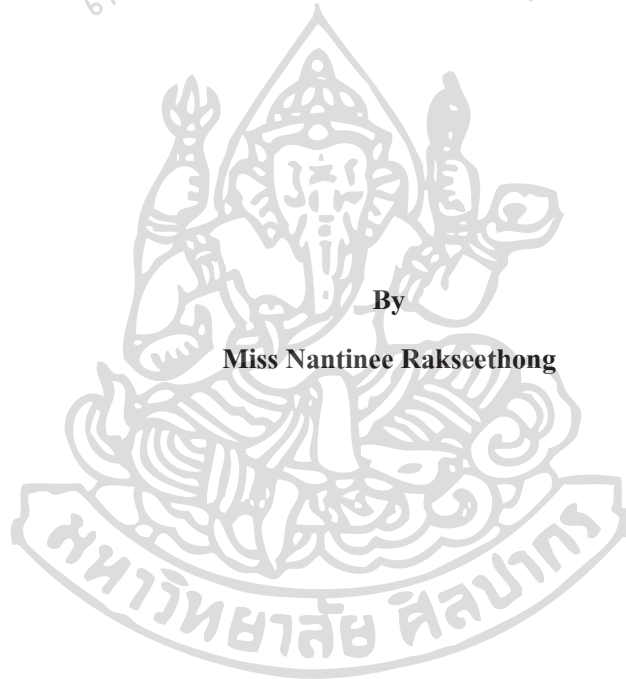




CONTROLLABILITY ANALYSIS OF ALKALI-CATALYZED BIODIESEL PROCESS

สำนักหอสมุดกลาง



By

Miss Nantinee Rakseethong

A Thesis Submitted in Partial Fulfillment of the Requirements for the Degree

Master of Engineering Program in Chemical Engineering

Department of Chemical Engineering

Graduate School, Silpakorn University

Academic Year 2012

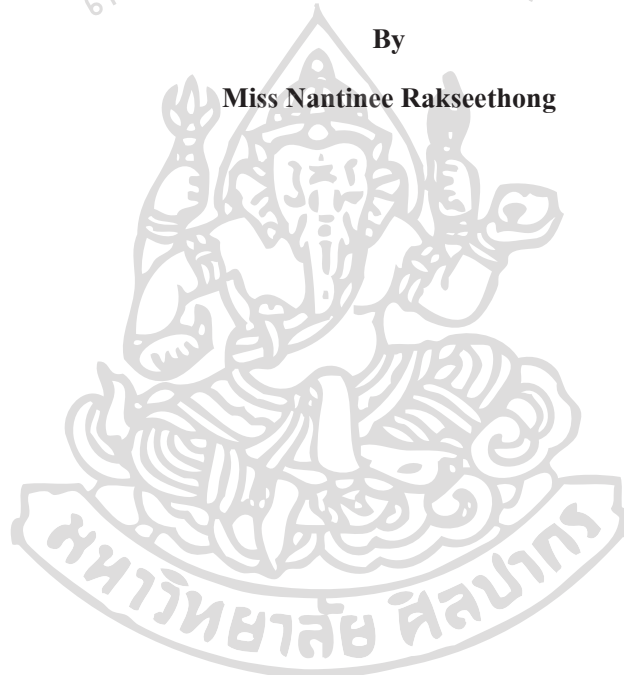
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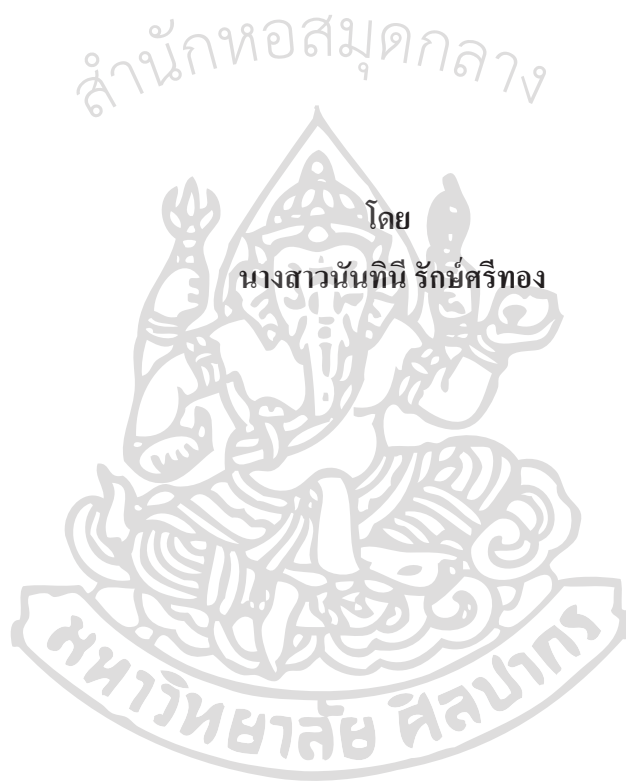
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การวิเคราะห์ความสามารถในการควบคุมได้ของกระบวนการผลิตไบโอดีเซลที่เร่งปฏิกิริยาด้วยเบส



วิทยานิพนธ์นี้เป็นส่วนหนึ่งของการศึกษาตามหลักสูตรปริญญาวิศวกรรมศาสตรมหาบัณฑิต

สาขาวิชาวิศวกรรมเคมี

ภาควิชาวิศวกรรมเคมี

บัณฑิตวิทยาลัย มหาวิทยาลัยศิลปากร

ปีการศึกษา 2555

ลิขสิทธิ์ของบัณฑิตวิทยาลัย มหาวิทยาลัยศิลปากร

The Graduate School, Silpakorn University has approved and accredited the Thesis title of “controllability analysis of alkali-catalyzed biodiesel process” submitted by Miss Nanthinee Rakseethong as a partial fulfillment of the requirements for the degree of Master of Engineering in Chemical Engineering

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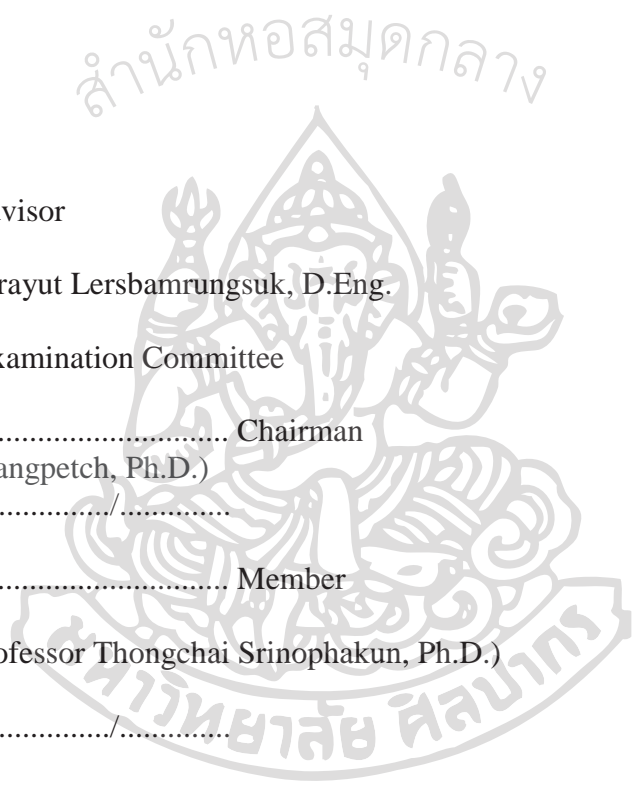
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LERSBAMRUNGSUK, D.Eng. 62 pp.

Controllability is an inherent property of process. Poor design can make the plant uncontrollable. This research focuses on controllability analysis of alkali-catalyzed biodiesel process, in particular transesterification section. The research is divided as two parts. The first part concerns design and control of transesterification reactor. Dynamic model of the reactor is developed and used for analysis of effects of important design parameters including residence time, reactor temperature, and methanol/oil molar ratio to conversion and controllability. The controllability is analyzed based on poles, zeros, and gain. The second part concerns design and control of reactor connected with methanol recovery column. To avoid difficulty of the modeling, Aspen Plus is chosen for the steady-state modeling of the system. Because the system is multivariable, relative gain array and singular value analysis are used as controllability analysis tool. Effects of design parameters including reactor temperature, methanol/oil molar ratio, distillate rate, and reboiler duty to the reactor conversion and methanol mole fraction and controllability are analyzed. Dynamic simulations on Aspen Dynamics are used to validate the analysis result.



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คำสำคัญ : ความสามารถในการควบคุมได้/ กระบวนการผลิตไบโอดีเซล/ กระบวนการผลิตไบโอดีเซลที่เร่งปฏิกิริยาด้วยเบส

บันทึกนี้ รัศม์ศรีทอง : การวิเคราะห์ความสามารถในการควบคุมได้ของกระบวนการผลิตไบโอดีเซลที่เร่งปฏิกิริยาด้วยเบส. อาจารย์ที่ปรึกษาวิทยานิพนธ์ : ดร.วิรัช เลิศบำรุงสุข. 62 หน้า.

วัตถุประสงค์ของงานวิจัยฉบับนี้คือการศึกษาศักยภาพในการควบคุมได้ของเครื่องปฏิกรณ์แบบทรานสเอสเทอริฟิเคชันและหอกลิ้นแยกเมทานอลในกระบวนการผลิตไบโอดีเซลที่เร่งปฏิกิริยาด้วยเบส โดยแบ่งการพิจารณาออกเป็นสองส่วนคือ 1. ศึกษาการออกแบบและควบคุมเครื่องปฏิกรณ์แบบทรานสเอสเทอริฟิเคชัน โดยการนำไดนามิคโมเดลของเครื่องปฏิกรณ์มาใช้ในการวิเคราะห์ผลกระทบที่เกิดจาก เรสซิเดนซ์ไทม์ อุณหภูมิของเครื่องปฏิกรณ์ และสัดส่วนโดยโมลของเมทานอลต่อน้ำมัน และวิเคราะห์ความสามารถในการควบคุมได้บนพื้นฐานของ โพล ซีโร และ เกน 2. ศึกษาการออกแบบและควบคุมเครื่องปฏิกรณ์แบบทรานสเอสเทอริฟิเคชันเชื่อมต่อกับหอกลิ้นแยกเมทานอล ในส่วนนี้เพื่อหลีกเลี่ยงความยากในการสร้างแบบจำลองจึงได้เลือกใช้ แอสเพน พลัส มาใช้ในการสร้างแบบจำลองสถานะคงที่ของระบบ และเนื่องจากเป็นระบบที่มีหลายตัวแปรจึงเลือก รีเลทีฟเกนอาร์เรย์ และ ซิงกูลาร์แวลูอานาไลซิส มาใช้ในการวิเคราะห์ความสามารถในการควบคุม โดยวิเคราะห์จากผลกระทบที่เกิดจากอุณหภูมิของเครื่องปฏิกรณ์ สัดส่วนโดยโมลของเมทานอลต่อน้ำมัน อัตราการกลั่น และ รีบอยเลอร์ดีวดี และใช้แอสเพน ไดนามิคมาใช้ในการตรวจสอบผลที่วิเคราะห์ได้

ภาควิชาวิศวกรรมเคมี

บัณฑิตวิทยาลัย มหาวิทยาลัยศิลปากร

ลายมือชื่อนักศึกษา.....

ปีการศึกษา 2555

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CHAPTER 1

INTRODUCTION

To be competitive in industries, plants are designed to have more process integration. Although the integration can increase plant's efficiency, interactions among the process units are required. Furthermore, the behavior of overall processes may change drastically comparing to those of individual units. As a result, the control consideration based on individual unit may fail to operate the plant properly. This motivates to a need of an efficient method for controllability design.

Chemical processes are usually highly nonlinear, complex, and multivariable system. Poor design without controllability consideration may result in uncontrollable plant. Morari (1997) pointed out that some simple controllability criteria should be included within synthesis procedures. Skogestad and Postlethwaite (2005) stated that "controllability is independent of the controller, and is a property of the plant (or process) alone. It can only be affected by changing the plant itself; that is, by (plant) design changes".

Controllability is an inherent property of the process itself, it should be considered at the design stage before the control system design is fixed. The most preferable way to consider controllability at the design stage is to include controllability as one of the design objectives just like traditional economic ones in the design optimization problem. The methods to evaluate controllability are complicated, and the required information is usually missing or unknown in the design stage. This is one of the prominent reasons why controllability study has not been performed when designing chemical processes (Lee et al., 2001).

Over the last few years, fatty acid methyl ester has become important as research has intensified on the utilization of vegetable oil and animal fats derivatives for liquid fuel, known as biodiesel (Noureddini and Zhu, 1997). The most common way to produce biodiesel is by transesterification, which refers to catalyzed chemical reaction involving vegetable oil and alcohol the yield fatty acid alkyl esters and glycerol. Methanol is the most commonly used alcohol because of its low cost (Zhang et al., 2003).

In biodiesel production plant, a transesterification reactor is one of the most important unit. Transesterification reaction is affected by the molar ratio of alcohol to oil, catalyst, reaction temperature, reaction time and the free fatty acid and water content of the oil or fats. To the best of our knowledge, there are only few researches studying on controllability of this unit. This work focuses on controllability of transesterification section including transesterification reactor and methanol recovery column. The design and operating condition of Zhang et al (2003) and

kinetic parameters of Nouredini and Zhu (1997) will be used in this research.

1.2 Research objective

The objective of the research is to analyze controllability of transesterification reactor and methanol recovery column in alkali-catalyzed biodiesel process.

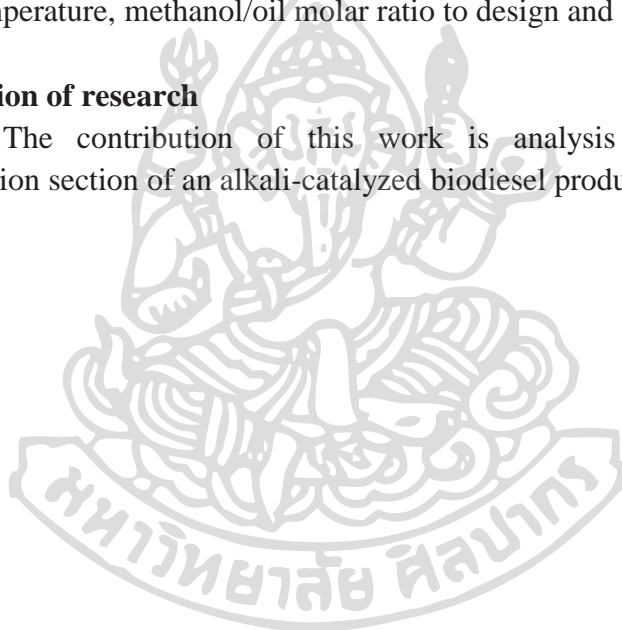
1.3 Scopes of research

The scopes of this work are listed below:

1. Consider alkali-catalyzed biodiesel process.
2. Analyze controllability of transesterification reactor.
3. Analyze controllability of transesterification reactor connected with methanol recovery column.
4. Study effects of design parameters such as residence time, reactor temperature, methanol/oil molar ratio to design and control of the system.

1.4 Contribution of research

The contribution of this work is analysis of controllability of transesterification section of an alkali-catalyzed biodiesel production.



CHAPTER 2

THEORY

This chapter provides some backgrounds necessary for understanding this research.

2.1 Biodiesel

2.1.1 Biodiesel production

Biodiesel is an alternative fuel which has many merits. It is derived from renewable, domestic resource, thereby relieving reliance on petroleum fuel imports. It is biodegradable and non-toxic (Zhang et al., 2003). There are four primary ways to make biodiesel, direct use blending, microemulsion, thermal cracking (pyrolysis) and transesterification (Ma and Hanna, 1999). The most common way to produce biodiesel is by transesterification, which refer to a catalyzed chemical reaction involving vegetable oil and an alcohol to yield fatty acid alkyl esters and glycerol. Triglyceride (TG) is the main component of vegetable oil that consists of three long chain fatty acids esterified to a glycerol backbone. When triglycerides react with an alcohol, the three fatty acid chains are released from the glycerol skeleton and combine with the alcohol to yield fatty acid alkyl esters. Glycerol is produced as a by-product. Methanol is the most commonly used alcohol because of its low cost. In general, a large excess of methanol is used to shift the equilibrium far to the right (figure 1).

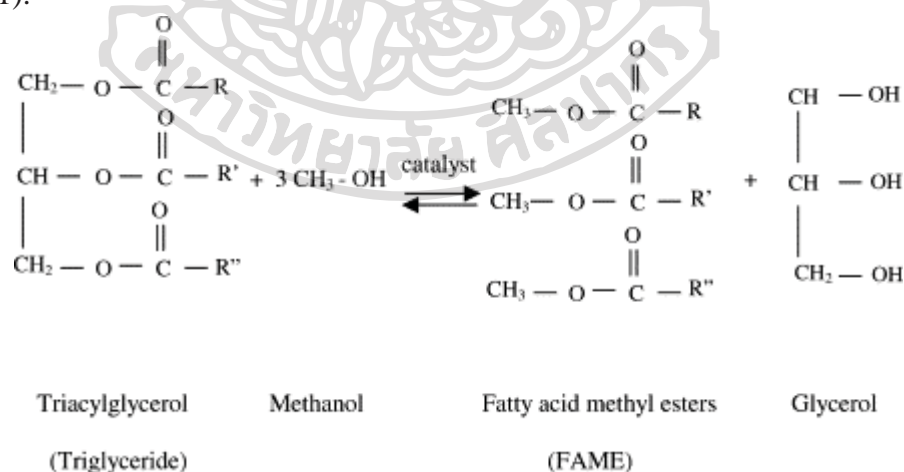


Figure 1 A schematic representation of the transesterification of triglyceride with methanol to produce fatty acid methyl esters.

Source: Zhang et al. "Biodiesel production from waste cooking oil: 1. Process design and technological assessment", **Bioresource Technology** 89 (2003): 1-16.

In the biodiesel reaction, the yield is affected by different parameters; however, researcher have considered four main factors; molar ratio between alcohol and triglycerides, reaction time, catalyst, and reactor temperature. For instance, an excess of alcohol can guarantee the complete conversion of fats or oils to esters in a short time. Usually, the molar ratio is 6:1 in presence of alkali catalyst. On the other hand, the conversion rate of fatty acids esters increase with time and it has been found that the yield reaches a maximum at the reaction time of less than 90 min. An excess of reaction time will decrease the yield due to the backward reactions and it will be cause more fatty acids to form soap. A higher reaction temperature can decrease the viscosities of oil resulting in an increase reaction rate and a shortened reaction time. Moreover, it is possible that the yield of biodiesel decreases when the reaction temperature rise beyond the optimal level since it accelerates the saponification reaction of triglyceride.

