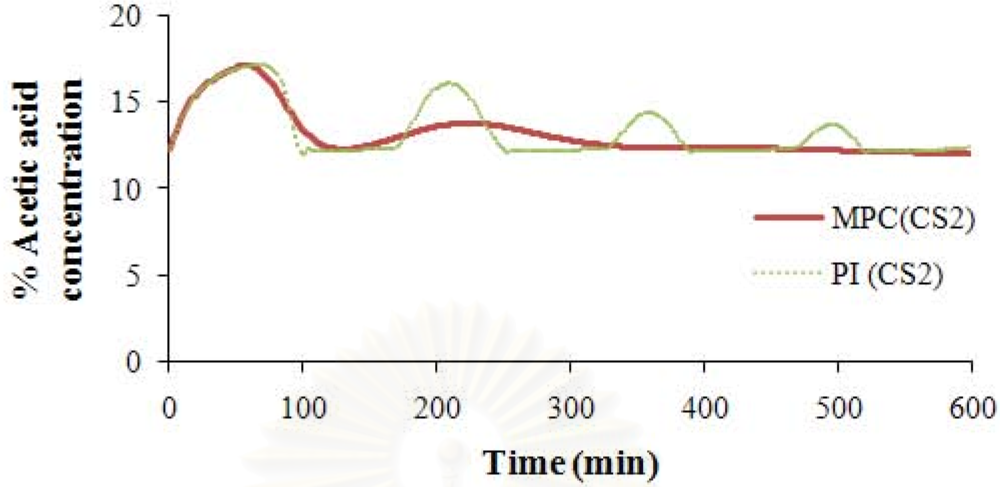
b)


% Butyl acetate purity
94
92
90
88
86
84
82
0
100
200
300
400
500
600
Time (min)
MPC(CS2)
PI (CS2)

c)


Reboiler duty (Mkcal/h)
2.8
2.6
2.4
2.2
2.0
1.8
1.6
0
100
200
300
400
500
600
Time (min)
MPC(CS2)
PI (CS2)

d)



e)

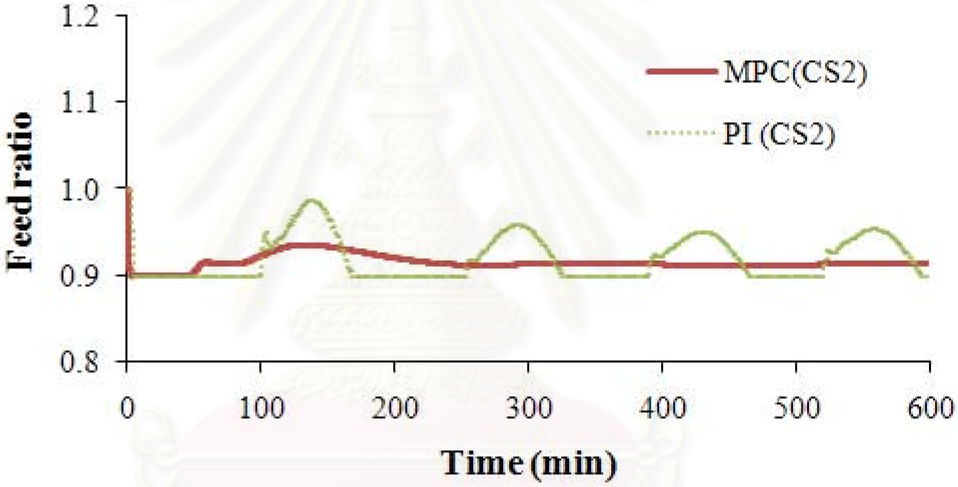
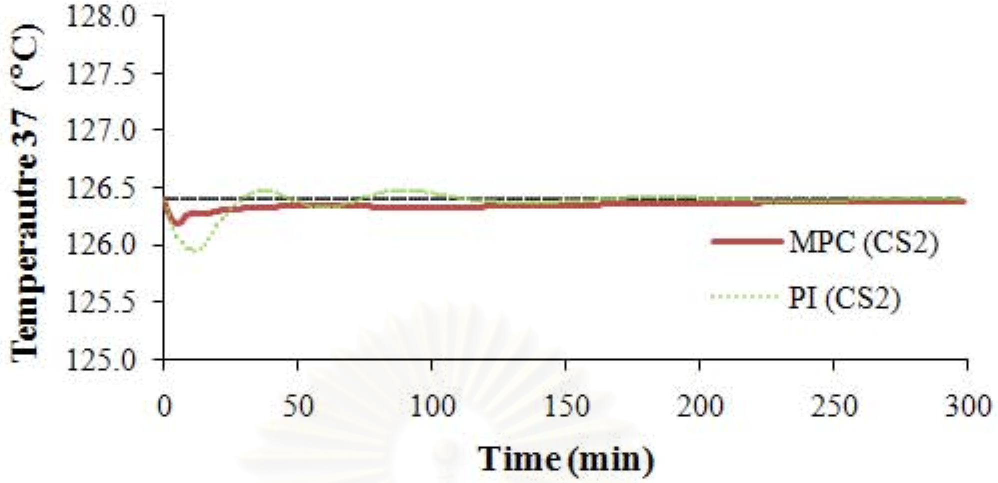


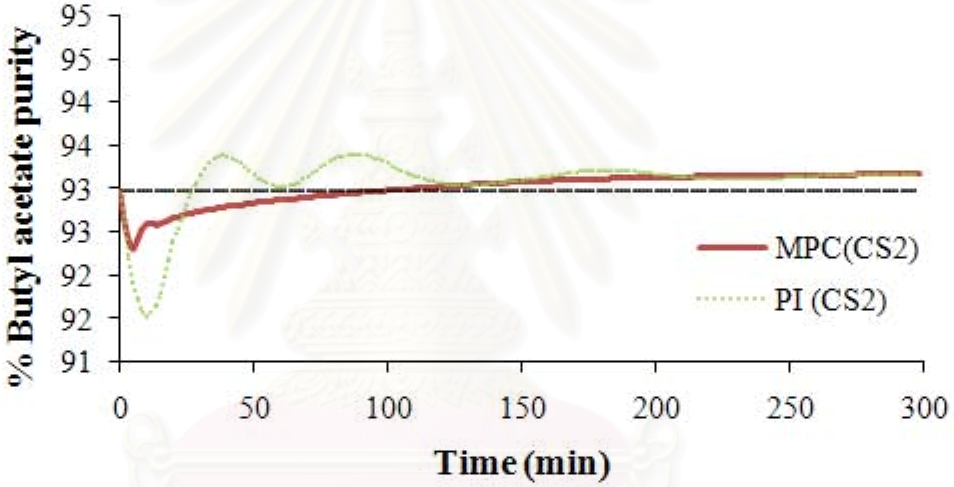
Figure 5-19 Comparison of MPC and PI control, closed-loop responses for -2 °C set point in temperature 37 (a) stage 37 temperature (b) % butyl acetate purity (c) reboiler duty (d) % acetic acid concentration at stage 17 (e) feed ratio

Figure 5-18 and Figure 5-19 show the dynamic responses resulting from set-point step changes in temperature 37 using MPC controller and PI controllers. For ± 2 °C set-point changes in temperature 37, it is obvious that the MPC ensures fast and smooth response, and the PI controller is clearly outperformed by the MPC controller because the PI controller has overshoot and oscillation for + 2 °C set point change and - 2 °C set point change, respectively. The corresponding values of the integral of the absolute error (IAE) criteria are shown in Table 5-2.

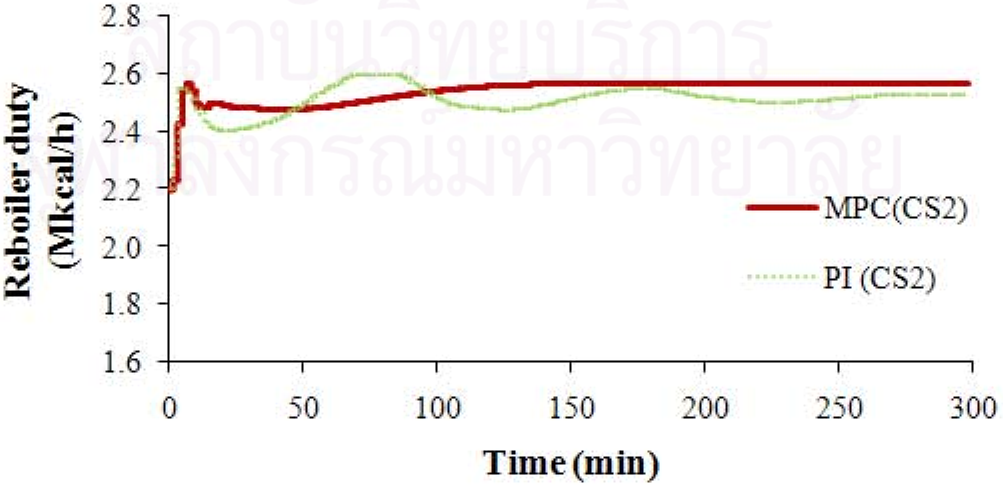
a)


Temperautre 37 (°C)
128.0
127.5
127.0
126.5
126.0
125.5
125.0
0
50
100
150
200
250
300
Time (min)
MPC (CS2)
PI (CS2)

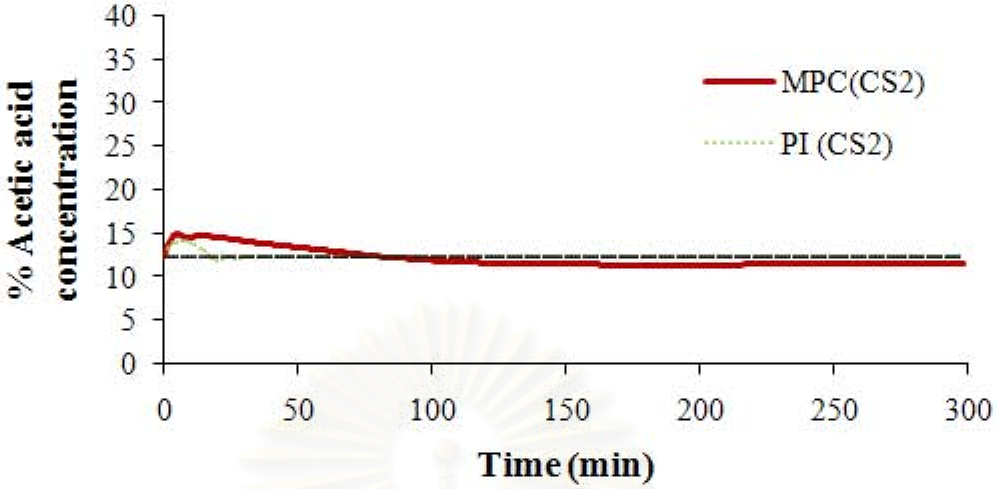
b)


% Butyl acetate purity
95
95
94
94
93
93
92
92
91
0
50
100
150
200
250
300
Time (min)
MPC(CS2)
PI (CS2)

c)


Reboiler duty (Mkcal/h)
2.8
2.6
2.4
2.2
2.0
1.8
1.6
0
50
100
150
200
250
300
Time (min)
MPC(CS2)
PI (CS2)

d)



e)