For instances, in the Malaysia Palm Oil Board (MPOB) the reactant feed temperature and the reaction pressure is used as the manipulated variables. The pressure is operated at pressure higher than the atmospheric pressure and it is controlled in order to keep the methanol in the liquid phase (Benavides and Dewikar, 2012).

2.1.2 Biodiesel production process

Alkali catalyzed biodiesel production

The flowsheet of alkali-catalyzed biodiesel production of Zhang et al. (2003a) is given in Figure 2. Description of each unit is given as follows:

Transesterification Reactor

Transesterification reactor was a device for reaction between methanol and virgin oil using sodium hydroxide as catalyst. The reaction was carried out with a 6:1 molar ratio of methanol to oil, 1% sodium hydroxide (based on oil). In reactor, 95% of oil was assumed to be converted to FAME, producing glycerol as a byproduct. Stream from the transesterification reactor was introduced to methanol recovery distillation.

Methanol Recovery Distillation

Methanol recovery distillation was designed with seven theoretical stages including reboiler and reflux drum. This unit was used for separation between methanol and other components. Stream methanol recovery was a pure methanol distillate, containing 94% of the total methanol in stream from the transesterification reactor. Bottom stream was sent to water washing column after being cooled in exchanger.

Water Washing Extraction

The purpose of this step was to separate the FAME from the glycerol, methanol and catalyst. A water washing column with five theoretical stages was used in this process. The FAME in stream EST3 was separated from the glycerol, methanol and catalyst by adding water. The amounts of unconverted oil, methanol and water in

removed in distillate stream WATMEOH. At the bottom, 92% glycerine was obtained as a high quality by-product.

2.2 Linear controllability analysis

2.2.1 Linear state-space model

State-space models have an important advantage: they provide a compact and useful representation of dynamic models that are in the form of set of ordinary differential equation (ODEs).

Consider a system with m input (vector u) and l output (vector y) which has internal description of n state (vector x). A natural way to represent many physical systems is by nonlinear state-space model of form

$$\dot{x} = f(x, u); y = g(x, u) \quad (1)$$

where $\dot{x} = dx/dt$ and f and g are nonlinear function.

Consider a linear state-space model

$$\dot{x}(t) = Ax(t) + Bu(t) \quad (2)$$

$$y(t) = Cx(t) + Du(t) \quad (3)$$

The elements of x are referred to as state variables. The elements of y are typically a subset of x , namely, the state variables that are measured. In general, x , u , d and y are functions of time. The time derivative of x is denoted by \dot{x} . Matrices A , B , C , and D are real matrices.

If equation (2) is derived by linearizing equation (1) then $A = \partial f / \partial x$ and $B = \partial f / \partial u$. A is sometimes called the state matrix. These equations provide a convenient means of describing the dynamic behavior of proper, rational, linear systems. They may be rewritten as

$$\begin{bmatrix} \dot{x} \\ y \end{bmatrix} = \begin{bmatrix} A & B \\ C & D \end{bmatrix} \begin{bmatrix} x \\ u \end{bmatrix} \quad (4)$$

2.2.2 Relative Gain Array (RGA)

One of the most importance factors, common to all process control applications, is the correct (best) pairing of the manipulated and controlled variables. A number of qualitative techniques are available to assist in the selection process. One of the earliest methods proposed was the Relative Gain Array (RGA). The original technique is based upon the open loop steady state of the process and provides two important items of information (Bristol, 1996):

1. A measure of process interactions.
2. A recommendation concerning the most effective pairing of controlled and manipulated variables

Consider the 2x2 system show in Figure 3. Suppose mv_2 remain constant, then a step change in mv_1 of magnitude Δmv_1 will produce a change Δcv_1 in output cv_1 . Thus, the gain between mv_1 and cv_1 when mv_2 is kept constant is given by

$$g_{11}|_{mv_2} = \frac{\Delta cv_1}{\Delta mv_1}|_{mv_2} \quad (5)$$

If instead of keeping mv_2 constant, cv_2 is now kept constant by closing the loop between cv_2 and mv_2 . A step change in mv_1 of magnitude Δmv_1 will result in another change in cv_1 . The gain in this case is denoted by:

$$g_{11}|_{y_2} = \frac{\Delta cv_1}{\Delta mv_1}|_{cv_2} \quad (6)$$

The gain relationships, equation (5) and (6) may have different values. If interaction exists, then the change in cv_1 due to change in mv_1 for two cases when mv_2 and cv_2 are kept constant, will be different.

$$\text{The ratio: } \lambda_{11} = \frac{g_{11}|_{mv_2}}{g_{11}|_{cv_2}} \quad (7)$$

The ratio is a dimensionless value and it defines the relative gain between the output cv_1 and the input mv_1 .

Interpretation of the relative gain follows as:

$\lambda_{ij} = 1$. There is no interaction with other control loops.

$\lambda_{ij} = 0$. Manipulated input, i , has no effect on output, j .

$\lambda_{ij} = 0.5$. There is a high degree of interaction. The other control loops have the same effect on the output, j , as the manipulated input, i .

$0.5 < \lambda_{ij} < 1$. There is interaction between the control loops. However, this would be the preferable pairing as it would minimize interactions.

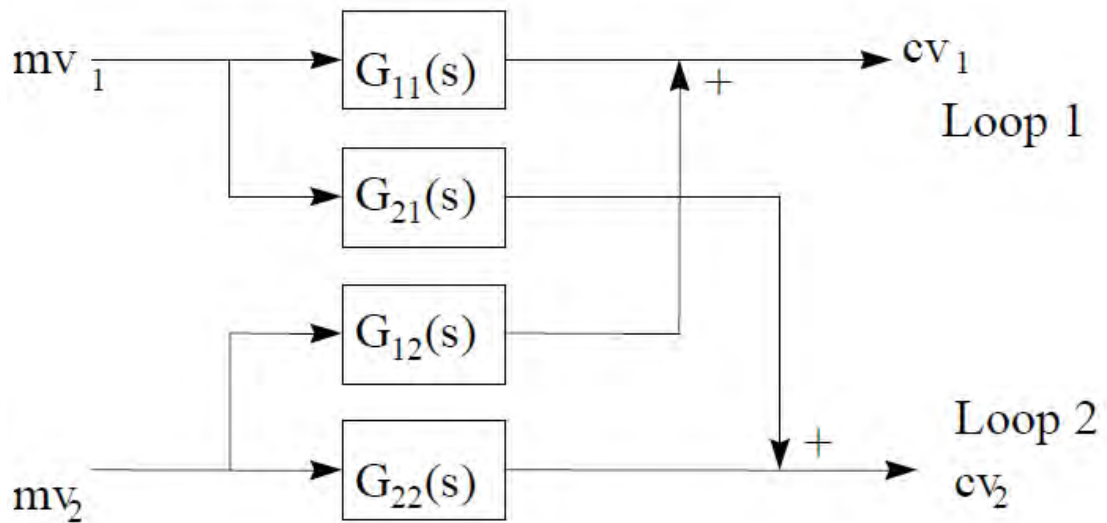


Figure 3 (2x2) Multivariable model structures
 Source: Willis M.J. "Multivariable control: An introduction", ncl.ac.uk (1999): 3.

$\lambda_{ij} > 1$. The interaction reduces the effect gain of the control loop. Higher controller gains are required.

$\lambda_{ij} > 10$. The pairing of variables with large RGA elements is undesirable. It can indicate a system sensitive to small variations in gain and possible problem applying model based control technique.

$\lambda_{ij} < 0$. Care must be taken with negative RGA elements. Negative off-diagonal elements indicate that closing the loop will change the sign of the effective gain. More importantly, negative diagonal elements can indicate 'integral instability' i.e. the control loop is unstable for any feedback controller.

Skogestad (1987) demonstrated that the RGA calculation can be expressed in matrix notation, facilitating computation for systems larger than 2x2

$$\text{RGA} = G \cdot (G^T)^{-1} \quad (8)$$

where, G is the process gain matrix, ' \cdot ' denotes element by element multiplication, $[\cdot]^{-1}$ denotes a matrix inverse, and $[\cdot]^T$ is the transpose operator (Willis M.J., 1999).

2.2.3 Singular Value Analysis

A very powerful set of techniques dealing with sets of equation or matrices that are either singular or numerically very close to singular is the so-called Singular value decomposition (SVD). SVD allows one to diagnose the problems in a given matrix and provide numerical answer as well when applied to the frequency response of a MIMO system $G(s)$ with m inputs and l outputs.

Consider a fixed frequency ω where $G(j\omega)$ is a constant $l \times m$ complex matrix, and denote $G(j\omega)$ by G for simplicity. Any matrix G may be decomposed into its SVD following:

$$G = U \Sigma V^H \quad (9)$$

where

Σ is $l \times m$ matrix with $k = \min \{l, m\}$ non-negative singular value

U is $l \times l$ unitary matrix of output singular vectors

V is $m \times m$ unitary matrix of input singular vectors

Some advantages of the SVD over the eigenvalue decomposition for analyzing gains and directionality of multivariable plants are:

1. The singular value gives better information about the gains of the plant.
2. The plant directions obtained from the SVD are orthogonal.
3. The SVD also applies directly to non-square plants.

2.2.4 Poles

The poles of a system are the eigenvalues of the state-space A -matrix, and this is the definition given below. More generally, the poles of $G(s)$ may be somewhat loosely defined as the finite value $s = p$ where $G(p)$ has singularity.

The pole polynomial corresponding to a minimal realization of a system with transfer function $G(s)$ is the least common denominator of all non-identically zero minors of all orders of $G(s)$.

The following theorem allows us to obtain the poles directly from the transfer function matrix $G(s)$ and is useful for hand calculations. It also has the advantage of yielding only the poles corresponding to a minimal realization of the system. For linear systems, the poles determine stability.

A linear dynamic system in equation (6) is stable if and only if all the poles are in the open left hand plane (LHP). A matrix A with such a property is said to be “stable”.

$$\dot{x} = Ax + Bu \quad (10)$$

2.2.5 Zeros

Zeros of a system arises when competing effects, internal to the system, are such that the output is zero even when the inputs (and the states) are not themselves identically zero. For a single-input single-output (SISO) system the zeros z_i are the solution to $G(z_i) = 0$. In general, it can be argued that zeros are values of s at which $G(s)$ loses rank (from rank 1 to rank 0 for a SISO system). This is the basic for the following definition of zeros for a multivariable system.

The zero polynomial, corresponding to a minimal realization of the system, is the greatest common divisor of all the numerators of all order- r minors of $G(s)$, where r is the normal rank of $G(s)$, provided that these minors have been adjusted in such a way as to have the pole polynomial as their denominator.

CHAPTER 3

LITERATURE REVIEWS

This chapter consists of two parts including the literature related to controllability of chemical process and biodiesel production

3.1 Controllability of chemical process

Hangzhou et al. (2011) studied the stability and phase behavior of chemical process systems when parameters are varied that separate in four cases (1) Stable minimum phase (S-MP). (2) Stable non-minimum phase (S-NMP). (3) Unstable minimum phase (US-MP). (4). Unstable non-minimum phase (US-NMP). In this situation, there are two continuous-stirred tank reactors (CSTRs) that work in series. Then, the conclusions can be drawn:

1. For a process, the steady state solutions change when the feed flow and recycle flow rates vary and at the same times, the system stability and phase behaviors change. The same situation can occur with other parameters in the process system
2. If the maximum conversion point in a process is unstable and has non-maximum phase behavior, it is difficult to design an appropriate control system at the optimized operating point.
3. Because individual unit operations or subsystem in a system differ from the global system in terms of these features, system, overall stability and phase behavior should be considered.

Hangzhou et al. (2011) presented chemical process is used to demonstrate how to analyze process stability and controllability. The conclusion is drawn that overall system stability and phase behaviors should be considered because the individual unit operations or subsystems differ from the total system in terms of these features. Therefore, the analysis of stability and controllability of process systems is important in terms of the design of inherently safer processes.

Cao et al. (1996) used a modified singular value analysis (SVA) technique and the new optimization-based method via based on nonlinear optimal control for predict the effect of manipulated variable constraints (MVCs) on closed loop performance. As a result, both methods give the same result. However, the dependence of the result on frequency needs to be investigated using the modified SVA

Shen et al. (2011) explored the design and control of biodiesel production process with phase separation and internal recycle in the reactor system in the case where vegetable oil reacts with methanol to form methyl esters and glycerol in the presence of homogeneous alkaline catalyst (NaOH). The result show if feed ratio of methanol to triglyceride (FR) decrease, the reaction system requires larger hold up. All designs at the lower FR meet the glycerol specification because the recycle ratio of glycerol can be adjusted to recycle more methanols to the reactors. So internal recycle of glycerol is attractive even though glycerol is a product because the glycerol contains a great of methanol

Wu and Yu (1996) studied the case of snowball effect, recycle system, process characteristics for individual units and the interconnected system and proposed disturbance rejection in plant wide control and a systematic controller tuning procedure. The process consists of a reactor and a distillation column in an interconnected structure. An irreversible first order reaction ($A \rightarrow B$) occurs in a continuous stirred tank reactor (CSTR). The reaction rate (k) is a function of temperature described by Arrhenius expression. This is an exothermic reaction and the reaction temperature (T) is controlled by manipulated cooling water flow rate. A mixture of A and B is fed into a 20-tray distillation column. The column has a partial reboiler and a total condenser. The product B is recycled back into the CSTR.

They consider in two cases as: individual unit and interconnected units. In the first case is divided into two units, separator and reactor. For separator, assuming that the top and the bottom composition (x_D and x_B) are controlled. The disturbances are feed flow rate (F) change and feed composition changes. For reactor, they consider CSTR with a first order reaction. The reactor conversion is a good measure of the reactor performance. In the second case, they consider the plant that has a reactor and separator connected under a recycle structure. The assumption of theoretical tray, equimolar overflow and constant relative volatility are made in modeling distillation column. They found the steady-state analyses provide useful information to validate this potential in plant wide control structure, balanced scheme, is proposed to overcome the snowball effect. Simulation results show that the balance control structure is effective in handling large load changes while maintaining good closed loop performance.

Skogestad (2000) presented a systematic procedure for finding suitable controlled variables based on only steady-state information. The first task in the plantwide control or control structure design problem:

1. Selection of control variables
2. Selection of manipulated variables
3. Selection of measurement
4. Selection of control configuration

5. Selection of the controlled type

He presented a stepwise procedure for selecting control variables:

Step 1: Degree of freedom

Step 2: Cost function and constraints

Step 3: Identify the most important disturbances

Step 4: Optimization

Step 5: Identify candidate control variables

Step 6: Evaluation of loss

Step 7: Further analysis and selection

The procedure requires a steady-state process model and clear definition of the cost function to be minimized during operation.

3.2 Biodiesel production

Noureddini and Zhu (1997) studied the effect of mixing on the kinetic of the transesterification of soybean oil with methanol via constant molar ratio of alcohol to oils 6:1 and the concentration of catalyst 0.2 wt% based on the mass of soybean oil while varied temperature and mixing intensity. Kinetic data were collected at three mixing intensities and five temperature levels. The temperature dependency of the reaction rate constants and the activation energies were determined for all the forward and reverse reactions. The dependency of the reaction rate constants on mixing intensity was also investigated. The rate constants for the first two reactions are increase with temperature for both the forward and reverse reactions. The rate of increase was much faster for the forward reaction than the reverse reaction. Therefore, in reaction (TG to DG) and (DG to MG), both forward reaction are favored at higher temperature. In the third reaction (MG to GL), the reaction rate constants are decrease as the reaction temperature is increased. This means a more favorable forward reaction than the reverse reaction in the third reaction at high temperature. The overall formation rate of the methyl ester is also more favorable at higher reaction temperature

Narvaze et al. (2007) studied in two case: 1) Effect of temperature was evaluated, keeping the molar ratio constant by consider at three temperatures were 50°C, 55°C, and 60°C at the molar ratio was 6:1. The results showed that the effect of temperature, within the rang studied, the influence of temperature over the palm oil methanolysis is low but perceptible that agree with Noureddini and Zhu who reported that the influence of temperature on the concentration is low around the boiling point of alcohol. 2) Effect of molar ratio while keeping temperature constant at 55°C were conducted with three molar ratio: 3:1, 4.5:1, and 6:1. In this case, the result showed the conversion of palm oil and productivity to methyl ester increased with the excess of methanol that mean the molar ratio that got the highest conversion is 6:1.

Zhang et al. (2003a) carried out process simulation on four flowsheets for continuous alkali- and acid-catalyzed using virgin vegetable oil or waste cooking oil. Analysis showed that the pretreatment required in the alkali-catalyzed process using waste cooking oil was not necessary in the other processes and operating condition of the process show in table 1. As a result, the alkali-catalyzed process using virgin oil (process I) was the simplest with the least amount of process equipment but had a higher raw material cost than other processes but the process that using waste cooking oil as raw material (process II), it was the most complex process with the greatest number of equipment pieces because of the addition of pretreatment unit for free fatty acid removal. The acid-catalyzed process using waste cooking oil (process III) had less equipment pieces than process II, but the large methanol requirement resulted in more and larger transesterification reactor, as well as larger methanol distillation column. The acid-catalyzed process to produce biodiesel from waste cooking oil using hexane extraction (process IV) had the same merits and limitations as process III. However, the addition of hexane and methanol/water solvents increased the number of equipment pieces and size of some separation unit. For process simplicity, the alkali-catalyzed process using virgin vegetable oil is recommended.

Table1 Operating condition and specification of reactor

Parameters	Symbol	Value	Unit
Reactor temperature	T_R	60	°C
Reactor pressure	P	4	bar
Reactant flow rate	F_{MeOH}	6.037	L/min
	F_{oil}	20.9	L/min
Initial concentration of MeOH	$C_{MeOH,in}$	20.915	mol/L
Initial concentration of TG	$C_{TG,in}$	1.005	mol/L
Reactor volume	V	13,734	L

Zhang et al (2003b) presented the economic feasibilities of four continuous processes to produce biodiesel. Economic evaluation was based on this assumption:

1. Each process was based on a plant capacity of 8,000 tonne/year
2. Operating hour for the biodiesel plant were assume to be 8,000 h/hour
3. Both waste cooking oil and virgin oil used, as the feedstock for biodiesel production, are free of water and any solid impurities
4. In the simulation, pump efficiency was assume to be 70%
5. Superheated, low and high-pressure stream were used as the heating media and water was the cooling medium

The basis of economic assessment showed the alkali-catalyzed process using virgin oil had the lowest total capital investment because of the relatively small

sizes and carbon steel construction of most of the process equipment. Sensitivity analysis of different processes for biodiesel production showed that plant capacity, the price of waste cooking oil and the price of the biodiesel were the major factor affecting the economic feasibility of the biodiesel production.

CHAPTER 4

METHODOLOGY

This chapter provided an overview of alkali catalyzed biodiesel production in the research and describes the proposed methodology for analysis of control-ability.