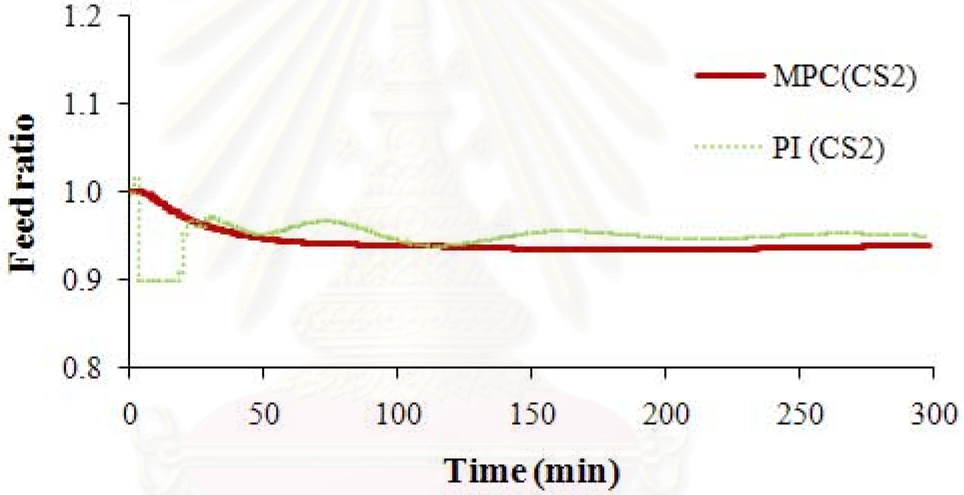
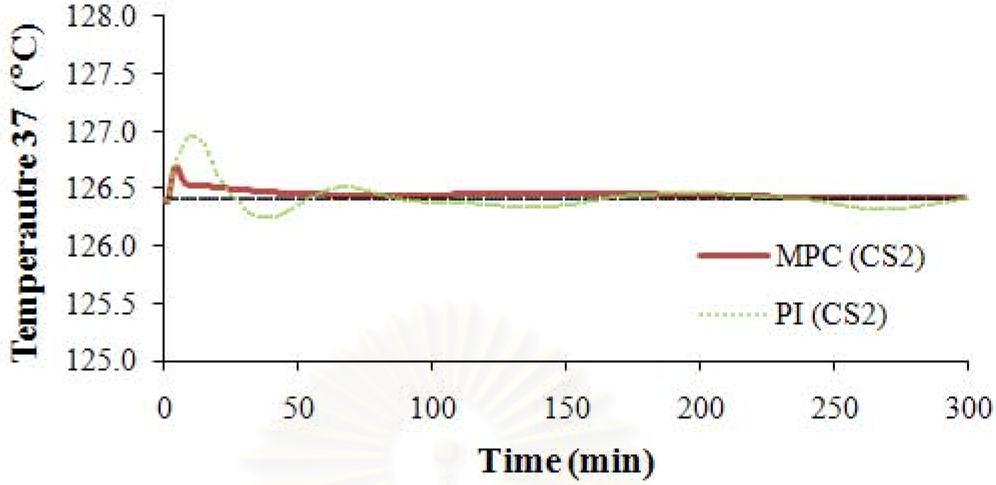


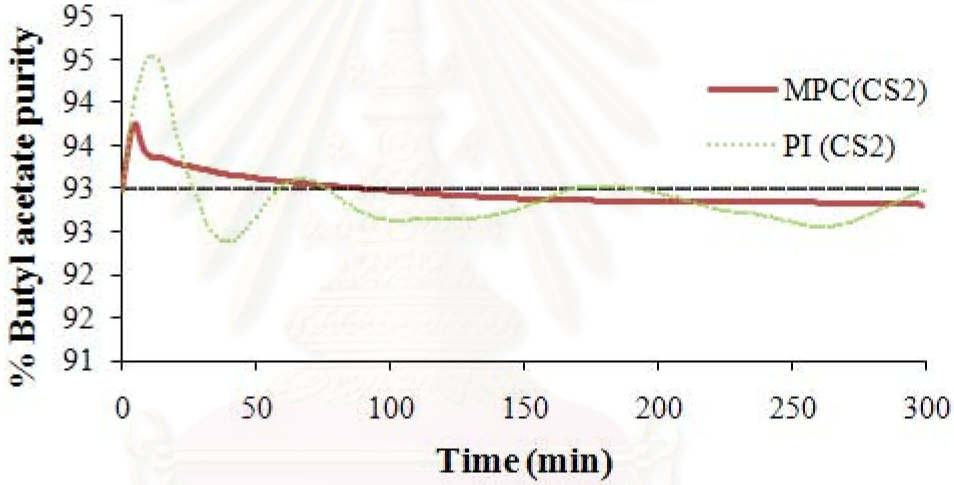
Figure 5-20 Comparison of MPC and PI control , closed-loop responses for +20 % butanol feed (a) stage 37 temperature (b) % butyl acetate purity (c) reboiler duty (d) % acetic acid concentration at stage 17 (e) feed ratio

For + 2 °C set point changes in temperature 37, the MPC controller provides the better control performance for temperature 37 control, which is primary control objective, but slightly poorer control performance for the secondary control objective which is acetic acid composition control at stage 17. For -2 °C set point changes in temperature 37, the MPC controller can give better control performance for both temperature 37 and acetic acid composition.

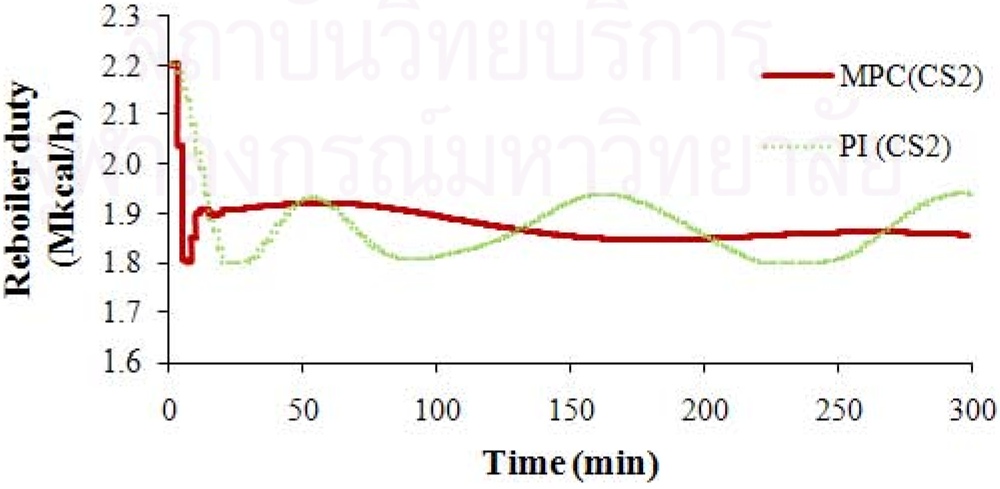
a)


Temperautre 37 (°C)
128.0
127.5
127.0
126.5
126.0
125.5
125.0
0
50
100
150
200
250
300
Time (min)
MPC (CS2)
PI (CS2)

b)


% Butyl acetate purity
95
95
94
94
93
93
92
92
91
0
50
100
150
200
250
300
Time (min)
MPC(CS2)
PI (CS2)

c)


Reboiler duty (Mkcal/h)
2.3
2.2
2.1
2.0
1.9
1.8
1.7
1.6
0
50
100
150
200
250
300
Time (min)
MPC(CS2)
PI (CS2)

d)

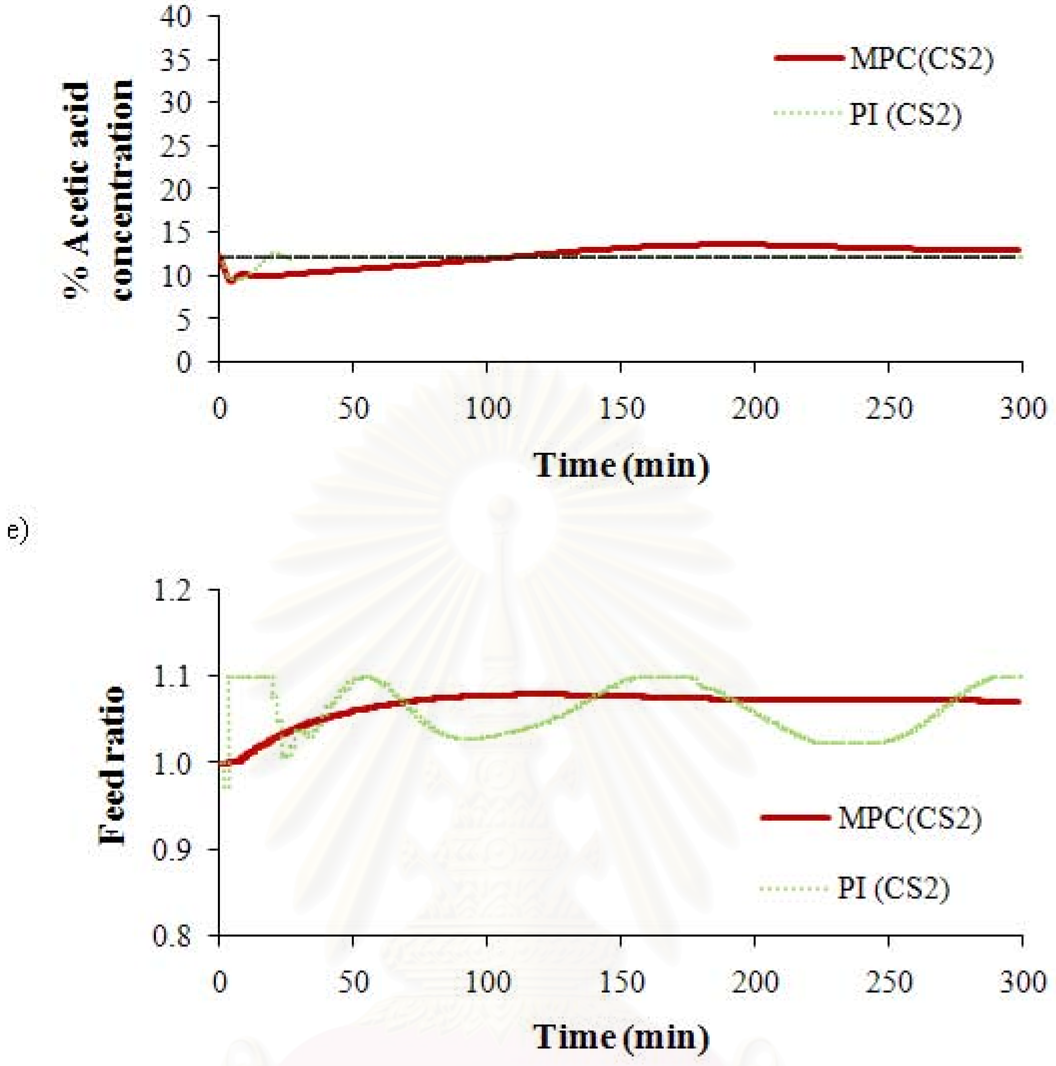
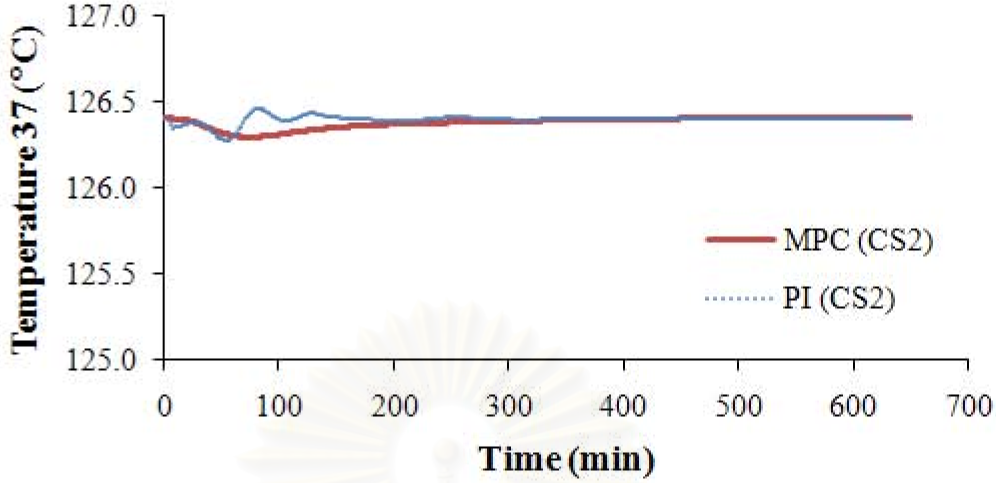