Methodology for analysis of controllability

This research is divided into two parts. The first part concerns design and control of the transesterification reactor and design and control transesterification reactor connected with methanol recovery column. The kinetic data are taken from Noureddini and Zhu (1997) while the capacity of the reactor is taken from Zhang et al. (2003).

4.1 Design and control of transesterification reactor

4.1.1 Design of transesterification reactor

Balat and Balat (2010) pointed out that there are three important design parameters of transesterification reactor: residence time, methanol/oil ratio, and reactor temperature. The design of these parameters can be given by solving a dynamic model deriving from component and energy balances of the reactor. The design objective is to have 0.98 conversion of fresh feed oil. In general, there will be multiple solutions to satisfy the design conversion.

4.1.2 Controllability of transesterification reactor

Among the solutions to satisfy the design conversion, there will be some conditions that are more controllable. Controllability will be analyzed based on linear system theory.

Controllability of the reactor can be given using zeros, poles, gain of the transfer matrix of the system,

$$P = C*([s*I-A]^{-1})*B \quad (11)$$

For controllability analysis, this research defines concentration of free fatty acid methyl ester (FAME) as controlled output, and methanol inlet flowrate and reactor temperature as inputs.

4.1.3 Dynamic control of transesterification reactor

The controllability analysis of the selected design in the previous section will be validated using dynamic simulation in Aspen Plus Dynamics v.7.3. Oil feed rate is defined as disturbance for the control problem.

4.2 Design and control of transesterification reactor connected with methanol recovery column

4.2.1 Design of transesterification reactor connected with methanol recovery column

Wills (2000) provided some basic ideas and rules that may be used to select a distillation control strategy. Generally, the variables in Table 2 need to be controlled. Hence, the addition design objective is to have 0.01 mole fraction of methanol in the column bottom while reboiler duty and distillate rate are considered as design parameters to be manipulated. Again, there will be multiple operations to satisfy the design objective and hence the most controllable operation should be chosen.

Table 2 Typical variables that have to be maintained in a distillation column

Controlled variable (CV)	Reason
Composition of the distillation stream, x_D	Product quality
Composition of the bottom stream, x_B	Product quality
Liquid level in the reflux drum	Maintain inventory
Liquid level at the base of the column	Maintain inventory
Pressure in the column	Maintain inventory

Source Wills. "Select a distillation column control strategy", ncl.ac.uk (2000)

4.2.2 Controllability of transesterification reactor connected with methanol recovery column

For controllability analysis, this research defines concentration of free fatty acid methyl ester (FAME) and mole fraction of methanol at the bottom as controlled outputs, and methanol/oil feed ratio, reactor temperature, reboiler duty, and distillate rate as inputs. Because the system is multivariable, relative gain array (RGA) and singular value analysis (SVA) will be used as controllability analysis tools. The process gain matrix will be given by perturbation of each input.

4.2.3 Dynamic control of transesterification reactor connected with methanol recovery column

The controllability analysis of the selected design in the previous section will be validated using dynamic simulation in Aspen Plus Dynamics v.7.3. Again, oil feedrate is defined as disturbance for the control problem.

CHAPTER 5

RESULTS AND DISCUSSION

This chapter describes the design and control of transesterification reactor and methanol recovery column in alkali-catalyzed biodiesel production.

5.1 Design and control of transesterification reactor

5.1.1 Design of transesterification reactor

Based on the American Society of Agriculture Engineers (ASAE) the overall biodiesel production line consists of a crude oil degumming, refining, transesterification, ester washing, glycerol refining and a methanol recovery sections. Among these sections, the transesterification is the most crucial section because any drifts in standard operating condition may lead to significant change in the process variables and production quality (Mjalli et al., 2009). The first part of this chapter will dedicate to design and control of this reactor. The reactor is shown in Figure4.

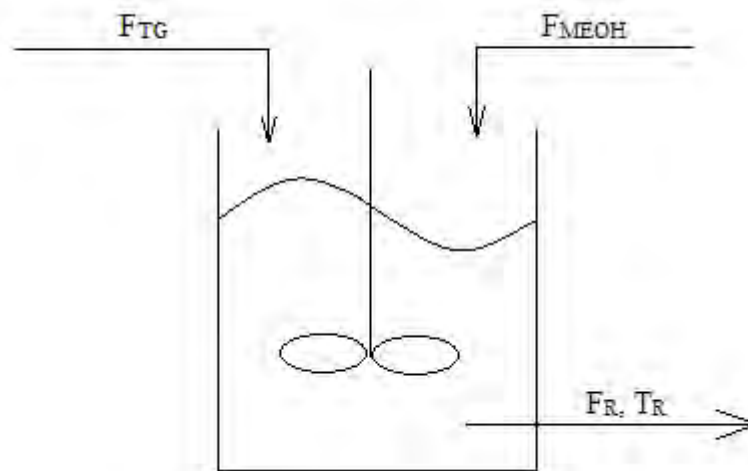
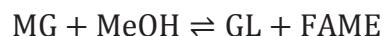
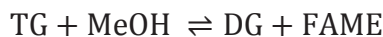


Figure 4 Transesterification reactor

To consider design and control of the reactor, dynamic model is needed. In transesterification, triglyceride (TG) reacts with methanol (MeOH) in the presence of an alkaline catalyst, to yield ester of fatty acids (FAME) and glycerol (GL). Diglyceride (DG) and Monoglyceride (MG) are the intermediates. The reaction steps are shown in equation (12)



The dynamic model of transesterification reactor is derived using the principles of component balances as shown in equations (13) - (18). Hypotheses are (a) the system is in a well mixed condition, (b) isothermal operation, and (c) virgin pure triolein is assumed as feedstock

$$\frac{dC_{\text{TG}}}{dt} = \frac{F_{\text{TG}}C_{\text{TG,in}} - F_{\text{R}}C_{\text{TG}}}{V} - r_1 \quad (13)$$

$$\frac{dC_{\text{DG}}}{dt} = \frac{-F_{\text{R}}C_{\text{DG}}}{V} + (r_1 - r_2) \quad (14)$$

$$\frac{dC_{\text{MG}}}{dt} = \frac{-F_{\text{R}}C_{\text{MG}}}{V} + (r_1 - r_3) \quad (15)$$

$$\frac{dC_{\text{MeOH}}}{dt} = \frac{F_{\text{MeOH}}C_{\text{MeOH,in}} - F_{\text{R}}C_{\text{MeOH}}}{V} - (r_1 + r_2 + r_3) \quad (16)$$

$$\frac{dC_{\text{FAME}}}{dt} = \frac{-F_{\text{R}}C_{\text{FAME}}}{V} + (r_1 + r_2 + r_3) \quad (17)$$

$$\frac{dC_{\text{GL}}}{dt} = \frac{-F_{\text{R}}C_{\text{GL}}}{V} + r_3 \quad (18)$$

The kinetic of reaction is taken from Nouredini and Zhu (1997) as shown in equations (19) - (21) and Table 3.

$$r_1 = k_{1f}C_{\text{TG}}C_{\text{MeOH}} - k_{1r}C_{\text{DG}}C_{\text{FAME}} \quad (19)$$

$$r_2 = k_{2f}C_{\text{DG}}C_{\text{MeOH}} - k_{2r}C_{\text{MG}}C_{\text{FAME}} \quad (20)$$

$$r_3 = k_{3f}C_{\text{MG}}C_{\text{MeOH}} - k_{3r}C_{\text{GL}}C_{\text{FAME}} \quad (21)$$

Table 3 Energy of activation (Ea, cal/mol) and reaction rate constant (k) from Modified Arrhenius equation at 323.15 K.

Reaction	Ea	k
TG → DG	11,707	0.050
DG → TG	8,482	0.110
DG → MG	18,439	0.215
MG → DG	13,433	1.228
MG → GL	7,937	0.242
GL → MG	10,992	0.007

Source: Nouredin H. and Zhu D. "Kinetics of transesterification of soybean oil." **JAACS**. (1997) Vol.74, pp.1457-1463

where Modified Arrhenius equation, $k=A\text{Te}^{(-E_a/RT)}$

The model is validated with the experimental result of Nouredini and Zhu (1997) that shows good prediction of conversion.

In the design of reactor, high conversion is usually required because this lowers the amount of triglyceride and makes the separation task easier. In this research, design specification is to achieve 0.98 conversion of fresh feed oil. Furthermore, to avoid large reactor size, residence time is assumed as 60 min.

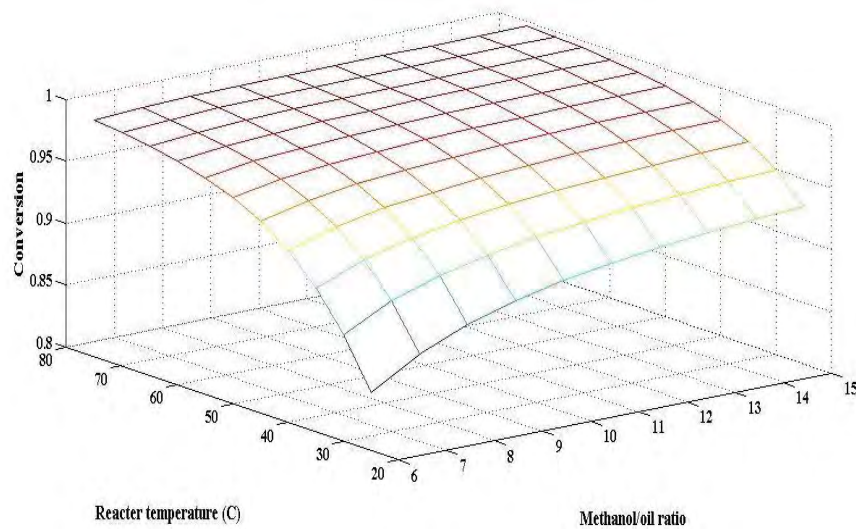


Figure 5 3D plot between feed ratio, temperature and conversion for 60 min residence time.

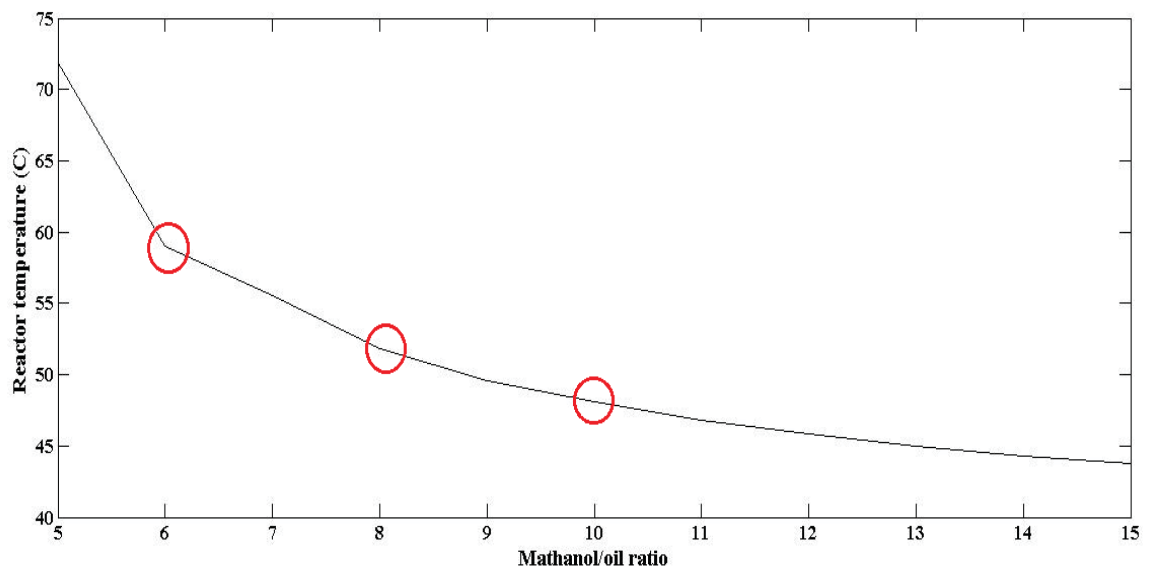


Figure 6 Contour plot of 98% oil conversion for 60 min residence time

By solving the model (13) – (18) at steady-state, the 3D plot of conversion as function of temperature and methanol/oil ratio can be given as shown in Figure 5 and the contour plot of 0.98 conversion is shown in Figure 6.

Figure 6 shows that there are several possible operating points to achieve 98% oil conversion. In term of design based on economic consideration, small methanol/oil ratio is required to reduce the separation task of methanol recovery column. However, in this work controllability will be considered. The three conditions corresponding to high temperature with low ratio, moderate temperature with moderate ratio, and low temperature with high ratio chosen for controllability analysis are

Point 1: FR = 6, T = 59.0916 °C

Point 2: FR = 8, T = 51.7996 °C

Point 3: FR = 10, T = 48.1042 °C

5.1.2 Controllability of transesterification reactor

To analyze controllability of the reactor, some variables need to be defined first. In this work, conversion of oil is considered as controlled variable (CV) while methanol/oil ratio (or methanol feed flowrate, F_{MeOH}) and reactor temperature (T_R) are considered as inputs and one of these inputs will be chosen as manipulated variables (MV).

By linearizing the model in equations (13) - (18), transfer function of the systems can be given using equation (11).

Table 4 shows zeros of the two inputs: F_{MeOH} and T_R . RHP-zeros occur for the input F_{MeOH} and hence, this should not be a good manipulated input for control. Hence, the reactor temperature will be chosen as a manipulated input.

Table 4 Zero location in the transfer function of methanol feed flowrate (F_{MeOH}) and reactor temperature (T_R) to conversion

Point	Input	Zeros				
1	T_R	-4.3344	-0.6389	-0.0563	-0.0167	-0.0167
	F_{MeOH}	-6.4523	-0.6495	0.1945	-0.0167	-0.0167
2	T_R	-3.4703	-0.6098	-0.0330	-0.0167	-0.0167
	F_{MeOH}	-4.6601	-0.5889	0.0123	-0.0167	-0.0167
3	T_R	-3.1830	-0.6170	-0.0167	-0.0167	-0.0165
	F_{MeOH}	-3.8873	-0.5722	-0.0603	-0.0167	-0.0167

The poles of the system are presented in Table 5. The results showed that all poles are negative real, and this implies the system is open-loop stable without oscillation. However, note that for the condition with higher the temperature and lesser the ratio (point 1) some poles are farther from the imaginary axis. In terms of controllability, this implies the system is more stable.

Table 5 Pole location in the transfer function of methanol feed flowrate (F_{MeOH}) and reactor temperature (T_R) to conversion

Point	Poles					
1	-6.6399	-0.6533	-0.1815	-0.0167	-0.0167	-0.0167
2	-4.7067	-0.5867	-0.1641	-0.0167	-0.0167	-0.0167
3	-3.9028	-0.5703	-0.1674	-0.0167	-0.0167	-0.0167

The last observation is the steady-state gain of the process. Table 6 shows the gain of F_{MeOH} and T_R to conversion. In general, large gain of manipulated input is required to avoid input saturation. From the result, point 3 has the highest gain of the temperature. Although the result on poles shows that the point 1 is more stable, the reactor is physically stable. Based on the analysis above for the individual reactor, the condition with lesser the temperature and higher the methanol/oil ratio is the most controllable. This is contrast to the recommended condition for economic purpose that usually requires the least excess methanol to reduce energy for methanol recovery.

Table 6 Gain of methanol feed flowrate (F_{MeOH}) and reactor temperature (T_R) to conversion

Point	Input	Gain
1	T_R	0.2194
	F_{MeOH}	0.3929
2	T_R	0.2450
	F_{MeOH}	0.2450
3	T_R	0.2602
	F_{MeOH}	0.1924

5.1.3 Dynamic control of transesterification reactor

In this work, oil feed flow rate is defined as disturbance variable while conversion of oil and reactor temperature are defined as controlled and manipulated variable, respectively. The steady-state flowsheet of this reactor is created using Aspen Plus v.7.3 and then exported to Aspen Plus Dynamics v.7.3 for dynamic simulation. The pressure drop of all valves is assumed as 0.5 bar. The reactor is sized using the information of residence time with the $L/D = 2$.

This research assumes TG can be detected online. Conversion of oil is observed by measuring the molar flow of TG before and after the reactor. Because the

reactor temperature cannot be directly manipulated, the cascade loop of oil conversion as primary loop sending targeting temperature to the secondary loop of reactor temperature and reactor duty is used. Other loops are (1) reactor level loop for stability, (2) ratio control of methanol/oil for keeping the ratio, and (3) oil feed flowrate loop for a given production rate. The controller tuning of all loops are fixed using default values.

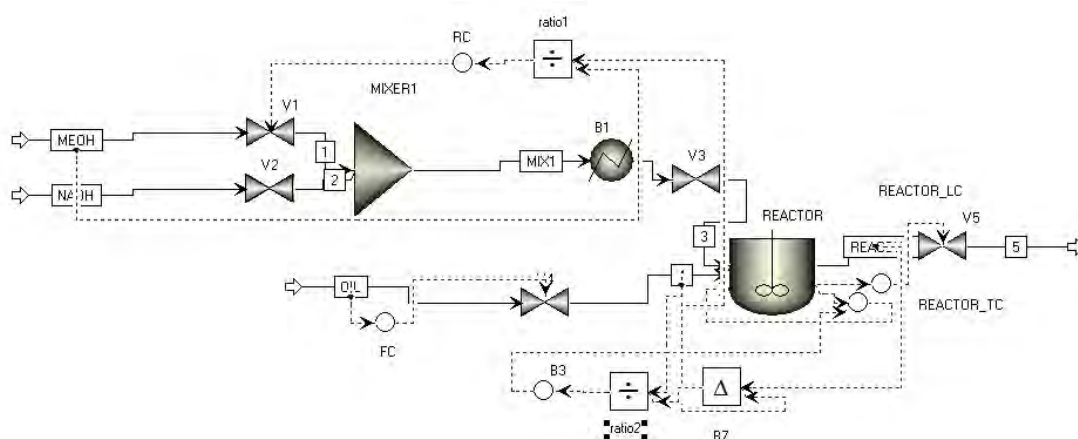


Figure 7 Control structure for optimal operation of the transesterification reactor

Controllability of the three operating points chosen in the section 5.1.2 will be validated by dynamic simulation. Dynamic simulation for the system with operating point 1, methanol/oil ratio = 6, $T_R = 59.0916$ °C is shown in Figures 8 – 10. In the first hour the system is operated at the normal condition of methanol/oil ratio 6, reactor temperature 59.0916 °C, and oil feedrate 1.1858 kmol/hr, after that the oil feedrate is changed to 1.065 kmol/hr. the reduction of the oil feedrate results in longer residence time and hence, the conversion is raised. However, with the existence of the control system, the conversion can be returned to the original setpoint. The conversion can be within $\pm 0.1\%$ of the setpoint in 13 hours and at the setpoint in 32 hours.

Dynamic simulation for the system with operating point 2, methanol/oil ratio = 8, $T_R = 51.7996$ °C is shown in Figures 11-13. The conversion can be within $\pm 0.1\%$ of the setpoint in 12 hours and at the setpoint in 32 hours.

Dynamic simulation for the system with operating point 3, methanol/oil ratio = 10, $T_R = 48.1042$ °C is shown in Figures 14-16. The conversion can be within $\pm 0.1\%$ of the setpoint in 12.5 hours and at the setpoint in 32 hours.

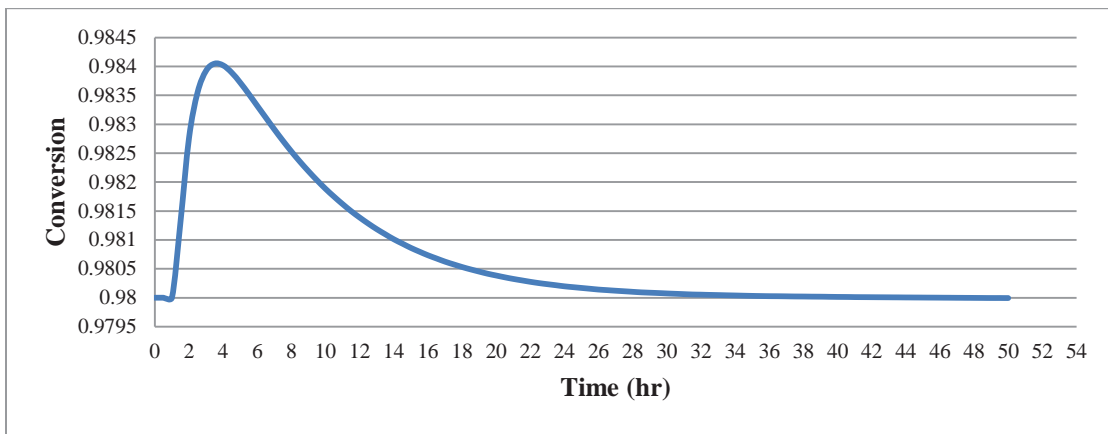


Figure 8 Effect of oil inlet to conversion for methanol/oil ratio = 6, $T_R = 59.0916\text{ }^\circ\text{C}$

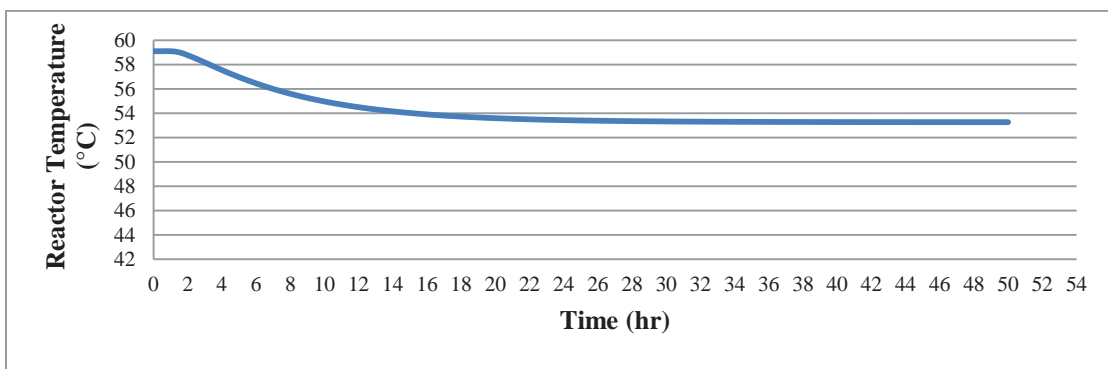


Figure 9 Reactor temperature change while oil inlet decrease for methanol/oil ratio = 6, $T_R = 59.0916\text{ }^\circ\text{C}$

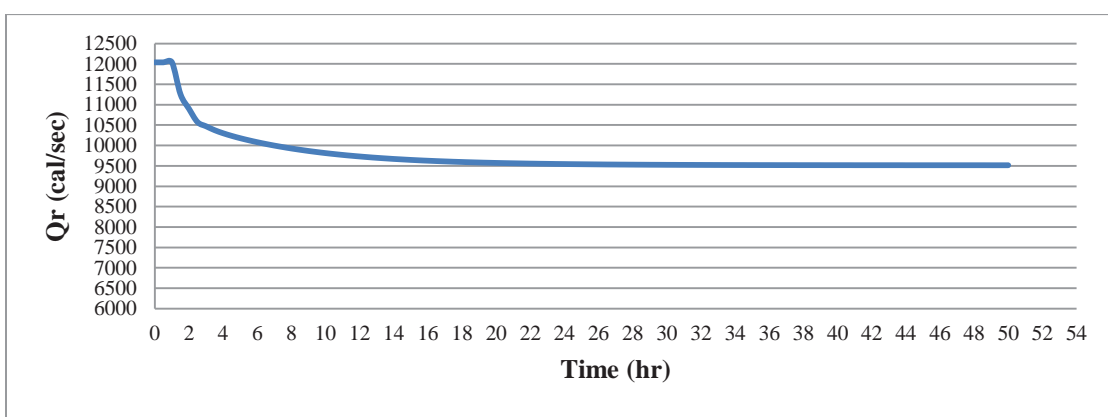


Figure 10 Heat inlet to reactor change while oil inlet decrease for methanol/oil ratio = 6, $T_R = 59.0916\text{ }^\circ\text{C}$

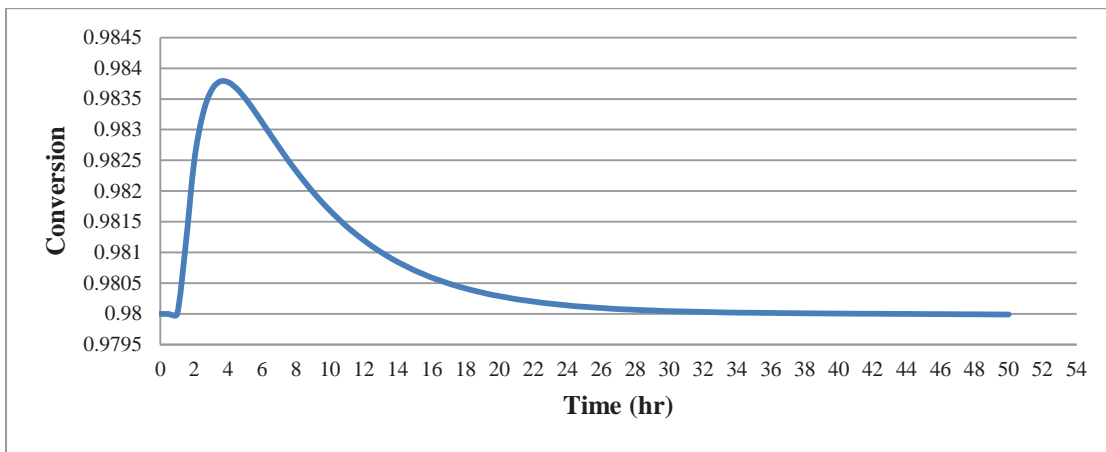


Figure 11 Effect of oil inlet to conversion for methanol/oil ratio = 8, $T_R = 51.7996\text{ }^\circ\text{C}$

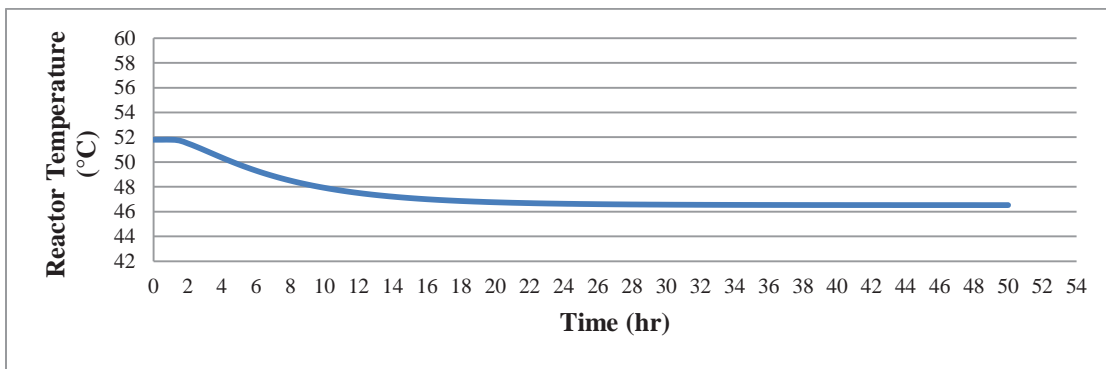


Figure 12 Reactor temperature change while oil inlet decrease for methanol/oil ratio = 8, $T_R = 51.7996\text{ }^\circ\text{C}$

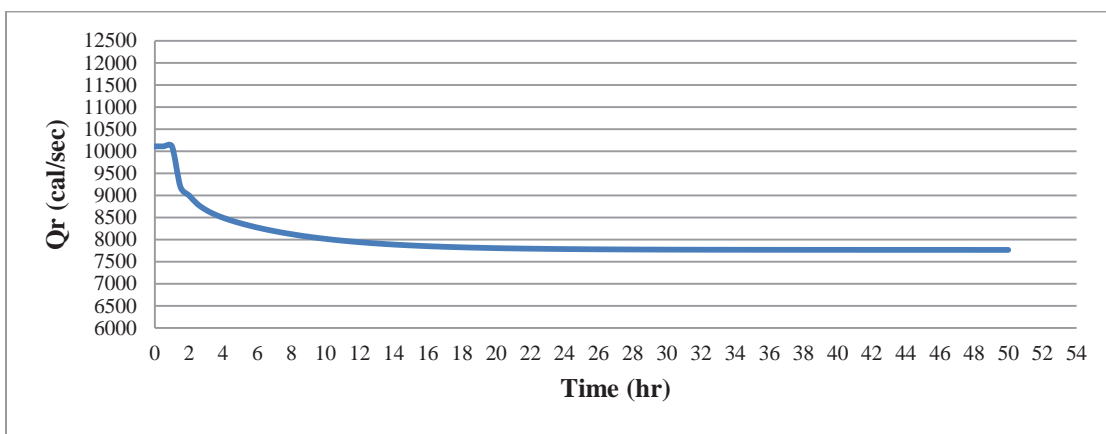


Figure 13 Heat inlet to reactor change while oil inlet decrease for methanol/oil ratio = 8, $T_R = 51.7996\text{ }^\circ\text{C}$

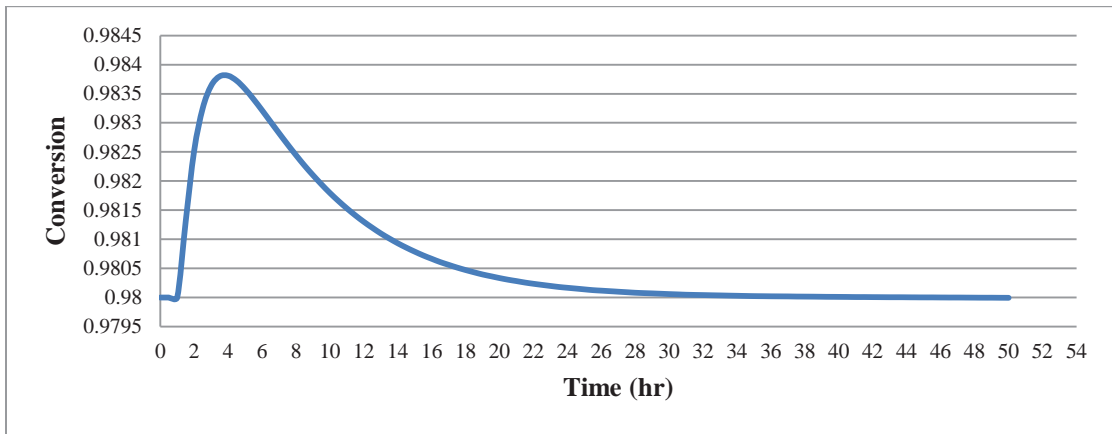


Figure 14 Effect of oil inlet to conversion for methanol/oil ratio = 10, $T_R = 48.1042\text{ }^\circ\text{C}$

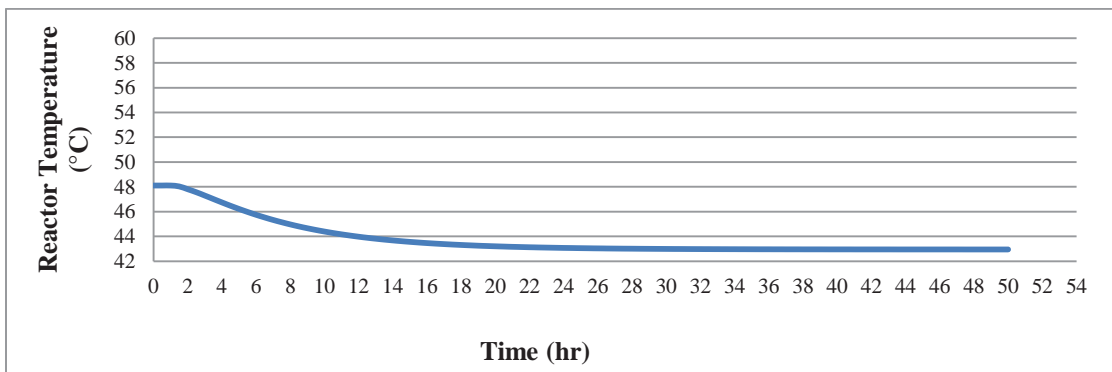


Figure 15 Reactor temperature change while oil inlet decrease for methanol/oil ratio = 10, $T_R = 48.1042\text{ }^\circ\text{C}$

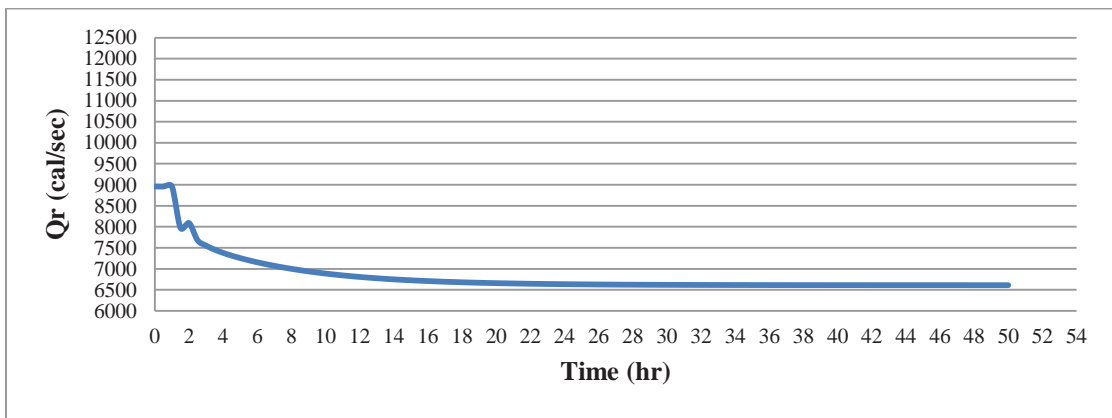


Figure 16 Heat inlet to reactor change while oil inlet decrease for methanol/oil ratio = 10, $T_R = 48.1042\text{ }^\circ\text{C}$

Change of reactor temperature and time constant are shown in Table 7. As expected, operation with the point 3 provides lesser change of reactor temperature

and faster response that corresponding with the controllability analysis of gain and poles (or time constant).

Table 7 Change reactor temperature (T_R , °C) and time constant (τ , hr)

Condition	ΔT_R	τ
Methanol/oil ratio = 6, $T_R = 59.0916$ °C	4.9408	4.5570
Methanol/oil ratio = 8, $T_R = 51.7996$ °C	4.4587	3.9092
Methanol/oil ratio = 10, $T_R = 48.1042$ °C	4.3642	3.8005

In addition, control performance of the system can be accounted from integral absolute error (IAE) as shown in equation 22. The time span for the calculation is from 1 to 14 hours. Note that the operating point 1 is the least controllable.

$$IAE = \sum_{t=i}^n |CV_t - \text{setpoint}| \quad (22)$$

Table 8 Integral absolute error (IAE) of transesterification reactor

Condition	IAE
Methanol/oil ratio = 6, $T_R = 59.0916$ °C	0.064624
Methanol/oil ratio = 8, $T_R = 51.7996$ °C	0.059139
Methanol/oil ratio = 10, $T_R = 48.1042$ °C	0.061427

5.2 Design and control of transesterification reactor connected with methanol recovery column

5.2.1 Design of transesterification reactor connected with methanol recovery column

Because dynamic model of distillation column is quite complex, only state-state controllability analysis is considered here. Furthermore, to make the result rigorous, Aspen Plus v7.3 will be used for the modeling task. RCSTR and RADFRAC will be used for modeling of transesterification reactor and methanol recovery column, respectively.

In the design of methanol recovery column, only operating variables including distillate rate and reboiler duty will be considered. Other design parameters such as number of stage, feed stage location will be followed the design of Zhang et al. (2003a)

Because the task of the column is to recover methanol from the product stream and hence the 0.01 methanol mole fraction at the bottom of column will be considered as design objective with distillation rate and reboiler duty as operating parameters. Tables 9 and 10 show the design conditions of methanol recovery column corresponding with the condition of transesterification reactor at points 1 and 2 in

section 5.1, respectively. Note that no feasible operation for the column corresponding with the point 3.

Table 9 Design condition of methanol recovery column for reactor condition: methanol/oil ratio 6 and reactor temperature 59.0916 °C

Distillate rate (kmol/hr)	Reboiler duty (cal/sec)
4	21,242.8491
3.9	24,521.0256
3.8	32,001.0228

Table 10 Design condition of methanol recovery column for reactor condition: methanol/oil ratio 8 and reactor temperature 51.7996 °C

Distillate rate (kmol/hr)	Reboiler duty (cal/sec)
6.3	37,757.8948
6.2	47,055.9827
6.1	78,370.2714

5.2.2 Controllability of transesterification reactor connected with methanol recovery column

Apart from the reactor, additional outputs and inputs corresponding to the column need to be defined. Methanol mole fraction at the bottom of the column is defined as controlled variable while distillate rate and reboiler duty are defined as input. The controllability consideration will be based on steady-state analysis. Important information requires is steady-state gain matrix that can be taken from Aspen Plus by changing the inputs and observing the outputs. The gain results are shown in Appendix A. The gain matrix will be used for the calculation of relative gain array (RGA) and singular value analysis (SVA).

RGA showed degree of control loop interaction if there is a nearly the identity matrix that control loop interaction is minimal (controllability also).

In singular value analysis (SVA), an operating windows need to be determined first. This is because SVA analysis depends strongly in the scaling (Skogestad and Postlethwaite, (2005)).