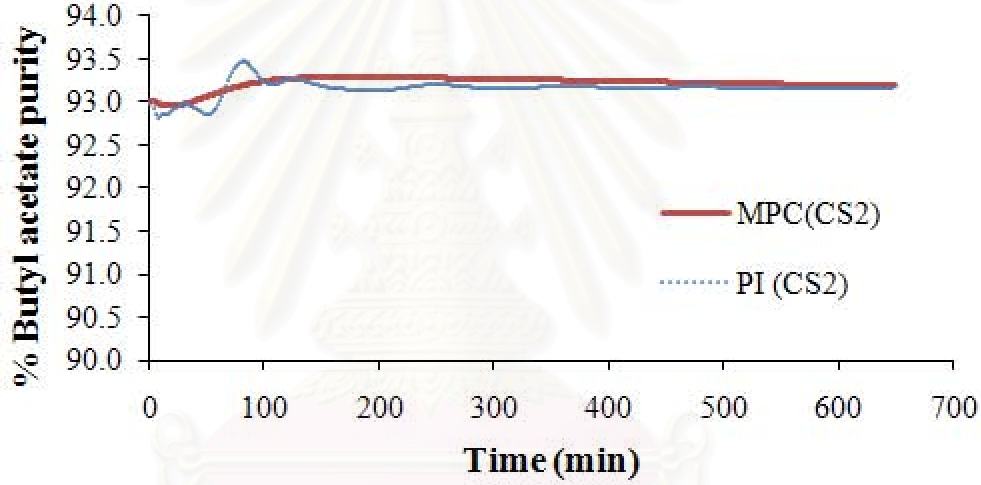
Figure 5-21 Comparison of MPC and PI control , closed-loop responses for -20 % butanol feed (a) stage 37 temperature (b) % butyl acetate purity (c) reboiler duty (d) % acetic acid concentration at stage 17 (e) feed ratio

In order to examine the disturbance rejection, three step changes have been considered; butanol feed flow rate increase of 20 % , butanol feed flow rate reduction of 20 % and a change in acetic feed composition from pure to 10 % water impurity in acetic acid feed flow rate. The dynamic changes resulting from +20 % step changes and -20 % step change in the butanol feed flow rate using the conventional PI controller and MPC are shown in Figure 5-20 and Figure 5-21, respectively. The figures show that the feed rate disturbance rejection of the MPC is superior to that of

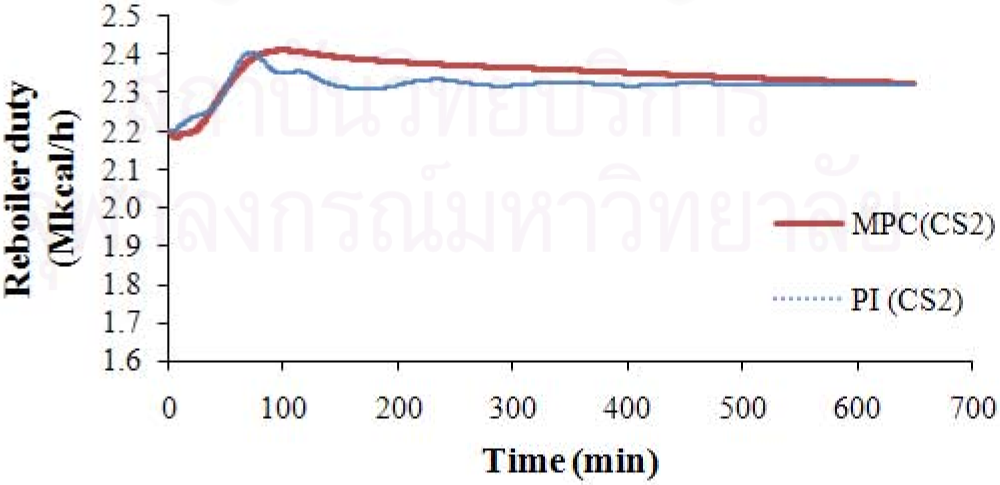
a)


Temperature 37 (°C)
127.0
126.5
126.0
125.5
125.0
0
100
200
300
400
500
600
700
Time (min)
MPC (CS2)
PI (CS2)