Scaling is very important in practical applications as it make model analysis and controller design much simpler. The scaling transfer functions as follow equation (48)

$$G = D_e^{-1} \hat{G} D_u \quad (48)$$

where G denotes scaling gain
 \hat{G} denotes unscaling gain
 D_u denotes largest allowed input change

D_e denotes largest allowed control error

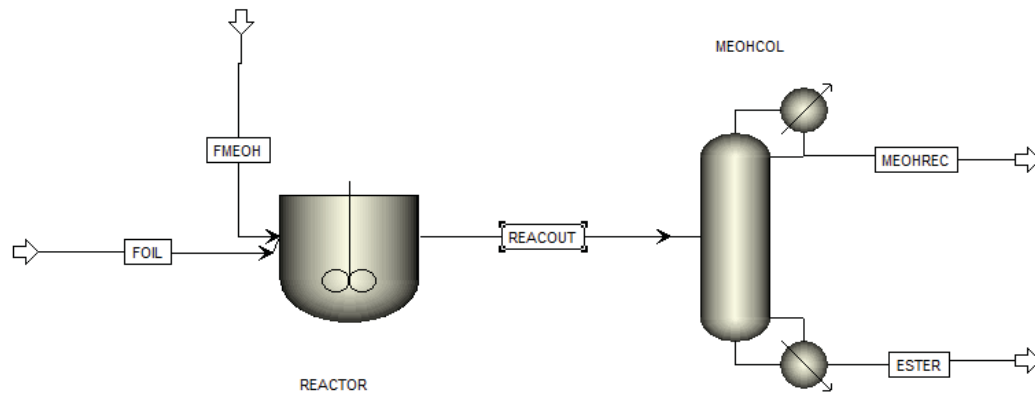


Figure 17 Transesterification reactor connected with methanol recovery column

The allowed input changes and control errors are defined as follow,

$$D_u = \begin{bmatrix} FR*0.1 & 0 & 0 & 0 \\ 0 & 2 & 0 & 0 \\ 0 & 0 & 0.2 & 0 \\ 0 & 0 & 0 & Q_{reb}*0.1 \end{bmatrix}$$

$$D_e = \begin{bmatrix} \text{conversion} \times 0.01 & 0 \\ 0 & x_{B,MeOH} * 0.1 \end{bmatrix}$$

Note that the diagonal elements D_u are corresponding with the allowed change of methanol/oil ratio, reactor temperature, distillate rate, and reboiler duty, respectively. The diagonal elements of D_e are corresponding with the allowed errors of conversion and mole fraction of the column bottom, respectively.

Based on the results in Tables 9 and 10, six cases are defined for the analysis,

Case 1: methanol/oil ratio = 6, $T_R = 59.0916$ °C, distillate rate (D) = 4 kmol/hr and reboiler duty (Q_{reb}) = 21,242.8491 cal/sec

Case 2: methanol/oil ratio = 6, $T = 59.0916$ °C, distillate rate = 3.9 kmol/hr and reboiler duty = 24,521.0256 cal/sec

Case 3: methanol/oil ratio = 6, $T = 59.0916$ °C, distillate rate = 3.8 kmol/hr and reboiler duty = 32,001.0228 cal/sec

Case 4: methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.3 kmol/hr and reboiler duty = 37,757.8948 cal/sec

Case 5: methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.2 kmol/hr and reboiler duty = 47,055.9827 cal/sec

Case 6: methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.1 kmol/hr and reboiler duty = 78,370.2714 cal/sec

For case 1, there are six possible pairings as listed in Table 11. However, based on the RGA result, suggested pairings are F_{MeOH} and D, F_{MeOH} and Q_{reb} , T_R and D, and T_R and Q_{reb} . Further analysis is required to determine the best pairing. Using SVA analysis, T_R and Q_{reb} should be selected due to its least condition number (CN).

Tables 11-16 show the results of RGA and SVA analysis for the other cases. Note that the best pairing for all cases is the pair T_R and Q_{reb} . The best possible condition number of each case is summarized in Table 17.

Table 11 RGA, SVA, and condition number (CN) for case 1

CV	MV	RGA	σ_{max}	σ_{min}	CN
Conversion and $x_{B,MeOH}$	F_{MeOH} and T_R	$\begin{bmatrix} 0.0143 & 0.9857 \\ 0.9857 & 0.0143 \end{bmatrix}$	64.5137	0.2255	286.0918
	F_{MeOH} and D	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	65.4139	0.0509	1,285.1454
	F_{MeOH} and Q_{reb}	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	64.5914	0.0154	4,194.2468
	T_R and D	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	10.8569	0.2218	48.9491
	T_R and Q_{reb}	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	3.3069	0.2175	15.2041
	D and Q_{reb}	$\begin{bmatrix} NaN & NaN \\ -Inf & -Inf \end{bmatrix}$	11.3086	0	Inf

Table 12 RGA, SVA, and condition number (CN) for case 2

CV	MV	RGA	σ_{max}	σ_{min}	CN
Conversion and $x_{B,MeOH}$	F_{MeOH} and D	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	2,503.7349	0.3071	8,152.8326
	F_{MeOH} and Q_{reb}	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	70.8132	0.0122	5,804.3607
	T_R and D	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	2,502.7346	0.2173	11,517.4165
	T_R and Q_{reb}	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	2.6392	0.2120	12.4491

Table 13 RGA, SVA, and condition number (CN) for case 3

CV	MV	RGA	σ_{\max}	σ_{\min}	CN
Conversion and $x_{B,MeOH}$	F_{MeOH} and D	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	2,504.6044	0.3071	8,155.6640
	F_{MeOH} and Q_{reb}	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	76.8612	0.0068	11,303.1176
	T_R and D	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	2,503.4254	0.2222	11,266.5400
	T_R and Q_{reb}	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	1.7894	0.2116	8.4565

Table 14 RGA, SVA, and condition number (CN) for case 4

CV	MV	RGA	σ_{\max}	σ_{\min}	CN
Conversion and $x_{B,MeOH}$	F_{MeOH} and D	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	2,502.0533	0.3071	8,147.3569
	F_{MeOH} and Q_{reb}	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	70.8165	0.0115	6,157.9565
	T_R and D	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	2,501.0524	0.2495	10,024.2581
	T_R and Q_{reb}	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	2.7213	0.2421	11.2404

Table 15 RGA, SVA, and condition number (CN) for case 5

CV	MV	RGA	σ_{\max}	σ_{\min}	CN
Conversion and $x_{B,MeOH}$	F_{MeOH} and D	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	2,505.1093	0.2057	12,178.4604
	F_{MeOH} and Q_{reb}	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	100.4010	0.0093	10,795.8065
	T_R and D	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	2,503.1008	0.2489	10,059.5151
	T_R and Q_{reb}	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	2.6392	0.2120	12.4481

From the result in this work, the best operating condition is methanol/oil ratio = 8, reactor temperature = 51.7996 °C, distillate rate = 6.1 kmol/hr, and reboiler duty = 78,370.2714 cal/sec with condition number = 6.0439

Table 16 RGA, SVA, and condition number (CN) for case 6

CV	MV	RGA	σ_{\max}	σ_{\min}	CN
Conversion and $x_{B,MeOH}$	F_{MeOH} and D	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	2,506.0585	0.2057	12,183.07486
	F_{MeOH} and Q_{reb}	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	106.0640	0.0016	66,290
	T_R and D	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	2,503.8133	0.2489	10,059.5151
	T_R and Q_{reb}	$\begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix}$	1.1290	0.1868	6.0439

Table 17 Condition number of six cases

MeOH/oil ratio	T_R (°C)	D (kmol/hr)	Q_{reb} (cal/sec)	CN
6	59.0916	4	21,242.8491	15.2041
		3.9	24,521.0256	12.4491
		3.8	32,001.0228	8.4565
8	51.7996	6.3	37,757.8948	11.2404
		6.2	47,055.9827	12.4481
		6.1	78,370.2714	6.0439

5.2.3 Dynamic control of transesterification reactor connected with methanol recovery column

Oil feed flow rate is defined as disturbance variable. Oil conversion and methanol mole fraction at column bottom are defined as controlled variables while reactor temperature and reboiler duty are defined as manipulated variables. Other control loops are oil federate, reactor level, methanol/oil molar ratio, column pressure, reflux drum level, and column level loops.

Dynamic simulation is used to validate the analysis results of the six cases considered in the previous section. The results are shown Figures 19-48. The oil federate is changed after one hour from 1.1858 to 1.0650 kmol/hr. The setpoints of the reactor conversion and the methanol mole fraction of the column are 0.98 and 0.01, respectively.

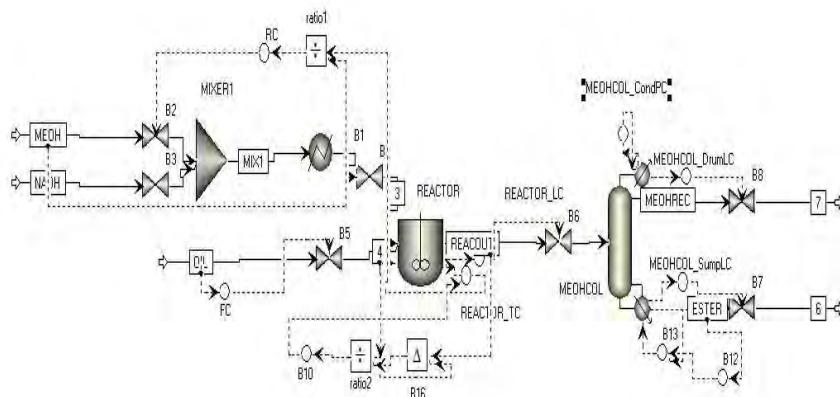


Figure 18 Control structure for optimal operation of the transesterification reactor connected with methanol recovery column

Case 1: methanol/oil ratio = 6, $T_R = 59.0916$ °C, distillate rate (D) = 4 kmol/hr and reboiler duty (Q_{reb}) = 21,242.8491 cal/sec

Figures 19 – 23 show the dynamic results from change of oil feedrate after 1 hour. The conversion reaches 0.981 ($\pm 0.1\%$ of the setpoint) in 11.5 hours and the reactor temperature change from 59.0916 °C to 54.1279 °C. The methanol mole fraction at the column bottom reaches 0.01 in 6 hours and reboiler duty change from 21,242.8491 to 20,659.0769 cal/sec.

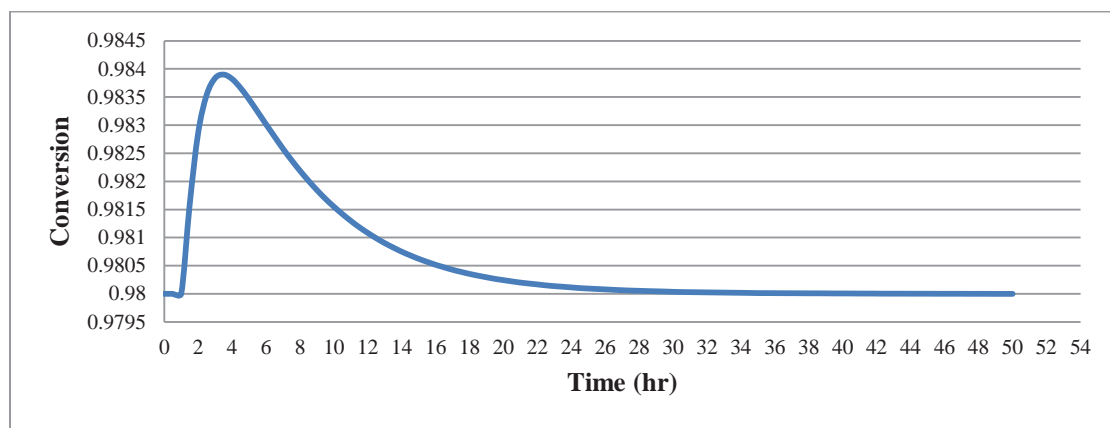


Figure 19 Effect of oil inlet to conversion for methanol/oil ratio = 6, $T_R = 59.0916$ °C, distillate rate = 4 kmol/hr and reboiler duty = 21,242.8491 cal/sec

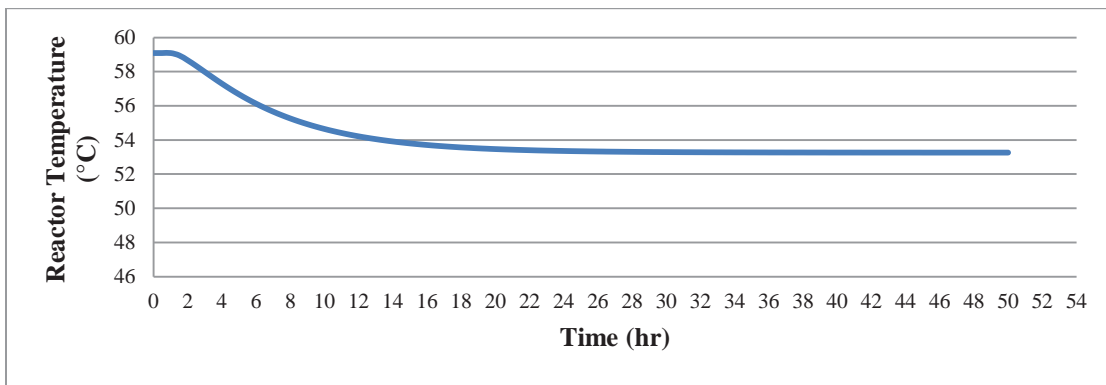


Figure 20 Reactor temperature change while oil inlet decrease for methanol/oil ratio = 6, $T_R = 59.0916$ °C, distillate rate = 4 kmol/hr and reboiler duty = 21,242.8491cal/sec

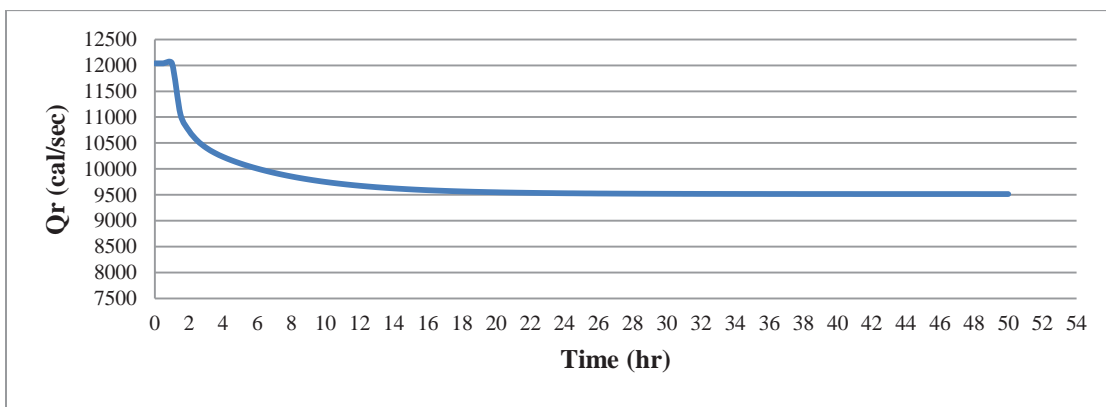


Figure 21 Heat inlet to reactor change while oil inlet decrease for methanol/oil ratio = 6, $T_R = 59.0916$ °C, distillate rate = 4 kmol/hr and reboiler duty = 21,242.8491cal/sec

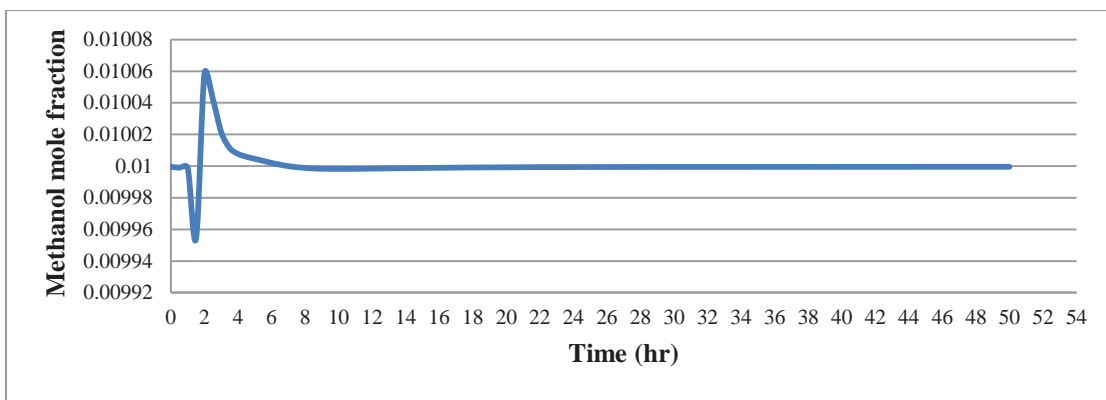


Figure 22 Effect of oil inlet to mole fraction of methanol in bottom of column for methanol/oil ratio = 6, $T_R = 59.0916$ °C, distillate rate = 4 kmol/hr and reboiler duty = 21,242.8491cal/sec

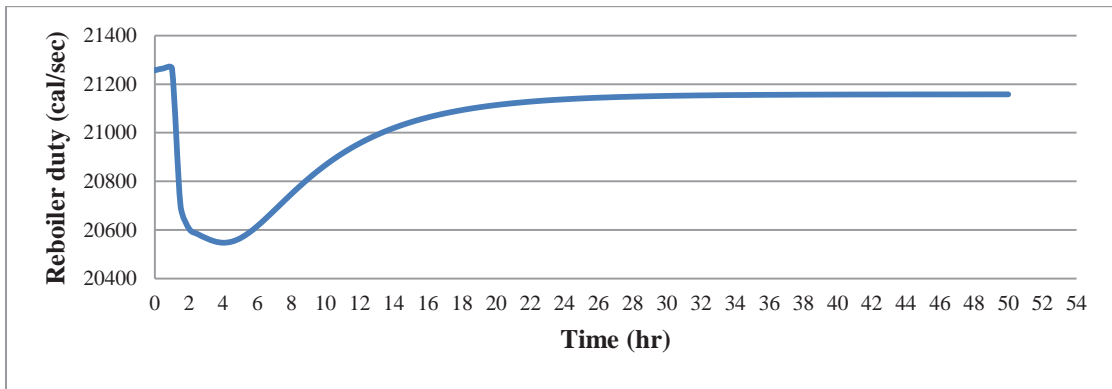


Figure 23 Reboiler duty change while oil inlet decrease for methanol/oil ratio = 6, $T_R = 59.0916$ °C, distillate rate = 4 kmol/hr and reboiler duty = 21,242.8491 cal/sec

Case 2: methanol/oil ratio = 6, $T = 59.0916$ °C, distillate rate = 3.9 kmol/hr and reboiler duty = 24,521.0256 cal/sec

Figures 24 – 28 show the dynamic results from change of oil feedrate after 1 hour. The conversion reaches 0.981 ($\pm 0.1\%$ of the setpoint) in 11.5 hours and the reactor temperature change from 59.0916 °C to 54.1512 °C. The methanol mole fraction at the column bottom reaches 0.01 in 7 hours and reboiler duty change from 24,521.0256 to 24,065.0553 cal/sec.

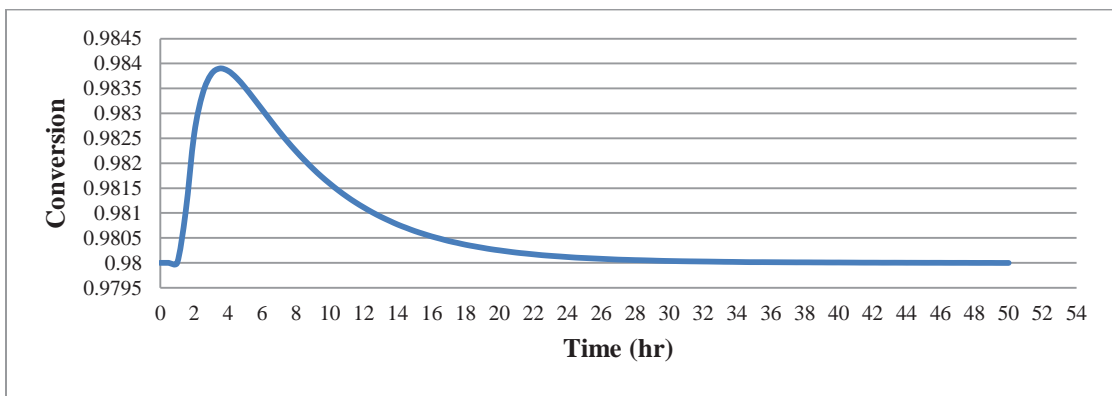


Figure 24 Effect of oil inlet to conversion for methanol/oil ratio = 6, $T_R = 59.0916$ °C, distillate rate = 3.9 kmol/hr and reboiler duty = 24,521.0256 cal/sec

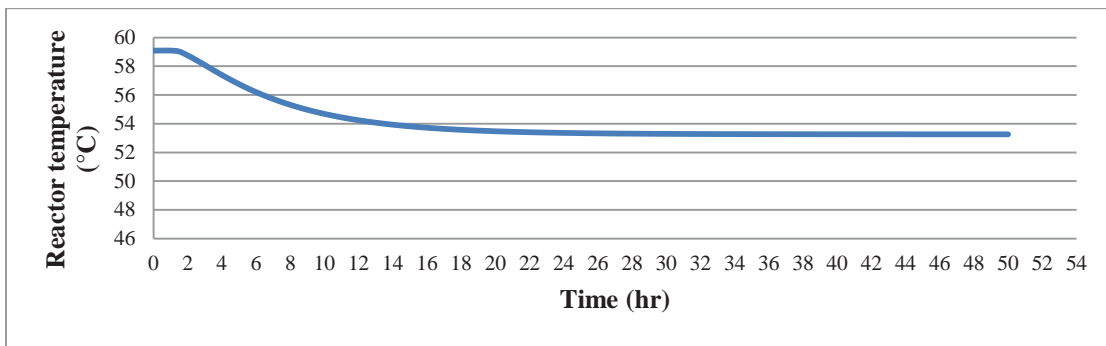


Figure 25 Reactor temperature change while oil inlet decrease for methanol/oil ratio 6, $T_R = 59.0916$ °C, distillate rate = 3.9 kmol/hr and reboiler duty = 24,521.0256 cal/sec

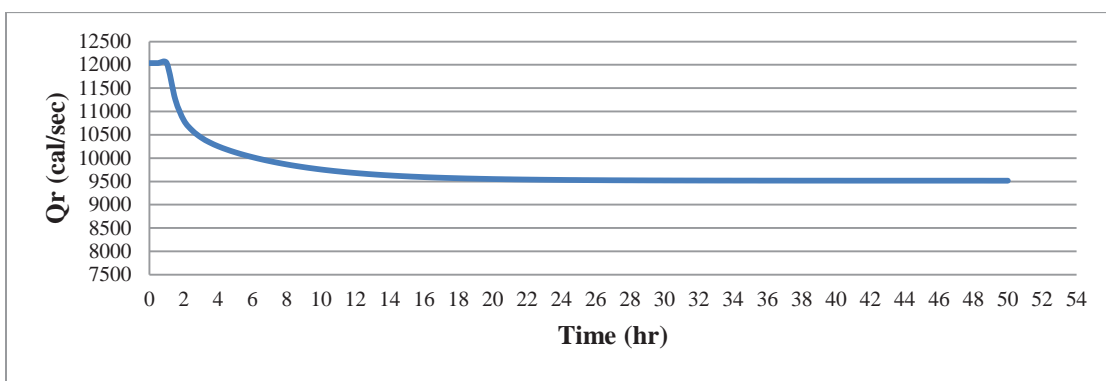


Figure 26 Heat inlet to reactor change while oil inlet decrease for methanol/oil ratio 6, $T_R = 59.0916$ °C, distillate rate = 3.9 kmol/hr and reboiler duty = 24,521.0256 cal/sec

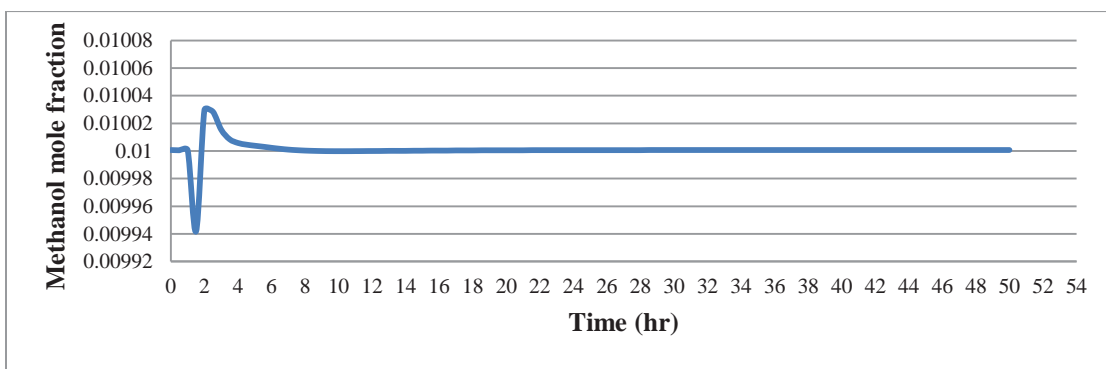


Figure 27 Effect of oil inlet to mole fraction of methanol in bottom of column for methanol/oil ratio = 6, $T_R = 59.0916$ °C, distillate rate = 3.9 kmol/hr and reboiler duty = 24,521.0256 cal/sec

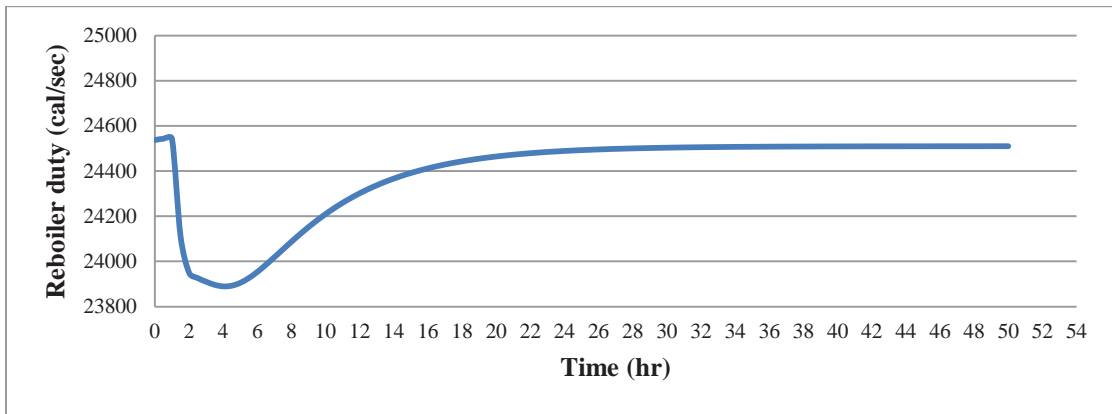


Figure 28 Reboiler duty change while oil inlet decrease for methanol/oil ratio 6, $T_R = 59.0916\text{ }^\circ\text{C}$, distillate rate = 3.9 kmol/hr and reboiler duty = 24,521.0256 cal/sec

Case 3: methanol/oil ratio = 6, $T = 59.0916\text{ }^\circ\text{C}$, distillate rate = 3.8 kmol/hr and reboiler duty = 32,001.0228 cal/sec

Figures 29 – 33 show the dynamic results from change of oil feedrate after 1 hour. The conversion reaches 0.981 ($\pm 0.1\%$ of the setpoint) in 11.5 hours and the reactor temperature change from 59.0916 $^\circ\text{C}$ to 54.1118 $^\circ\text{C}$. The methanol mole fraction at the column bottom reaches 0.01 in 6 hours and reboiler duty change from 32,001.0228 to 31,607.0612 cal/sec.

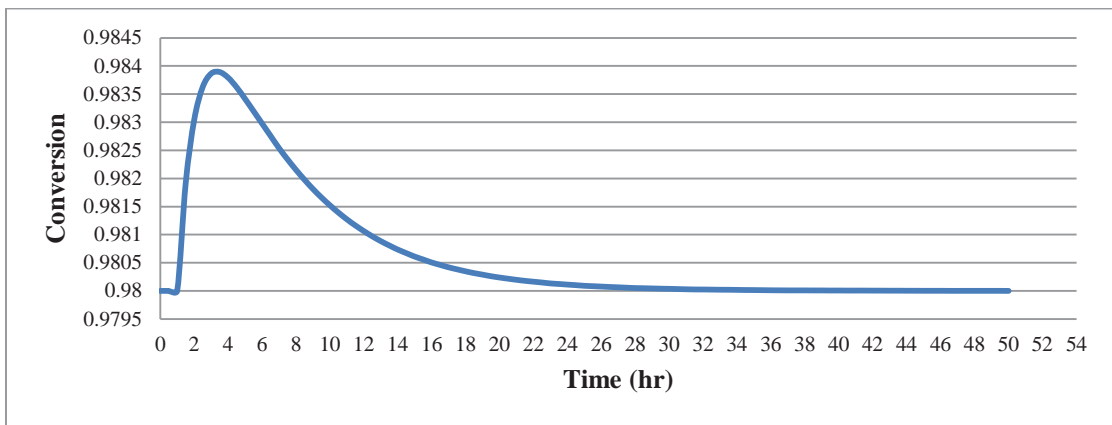


Figure 29 Effect of oil inlet to conversion for methanol/oil ratio = 6, $T_R = 59.0916\text{ }^\circ\text{C}$, distillate rate = 3.8 kmol/hr and reboiler duty = 32,001.0228 cal/sec

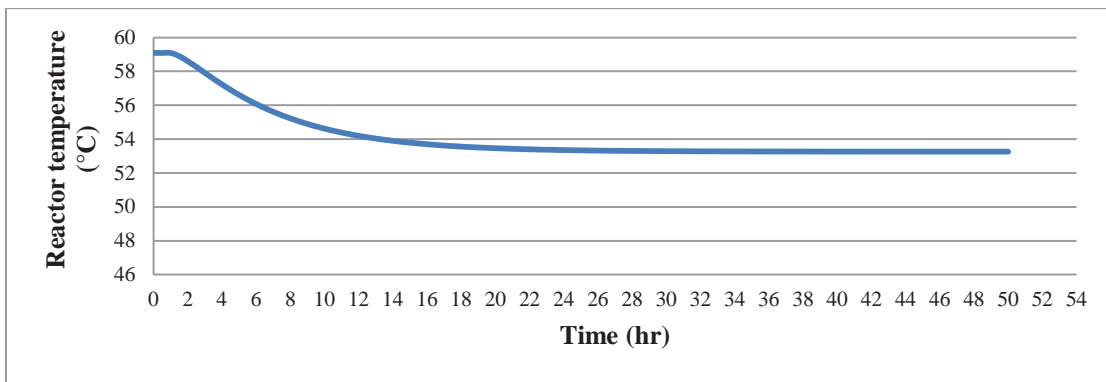


Figure 30 Reactor temperature change while oil inlet decrease for methanol/oil ratio = 6, $T_R = 59.0916$ °C, distillate rate = 3.8 kmol/hr and reboiler duty = 32,001.0228 cal/sec

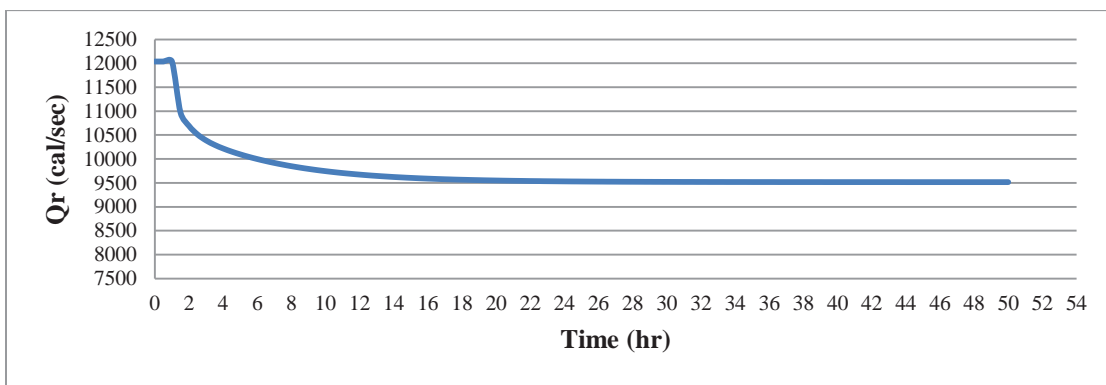


Figure 31 Heat inlet to reactor change while oil inlet decrease for methanol/oil ratio = 6, $T_R = 59.0916$ °C, distillate rate = 3.8 kmol/hr and reboiler duty = 32,001.0228 cal/sec

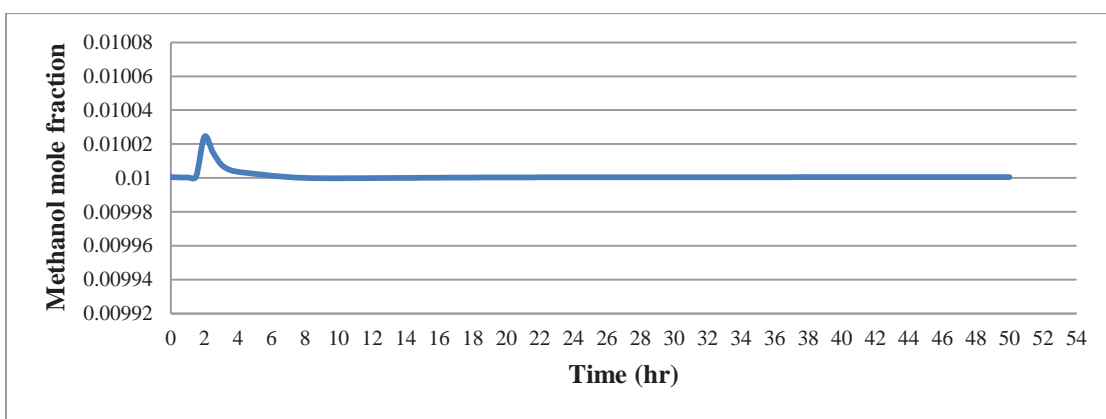


Figure 32 Effect of oil inlet to mole fraction of methanol in bottom of column for methanol/oil ratio = 6, $T_R = 59.0916$ °C, distillate rate = 3.8 kmol/hr and reboiler duty = 32,001.0228 cal/sec

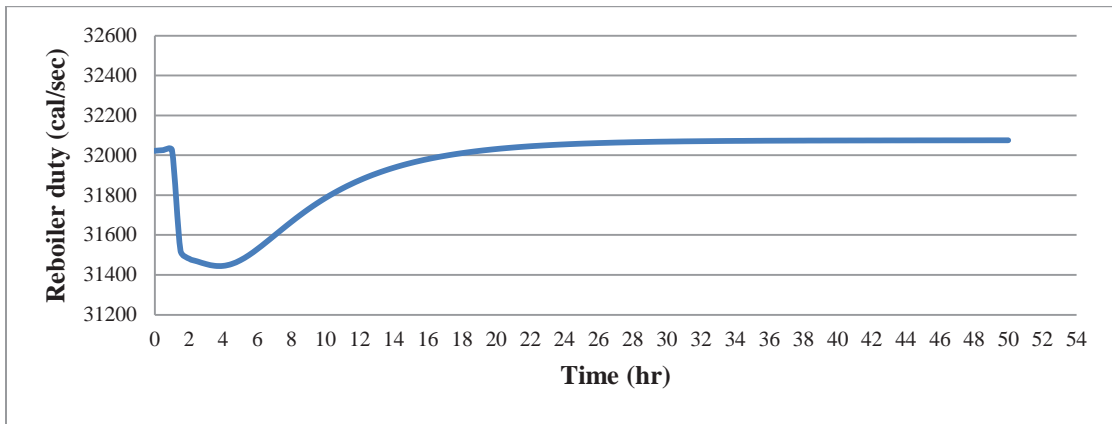


Figure 33 Reboiler duty change while oil inlet decrease for methanol/oil ratio = 6, $T_R = 59.0916$ °C, distillate rate = 3.8 kmol/hr and reboiler duty = 32,001.0228 cal/sec

Case 4: methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.3 kmol/hr and reboiler duty = 37,757.8948 cal/sec

Figures 34 – 38 show the dynamic results from change of oil feedrate after 1 hour. The conversion reaches 0.981 ($\pm 0.1\%$ of the setpoint) in 12 hours and the reactor temperature change from 51.7996 °C to 47.3105 °C. The methanol mole fraction at the column bottom reaches 0.01 in 7.5 hours and reboiler duty change from 37,757.8948 to 36,633.8368 cal/sec.