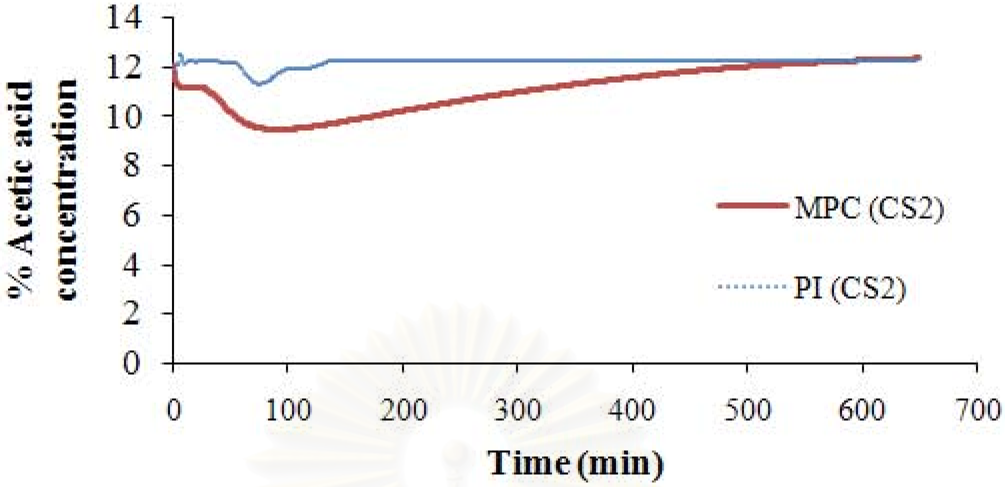
b)


% Butyl acetate purity
94.0
93.5
93.0
92.5
92.0
91.5
91.0
90.5
90.0
0
100
200
300
400
500
600
700
Time (min)
MPC(CS2)
PI (CS2)

c)


Reboiler duty (Mkcal/h)
2.5
2.4
2.3
2.2
2.1
2.0
1.9
1.8
1.7
1.6
0
100
200
300
400
500
600
700
Time (min)
MPC(CS2)
PI (CS2)

d)



e)

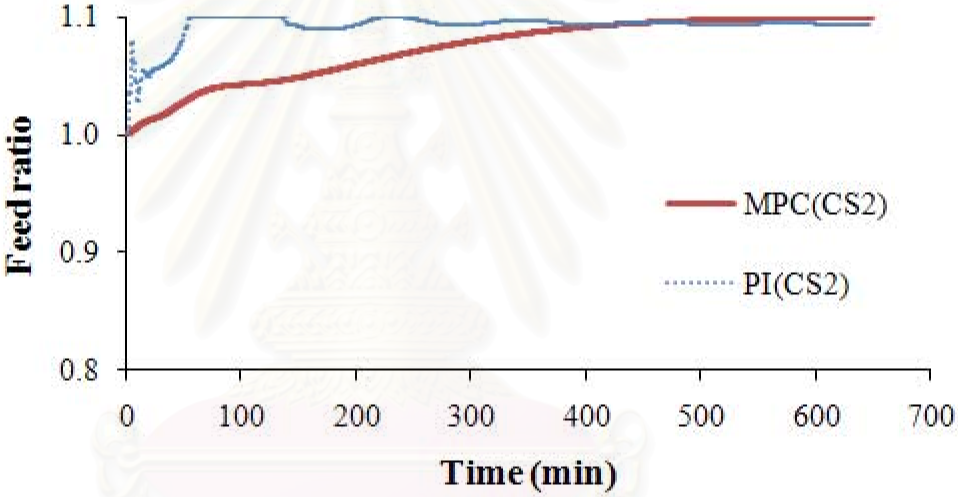


Figure 5-22 Comparison of MPC and PI control , for + 10 % water impurity in acetic acid feed (a) stage 37 temperature (b) % butyl acetate purity (c) reboiler duty (d) % acetic acid concentration at stage 17 (e) feed ratio

the conventional PI controller for temperature 37 control because the MPC results in less overshoot and faster settling time. However, for acetic acid composition control at stage 17, PI controller can give much faster settling time than MPC without offset in controlled variable. Table 5-2 summarizes the comparisons the control criteria between PI control and MPC for butanol feed flow rate changes. Note that in this case, the MPC controller provides the better control the temperature 37, but PI controller performs better than MPC for acetic acid composition control.

The dynamic changes resulting from step changes in the acetic acid feed composition using the PI and MPC are shown in Figures 5-22. In this case, the PI controller slightly outperforms the MPC controller for temperature 37 control but for acetic acid composition control at stage 17 PI control clearly outperforms the MPC controller. The IAE criteria in Table 5-2 shows that the conventional PI controller rejects the water impurity disturbance in acetic acid feed flow rate better than the MPC controller does. The reason why PI controller gives better performance than MPC controller in this case is that the MPC controller use model of the process to calculate control action. Due to the nonlinearity of reactive distillation columns for butyl acetate production, the process gain and dynamic behavior of the process can change significantly due to changes in the operating conditions. As a result, when MPC is applied to such a process, the errors in the models become amplified since all the process models are used to calculate the control action. On the other hand, for a PI controller, while it too is affected by process nonlinearity, it is less susceptible than the MPC controller because its control action is based on the error from setpoint and not a model of the process.