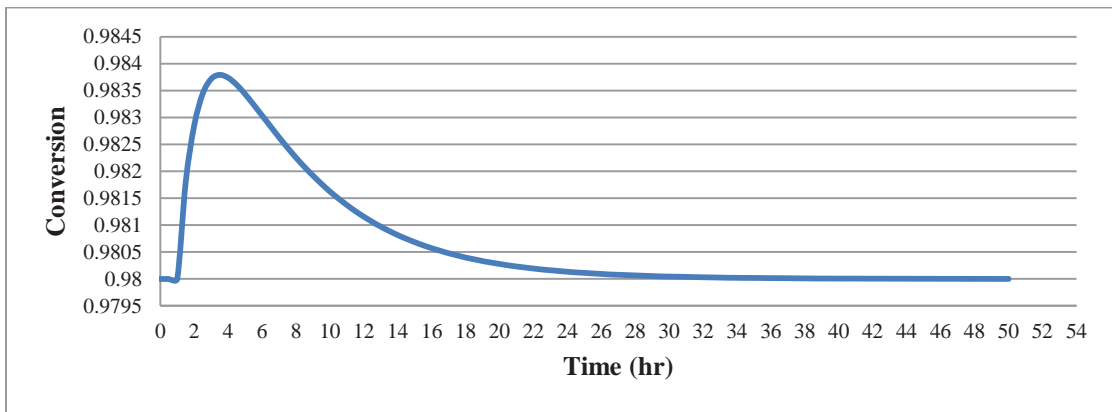


Figure 34 Effect of oil inlet to conversion for methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.3 kmol/hr and reboiler duty = 37,757.8948 cal/sec

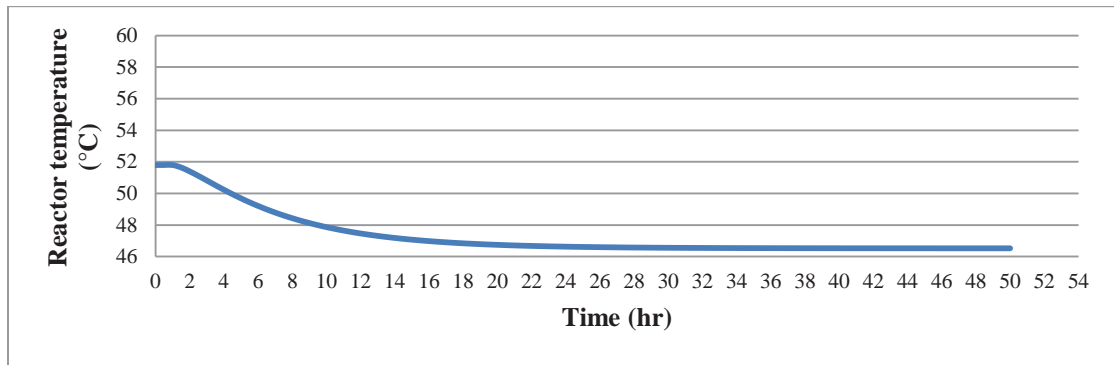


Figure 35 Reactor temperature change while oil inlet decrease for methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.3 kmol/hr and reboiler duty = 37,757.8948 cal/sec

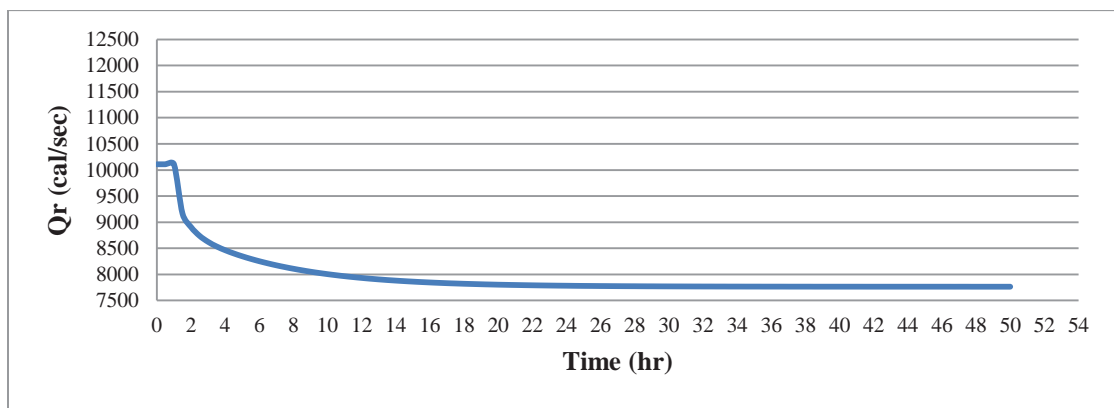


Figure 36 Heat inlet to reactor change while oil inlet decrease for methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.3 kmol/hr and reboiler duty = 37,757.8948 cal/sec

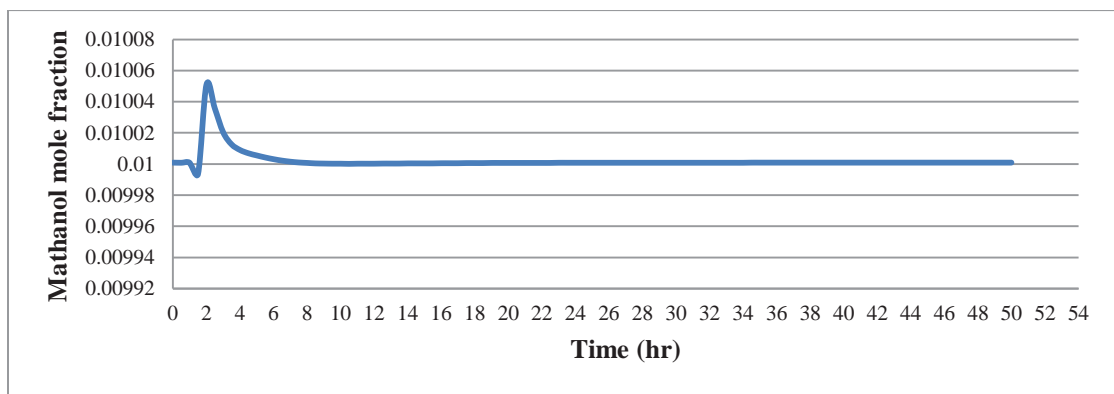


Figure 37 Effect of oil inlet to mole fraction of methanol in bottom of column for methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.3 kmol/hr and reboiler duty = 37,757.8948 cal/sec

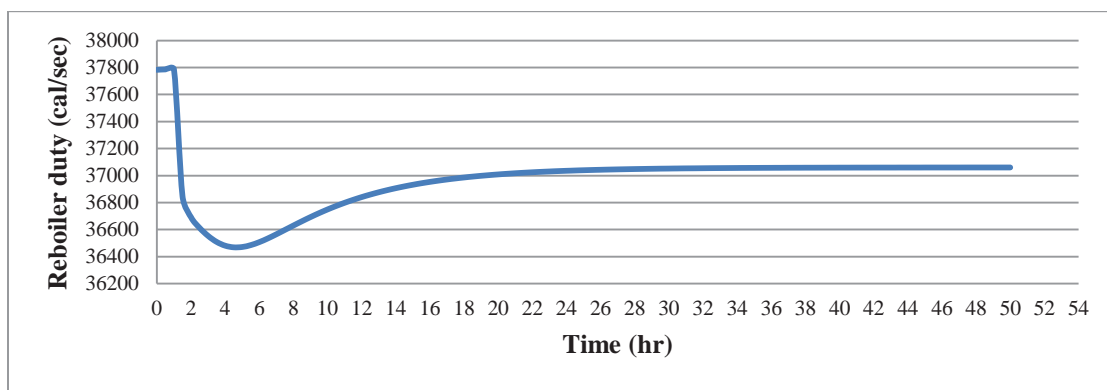


Figure 38 Reboiler duty change while oil inlet decrease for methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.3 kmol/hr and reboiler duty = 37,757.8948 cal/sec

Case 5: methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.2 kmol/hr and reboiler duty = 47,055.9827 cal/sec

Figures 39 – 43 show the dynamic results from change of oil feedrate after 1 hour. The conversion reaches 0.981 ($\pm 0.1\%$ of the setpoint) in 12 hours and the reactor temperature change from 51.7996°C to 47.3134°C. The methanol mole fraction at the column bottom reaches 0.01 in 6.5 hours and reboiler duty change from 47,055.9827 to 45,936.1727 cal/sec.

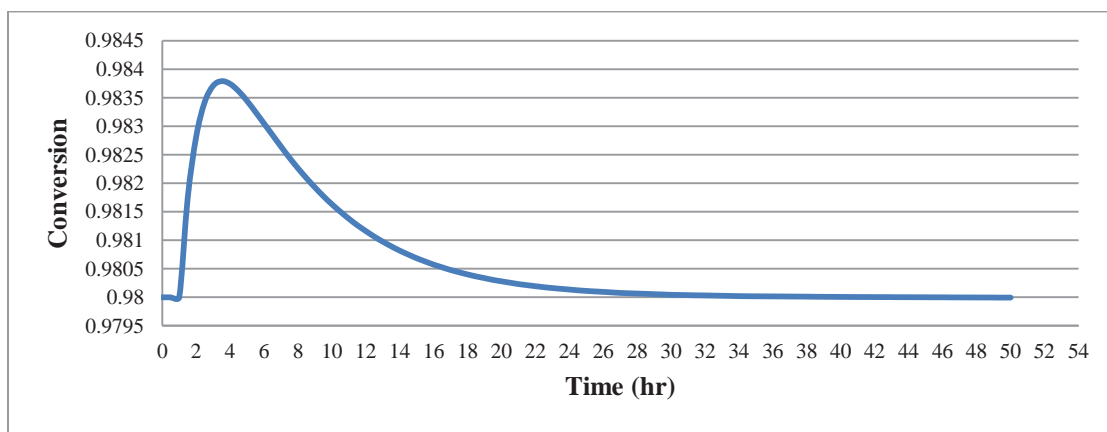


Figure 39 Effect of oil inlet to conversion for methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.2 kmol/hr and reboiler duty = 47,055.9827 cal/sec

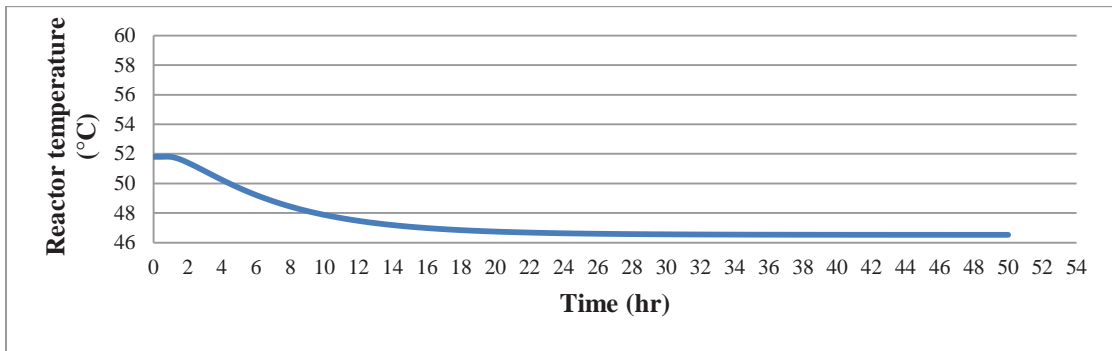


Figure 40 Reactor temperature change while oil inlet decrease for methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.2 kmol/hr and reboiler duty = 47,055.9827 cal/sec

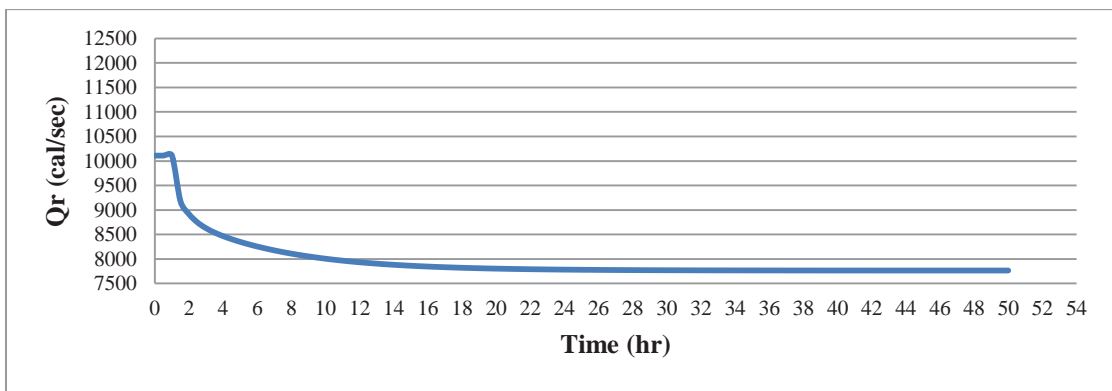


Figure 41 Heat inlet to reactor change while oil inlet decrease for methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.2 kmol/hr and reboiler duty = 47,055.9827 cal/sec

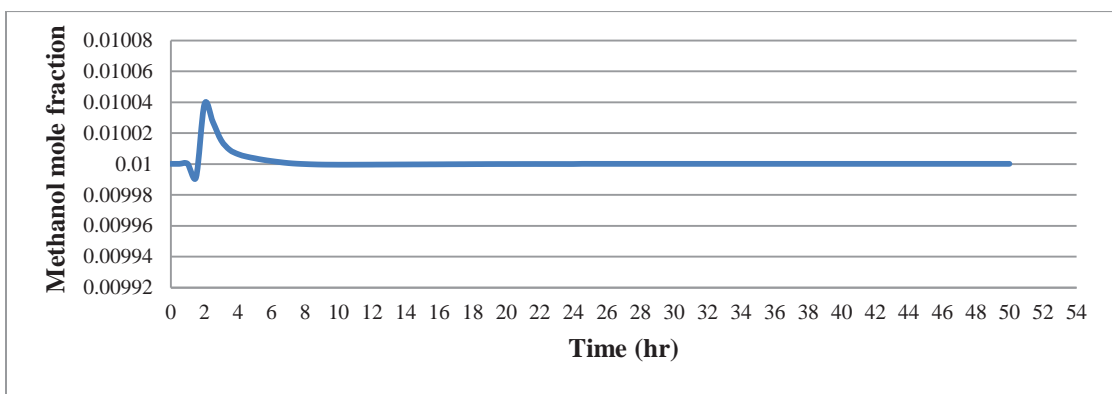


Figure 42 Effect of oil inlet to mole fraction of methanol in bottom of column for methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.2 kmol/hr and reboiler duty = 47,055.9827 cal/sec

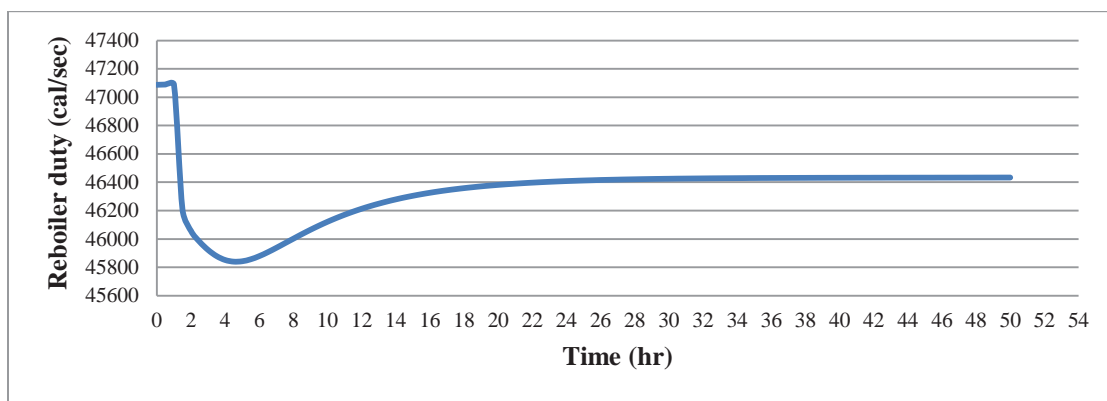


Figure 43 Reboiler duty change while oil inlet decrease for methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.2 kmol/hr and reboiler duty = 47,055.9827 cal/sec

Case 6: methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.1 kmol/hr and reboiler duty = 78,370.2714 cal/sec

Figures 44 – 48 show the dynamic results from change of oil feedrate after 1 hour. The conversion reaches 0.981 ($\pm 0.1\%$ of the setpoint) in 12 hours and the reactor temperature change from 51.7996°C to 47.3134°C. The methanol mole fraction at the column bottom reaches 0.01 in 6 hours and reboiler duty change from 78,370.2714 to 77,301.3834 cal/sec.

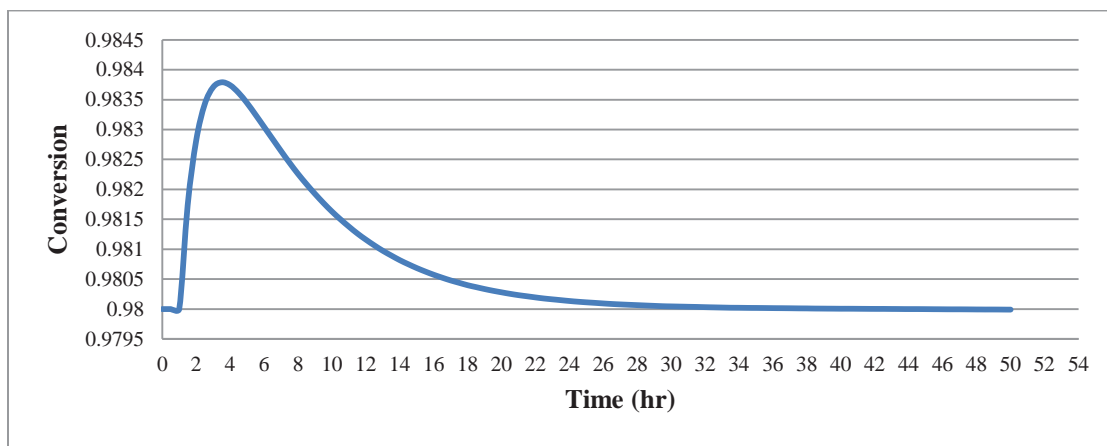


Figure 44 Effect of oil inlet to conversion for methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.1 kmol/hr and reboiler duty = 78,370.2714 cal/sec

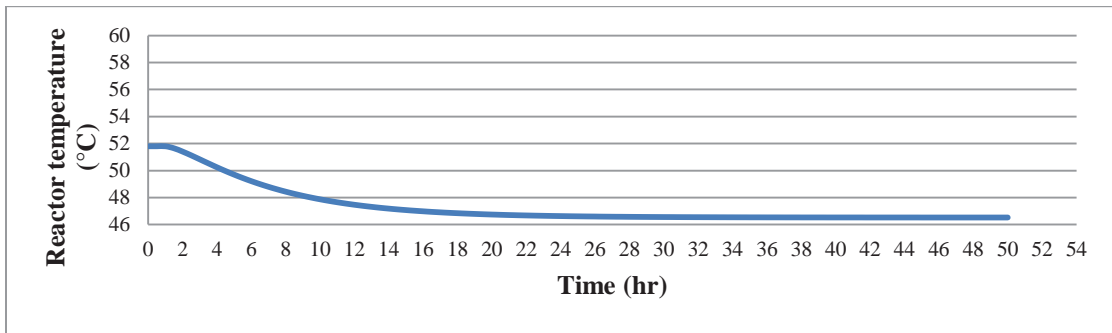


Figure 45 Reactor temperature change while oil inlet decrease for methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.1 kmol/hr and reboiler duty = 78,370.2714 cal/sec