CHAPTER VI

CONCLUSION AND RECOMMENDATIONS

6.1 Conclusion

In this work, a dynamic model of a reactive distillation with 39 stages and 4 components has been developed based on material balance, energy balance, vapor-liquid equilibrium relation and reaction kinetics. The MATLAB solver is used to implement the full dynamic simulation of reactive distillation for butyl acetate production. The accuracy of the dynamic simulation results under this work is verified by comparing with the literature. Most of the simulation results are good agreement with the literature and this confirm that the dynamic model developed in this work is reliable. The mismatch in some data can possibly be due to simplifications of mathematical model, differences of the kinetic and vapor-liquid equilibrium description for highly nonideal and multicomponent system. Then, the effects of operating condition such as reboiler duty, feed rate and feed composition on the dynamic behavior of reactive distillation are investigated under open-loop operation to design control strategy of reactive distillation for butyl acetate production.

Two alternative control structures with using MPC controller of this process are proposed. The control objectives are to maintain bottom product composition at desired purity and to prevent accumulation of unreacted reactants in reactive zone. The first control structure CS1 uses the two-point temperature control to infer butyl acetate bottom product purity and acetic acid compoisition within reactive zone by adjusting reboiler heat duty and feed ratio, respectively. The second control structure CS2 uses the one-point temperature control to infer butyl acetate bottom product purity by manipulating reboiler heat duty and the direct control of acetic acid composition within reactive zone by manipulating feed ratio. Finding proper candidate for the control variables is determined through open-loop sensitivity analysis to choose one controlled variable at maximum sensitivity while still exhibits near linear dynamic behavior. Then, the full dynamic model of a reactive distillation for butyl acetate production is utilized in performing step testing to generate input-output dynamic data from step changes in reboiler duty and feed ratio at the nominal condition. A simplified input-output dynamic model is generated from input-output dynamic data of the nonlinear reactive distillation process model using System Identification Toolbox in MATLAB software. This simplified input-output dynamic model of the reactive distillation column can be approximated by first order equation and is applied in the model predictive control toolbox in Matlab.

The control performance of the MPC controller for disturbance rejection and set-point tracking of CS1 and CS2 structures are investigated. The disturbances considered in this study include changes in butanol feed flow rate and changes in acetic acid feed composition with some water impurity. Although both two control structures have no on-line composition measurement for bottom product, they are able to hold product specification around desired purity despite changes in butanol feed flow rate and acetic acid feed composition. However, it is demonstrated that the disturbance rejection and set-point tracking abilities of CS2 structure is superior for that of CS1 structure. The CS2 structure can handle the set-point and disturbance changes without any oscillation. The direct control of acetic acid composition in reactive zone of CS2 structure can help the control system having faster and smoother response to set-point and disturbance changes. This is because, when there are changes in feed flow rate and feed composition, the acetic acid composition at stage 17 will be affected directly with these changes. This make controller can see the changes of acetic acid composition at stage 17 and can adjust feed ratio to prevent accumulation of unreacted reactants accurately. For CS1 structure, although the controller can adjust feed ratio to maintain temperature in reactive zone, it doesn’t guarantee keeping in correct stoichiometric ratio of acitic acid and butanol during operation of a reactive distillation. Therefore, the performance of CS1 structure is not as good as CS2 structure.

The performance of MPC controller is evaluated and compared to that of conventional PI controller using CS2 structure. The 2x2 PI controllers are used to control the reactive distillation for butyl acetate production. The performances of both controllers are evaluated for changes in the set-point as well as in the feed rate and feed composition. For set-point changes in temperature 37, the results show that the MPC controller ensures fast and smooth response, and the PI controller is clearly outperformed by the MPC controller because it has overshoot and oscillation. The control criteria shows that MPC controller provides the better control performance for temperature 37, which is more important control objective, but poorer control performance for the less important acetic acid composition at stage 17. For butanol feed flow rate changes, the results shows that the disturbance rejection of the MPC is superior to that of the conventional PI controller. The MPC controller results in less overshoot and faster settling time. Also, when considering the control criteria between PI control and MPC for butanol feed flow rate change, MPC controller provides the better control the temperature 37, which was the primary control objective but PI control perform better than MPC for acetic acid composition control, which was the secondary control objective. It is observed that MPC give better performance for temperature 37 control but worse performance for acetic acid composition control.

For step changes in acetic acid feed composition, PI controller slightly outperforms the MPC controller for temperature 37 control but for acetic acid composition control PI control clearly outperforms the MPC controller when considering from results and control criteria. The reason why PI controller give better performance than MPC controller in this case is that the MPC controller use model of the process to calculate control action. Due to the nonlinearity of reactive distillation columns for butyl acetate production, the process gain and dynamic behavior of the process can change significantly due to changes in the operating conditions. As a result, when MPC is applied to such a process, the errors in the models become amplified since all the process models are used to calculate the control action. On the other hand, for a PI controller, while it too is affected by process nonlinearity, it is less susceptible than the MPC controller because its control action is based on the error from setpoint and not a model of the process.

6.2 Recommendations

In this work, we used a simplified input-output dynamic model, which is generated from the full dynamic simulation of nonlinear reactive distillation in our interest operating condition range, as model in MPC controller algorithm. A simplified model which is in the form of the transfer function is approximated by first order equation. However, the appropriate range of fitting a linear transfer function should be one where changes in manipulated variable in both directions can result in a response of the controlled variable in one direction which is approximately the mirror image of the other. In this case the fitted linear transfer function is appropriate. However, due to the nonlinearity of reactive distillation process, the responses of output from changes in manipulated variable in both directions are different. Thus, it is difficult to fit the appropriate linear transfer function for this case. When fitted model is not appropriate, it generates model-plant mismatch in MPC controller algorithm and can make the performance of MPC poorer. One of examples which is used for solving this problem is applying an adaptive controller with variable process model parameters which this method is convenient and appropriate for control of nonlinear reactive distillation column. Another solution is the application of nonlinear model predictive control for the control of reactive distillation for butyl acetate production, but this method will use more energy to solve the optimization problem of nonlinear model predictive control algorithm.

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APPENDICES

APPENDIX A

UNIQUAC METHOD

The UNIQUAC method, introduced by the mid 70's by Abrams and Prausnitz (1975), is one of the most frequently employed tools in correlation experimental data of phase equilibria. The basic equation of the method stems out from an essentially heuristic approach based on an extension of the quasi-chemical theory of Guggenheim (1952) which includes the introduction of the local area fraction as the primary concentration variable and the use of a combinatiorial factor proposed by Staverman (1963).

Within this approach a liquid is considered as a three - dimensional lattice of equi-spaced sites in which the immediate vicinity of a site is called a *cell*. Each molecule in the liquid is divided into attached segments such that each segment occupies one cell and it is assumed that the total number of cells is equal to the total number of segments. Thus, for a binary solution of components 1 and 2, it is considered that there are, respectively, r_1 and r_2 segments for molecule of each component. Also, it is assumed that all segments have the same size but different external contact area. Hence, for any component **i** the number of nearest neighbours is zqi where z is the coordination number of the lattice and q_i is a parameter proportional to the molecules external surface area. Assuming that only nearest neighbours can be considered, the following equation is obtained for the activity coefficient of component i (γ_i) in a multicomponent mixture with molar fraction x_i:

$$\ln \gamma_i = \ln \gamma_i^C + \ln \gamma_i^R \tag{A-1}$$

$$\ln \gamma_i^C = \ln \frac{\varphi_i}{x_i} + \frac{z}{2} q_i \ln \frac{\nu_i}{\varphi_i} + l_i - \frac{\varphi_i}{x_i} \sum_j x_j l_j \tag{A-2}$$

$$\ln \gamma_i^R = q_i \left[1 - \ln \left(\sum_{j=1}^{m} \nu_j \tau_{ji} \right) - \sum_{j=1}^{m} \frac{\nu_j \tau_{ij}}{\sum_{k=1}^{m} \nu_k \tau_{kj}} \right] \tag{A-3}$$

where ν_i is the average area fraction, defined by:

$$\nu_i = \frac{q_i x_i}{\sum_j (q_j x_j)} \tag{A-4}$$

φ_i is the average segment fraction:

$$\varphi_i = \frac{r_i x_i}{\sum_j (r_j x_j)} \tag{A-5}$$

l_j is given by:

$$l_i = \frac{z}{2}(r_i - q_i) - (r_i - 1) \qquad z = 10 \tag{A-6}$$

and τ_{ij} is defined by:

$$\tau_{ij} = \exp\{-\frac{[u_{ij} - u_{jj}]}{RT}\} \tag{A-7}$$

where u_{ij} is the interaction energy.

The structural parameters r and q are defined, respectively, as the van der Waals volume and area of the molecule relative to those of a standard segment which, in turn is, as the authors state, "somewhat arbitrarily" chosen through an approximation (Abrams and Prausnitz (1975)).

With these equations, and assuming a certain value for z it is possible to calculate activity coefficients for phase equilibrium of multicomponent systems employing only two adjustable parameters per binary mixture and, hence, the composition of coexisting phases could be predicted.

In this work, the UNIQUAC model was used to determine the activity coefficients of butyl acetate system in the liquid phase. The UNIQUAC binary interaction parameters and Van der Waals properties, which obtained from the DECHEMA database and the Dortmund Data Bank, are given in TableA-1 and TableA-2, respectively.

Table A-1 Binary interaction parameters for the UNIQUAC equation (cal/mol)

$U_{11} = 0.0$	$U_{21} = 148.2833$	$U_{31} = 712.2349$	$U_{41} = 527.9269$
$U_{12} = -131.769$	$U_{22} = 0.0$	$U_{32} = 24.6386$	$U_{42} = 581.9269$
$U_{13} = -298.433$	$U_{23} = 82.5336$	$U_{33} = 0.0$	$U_{43} = 461.4747$
$U_{14} = -343.593$	$U_{24} = 68.0083$	$U_{34} = 685.71$	$U_{44} = 0.0$

Table A-2 Area and volume parameters for the UNIQUAC equation

Component	**r**	**q**
Acetic acid (1)	2.2024	2.072
Butanol (2)	3.4543	3.052
Butyl acetate (3)	4.8274	4.196
Water (4)	0.92	1.4

APPENDIX B

CONTROL PERFORMANCE INDICES

Integral error measures indicate the cumulative deviation of the controlled variable from its set point during the transient response. The following formulations of the integral can be proposed.

Figure B-1 Definition of error integrals

Integral of the absolute value of error (IAE)

$$IAE = \int_0^\infty \left| e(t) \right| dt \qquad \text{(B-1)}$$

Integral of the square of error (ISE)

$$ISE = \int_0^\infty \left| e^2(t) \right| dt \qquad \text{(B-2)}$$

Integral of time-weighted absolute error (ITAE)

$$ITAE = \int_0^\infty \left| e(t) \right| t dt \qquad \text{(B-3)}$$

where, *e* is the usual error between set point and control variable.

Each formulations of the error have different purposes. The ISE will penalize the response that has large errors, which usually occur at the beginning of a response, because the error is squared. The ITAE will penalize a response which has errors that persist for a long time. The IAE will be less severe in penalizing a response for large errors and treat all errors (large or small, persist for a long or short time) in a uniform manner.

VITA

Mr Teerasit Komkrajang was born on March 17th, 1983 in Chonburi, Thailand. He finished high school from Assumption College Sriracha, Chonburi and received bachelor's degree in Engineering from the department of Chemical Engineering, Faculty of Engineering, Chulalongkorn University, Bangkok, Thailand in 2006. He began his graduate study in June, 2006 and received her Master degree in Engineering from Chemical Engineering Department, Faculty of Engineering, Chulalongkorn University in 2008.