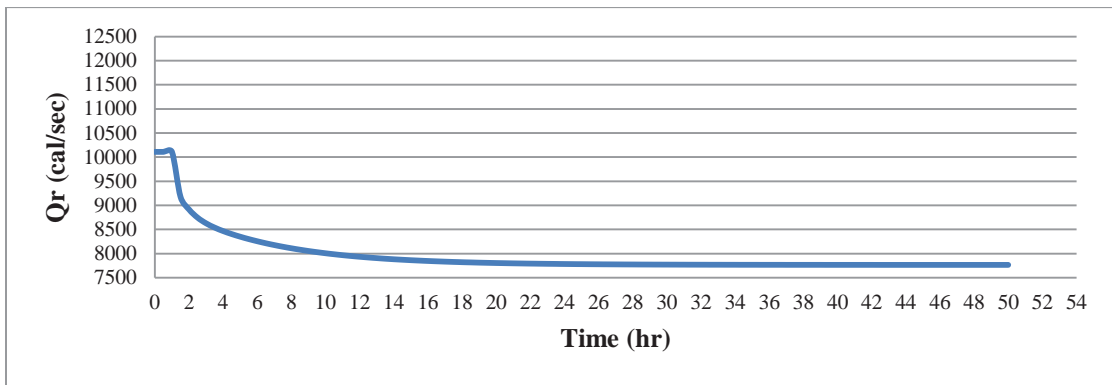


Figure 46 Heat inlet to reactor change while oil inlet decrease for methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.1 kmol/hr and reboiler duty = 78,370.2714 cal/sec

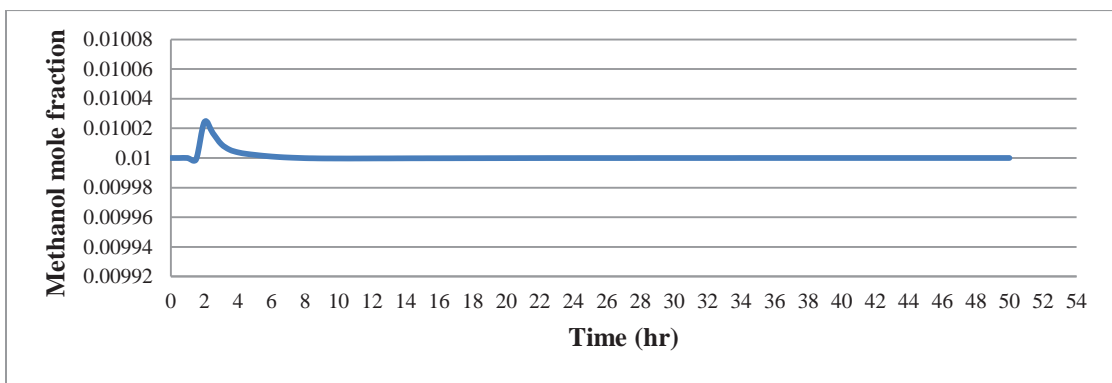


Figure 47 Effect of oil inlet to mole fraction of methanol in bottom of column for methanol/oil ratio = 8, $T_R = 51.7996$ °C, distillate rate = 6.1 kmol/hr and reboiler duty = 78,370.2714 cal/sec

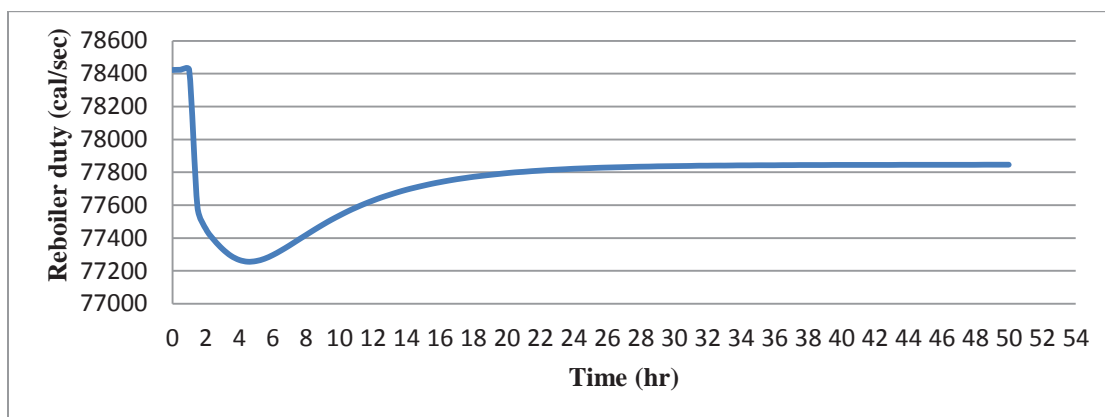


Figure 48 Reboiler duty change while oil inlet decrease for methanol/oil ratio= 8, $T_R = 51.7996$ °C, distillate rate = 6.1 kmol/hr and reboiler duty = 78,370.2714 cal/sec

Table 18 summarizes change of reactor temperature and reboiler duty for the control and Table 19 summarizes the IAE for the operation with all conditions. Note that there is not much different for the IAE results.

Table 18 Change of reactor temperature and reboiler duty for control

Conditions				ΔT_R (°C)	ΔQ_{reb} (cal/sec)
Methanol/oil ratio	T_R (°C)	D (kmol/hr)	Q_{reb} (cal/sec)		
6	59.0916	4.0	21,242.8491	4.9637	583.7722
		3.9	24,521.0256	4.9404	455.9703
		3.8	32,001.0228	4.4862	393.9616
8	51.7996	6.3	37,757.8948	4.4891	1,124.0580
		6.2	47,055.9827	4.4862	1,119.8100
		6.1	78,370.2714	4.4862	1.068.8800

Table 19 IAE of transesterification reactor connected with methanol recovery column process

Condition				IAE	
Methanol/oil ratio	T_R (°C)	D (kmol/hr)	Q_{reb} (cal/sec)	Conversion of oil	$X_{B,MeOH}$
6	59.0916	4.0	21,242.8491	0.0568	2.0737×10^{-4}
		3.9	24,521.0256	0.0565	1.6300×10^{-4}
		3.8	32,001.0228	0.0569	6.6910×10^{-5}
8	51.7996	6.3	37,757.8948	0.0575	1.6000×10^{-4}
		6.2	47,055.9827	0.0575	1.2100×10^{-4}
		6.1	78,370.2714	0.0575	6.9590×10^{-5}

CHAPTER 6

CONCLUSIONS

Controllability is an inherent property of process. Poor design can make the plant uncontrollable. This research focuses on controllability analysis of alkali-catalyzed biodiesel process, in particular, transesterification section. The research is divided as two parts. The first part concerns design and control of transesterification reactor. Dynamic model of the reactor is developed and used for analysis of effects of important design parameters including residence time, reactor temperature, and methanol/oil molar ratio to conversion and controllability. The controllability is analyzed based on poles, zeros, and gain. Note that the reactor is stable.

The second part concerns design and control of the reactor connected with a methanol recovery column. To avoid difficulty of the modeling, Aspen Plus is chosen for the steady-state modeling of the system. Because the system is multivariable, relative gain array and singular value analysis are used as controllability analysis tools. Effects of design parameters including reactor temperature, methanol/oil molar ratio, distillate rate, and reboiler duty to the reactor conversion and methanol mole fraction and controllability are analyzed. Dynamic simulation shows there is not much difference on the control performance for each operation.

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APPENDIX

APPENDIX A

GAIN

Gain

Table A1 Gain of methanol/oil feed flowrate (F_{MeOH}), reactor temperature (T_R), distillate rate and reboiler duty to conversion and mole fraction of methanol in the bottom of case 1

CV\MV	F_{MeOH}	T_R	D	Q_{reb}
Conversion	0.3074	0.2222	0	0
$x_{\text{B,MeOH}}$	64.5095	-0.6785	-10.8357	-3.2362

Table A2 Gain of methanol/oil feed flowrate (F_{MeOH}), reactor temperature (T_R), distillate rate and reboiler duty to conversion and mole fraction of methanol in the bottom of case 2

CV\MV	F_{MeOH}	T_R	D	Q_{reb}
Conversion	0.3072	0.2173	0	0
$x_{\text{B,MeOH}}$	70.7657	-0.5772	-2,502.7345	-2.5748

Table A3 Gain of methanol/oil feed flowrate (F_{MeOH}), reactor temperature (T_R), distillate rate and reboiler duty to conversion and mole fraction of methanol in the bottom of case 3

CV\MV	F_{MeOH}	T_R	D	Q_{reb}
Conversion	0.3072	0.2222	0	0
$x_{\text{B,MeOH}}$	76.8417	-0.5432	-2,503.4254	-1.7036

Table A4 Gain of methanol/oil feed flowrate (F_{MeOH}), reactor temperature (T_R), distillate rate and reboiler duty to conversion and mole fraction of methanol in the bottom of case 4

CV\MV	F_{MeOH}	T_R	D	Q_{reb}
Conversion	0.3072	0.2495	0	0
$x_{\text{B,MeOH}}$	70.7666	-0.6568	-2,501.0524	-2.6401

Table A5 Gain of methanol/oil feed flowrate (F_{MeOH}), reactor temperature (T_R), distillate rate and reboiler duty to conversion and mole fraction of methanol in the bottom of case 5

CV\MV	F_{MeOH}	T_R	D	Q_{reb}
Conversion	0.2059	0.2489	0	0
$x_{\text{B,MeOH}}$	100.2979	-0.6435	-2,503.1007	-4.5449

Table A6 Gain of methanol/oil feed flowrate (F_{MeOH}), reactor temperature (T_R), distillate rate and reboiler duty to conversion and mole fraction of methanol in the bottom of case 6

CV\MV	F_{MeOH}	T_R	D	Q_{reb}
Conversion	0.2059	0.2489	0	0
$x_{B,\text{MeOH}}$	106.0604	-0.7276	-2,503.8132	-0.8475

APPENDIX B
State Space Model

State Space Model

Linearization of equation (13) – (18)

$$\begin{aligned}\frac{dC_{TG}}{dt} &= \frac{F_{TG}C_{TG,in} - F_R C_{TG}}{V} - r_1 = f_1 \\ \frac{dC_{DG}}{dt} &= \frac{-F_R C_{DG}}{V} + (r_1 + r_2) = f_2 \\ \frac{dC_{MG}}{dt} &= \frac{-F_R C_{MG}}{V} + (r_1 - r_3) = f_3 \\ \frac{dC_{MeOH}}{dt} &= \frac{F_{MeOH}C_{MeOH,in} - F_R C_{MeOH}}{V} - (r_1 + r_2 + r_3) = f_4 \\ \frac{dC_{FAME}}{dt} &= \frac{-F_R C_{FAME}}{V} + (r_1 + r_2 + r_3) \\ \frac{dC_{GL}}{dt} &= \frac{-F_R C_{GL}}{V} + r_3\end{aligned}$$

$$\frac{\partial f_1}{\partial C_{TG}} = a_{11} = \frac{-F_R}{V} - k_1 f^* C_{MeOH}$$

$$\frac{\partial f_2}{\partial C_{TG}} = a_{12} = k_1 r^* C_{FAME}$$

$$\frac{\partial f_3}{\partial C_{TG}} = a_{13} = 0$$

$$\frac{\partial f_4}{\partial C_{TG}} = a_{14} = -k_1 f C_{TG}$$

$$\frac{\partial f_5}{\partial C_{TG}} = a_{15} = k_1 r C_{DG}$$

$$\frac{\partial f_6}{\partial C_{TG}} = a_{16} = 0$$

$$\frac{\partial f_1}{\partial C_{DG}} = a_{21} = k_1 f C_{MeOH}$$

$$\frac{\partial f_2}{\partial C_{DG}} = a_{22} = \frac{-F_R}{V} - k_1 r C_{FAME} - k_2 f C_{MeOH}$$

$$\frac{\partial f_3}{\partial C_{DG}} = a_{23} = k_2 r C_{FAME}$$

$$\frac{\partial f_4}{\partial C_{DG}} = a_{24} = k_1 f C_{TG} - k_2 f C_{DG}$$

$$\frac{\partial f_5}{\partial C_{DG}} = a_{25} = -k_1 r_{C_{DG}} + k_2 r_{C_{MG}}$$

$$\frac{\partial f_6}{\partial C_{DG}} = a_{26} = 0$$

$$\frac{\partial f_1}{\partial C_{MG}} = a_{31} = 0$$

$$\frac{\partial f_2}{\partial C_{MG}} = a_{32} = k_2 f_{C_{MeOH}}$$

$$\frac{\partial f_3}{\partial C_{MG}} = a_{33} = \frac{-F_R}{V} - k_2 r_{C_{FAME}} - k_3 f_{C_{MeOH}}$$

$$\frac{\partial f_4}{\partial C_{MG}} = a_{34} = k_2 f_{C_{DG}} - k_3 f_{C_{MG}}$$

$$\frac{\partial f_5}{\partial C_{MG}} = a_{35} = -k_2 r_{C_{MG}} + k_3 r_{C_{GL}}$$

$$\frac{\partial f_6}{\partial C_{MG}} = a_{36} = k_3 r_{C_{FAME}}$$

$$\frac{\partial f_1}{\partial C_{MeOH}} = a_{41} = -k_1 f_{C_{MeOH}}$$

$$\frac{\partial f_2}{\partial C_{MeOH}} = a_{42} = k_1 r_{C_{FAME}} - k_2 f_{C_{MeOH}}$$

$$\frac{\partial f_3}{\partial C_{MeOH}} = a_{43} = k_2 r_{C_{FAME}} - k_3 f_{C_{MeOH}}$$

$$\frac{\partial f_4}{\partial C_{MeOH}} = a_{44} = \frac{-F_R}{V} - (k_1 f_{C_{TG}} + k_2 f_{C_{DG}} + k_3 f_{C_{MG}})$$

$$\frac{\partial f_5}{\partial C_{MeOH}} = a_{45} = k_1 r_{C_{DG}} + k_2 r_{C_{MG}} + k_3 r_{C_{GL}}$$

$$\frac{\partial f_6}{\partial C_{MeOH}} = a_{46} = k_3 r_{C_{FAME}}$$

$$\frac{\partial f_1}{\partial C_{FAME}} = a_{51} = k_1 f_{C_{MeOH}}$$

$$\frac{\partial f_2}{\partial C_{FAME}} = a_{52} = -k_1 r C_{FAME} + k_2 f C_{MeOH}$$

$$\frac{\partial f_3}{\partial C_{FAME}} = a_{53} = -k_2 r C_{FAME} + k_3 f C_{MeOH}$$

$$\frac{\partial f_4}{\partial C_{FAME}} = a_{54} = k_1 f C_{TG} + k_2 f C_{DG} + k_3 f C_{MG}$$

$$\frac{\partial f_5}{\partial C_{FAME}} = a_{55} = \frac{-F_R}{V} - (k_1 r C_{DG} + k_2 r C_{MG} + k_3 r C_{GL})$$

$$\frac{\partial f_6}{\partial C_{FAME}} = a_{56} = -k_3 r C_{FAME}$$

$$\frac{\partial f_1}{\partial C_{GL}} = a_{61} = 0$$

$$\frac{\partial f_2}{\partial C_{GL}} = a_{62} = 0$$

$$\frac{\partial f_3}{\partial C_{GL}} = a_{63} = k_3 f C_{MeOH}$$

$$\frac{\partial f_4}{\partial C_{GL}} = a_{64} = k_3 f C_{MG}$$

$$\frac{\partial f_5}{\partial C_{GL}} = a_{65} = -k_3 f C_{GL}$$

$$\frac{\partial f_6}{\partial C_{GL}} = a_{66} = \frac{-F_R}{V} - k_3 r C_{FAME}$$

$$\begin{aligned} \frac{\partial f_1}{\partial T} = bu_{11} = & -C_{TG} C_{MeOH} \left(A_1 f \exp\left(\frac{-E_1 f}{RT}\right) \right) \left(\frac{E_1 f}{RT} + 1 \right) \\ & + C_{DG} C_{FAME} \left(A_1 r \exp\left(\frac{-E_1 r}{RT}\right) \right) \left(\frac{E_1 r}{RT} + 1 \right) \end{aligned}$$

$$\frac{\partial f_1}{\partial F_{MeOH}} = bu_{12} = \frac{-C_{TG}}{V}$$

$$\begin{aligned} \frac{\partial f_2}{\partial T} = \text{bu}_{21} = & C_{\text{TG}} C_{\text{MeOH}} \left(A_{1\text{f}} \exp\left(\frac{-E_{1\text{f}}}{RT}\right) \right) \left(\frac{E_{1\text{f}}}{RT} + 1 \right) \\ & - C_{\text{DG}} C_{\text{FAME}} \left(A_{1\text{r}} \exp\left(\frac{-E_{1\text{r}}}{RT}\right) \right) \left(\frac{E_{1\text{r}}}{RT} + 1 \right) \\ & - C_{\text{DG}} C_{\text{MeOH}} \left(A_{2\text{f}} \exp\left(\frac{-E_{2\text{f}}}{RT}\right) \right) \left(\frac{E_{2\text{f}}}{RT} + 1 \right) \\ & + C_{\text{MG}} C_{\text{FAME}} \left(A_{2\text{r}} \exp\left(\frac{-E_{2\text{r}}}{RT}\right) \right) \left(\frac{E_{2\text{r}}}{RT} + 1 \right) \end{aligned}$$

$$\frac{\partial f_2}{\partial F_{\text{MeOH}}} = \text{bu}_{22} = \frac{-C_{\text{DG}}}{V}$$

$$\begin{aligned} \frac{\partial f_3}{\partial T} = \text{bu}_{31} = & C_{\text{DG}} C_{\text{MeOH}} \left(A_{2\text{f}} \exp\left(\frac{-E_{2\text{f}}}{RT}\right) \right) \left(\frac{E_{2\text{f}}}{RT} + 1 \right) \\ & - C_{\text{MG}} C_{\text{FAME}} \left(A_{2\text{r}} \exp\left(\frac{-E_{2\text{r}}}{RT}\right) \right) \left(\frac{E_{2\text{r}}}{RT} + 1 \right) \\ & - C_{\text{MG}} C_{\text{MeOH}} \left(A_{3\text{f}} \exp\left(\frac{-E_{3\text{f}}}{RT}\right) \right) \left(\frac{E_{3\text{f}}}{RT} + 1 \right) \\ & + C_{\text{GL}} C_{\text{FAME}} \left(A_{3\text{r}} \exp\left(\frac{-E_{3\text{r}}}{RT}\right) \right) \left(\frac{E_{3\text{r}}}{RT} + 1 \right) \end{aligned}$$

$$\frac{\partial f_3}{\partial F_{\text{MeOH}}} = \text{bu}_{32} = \frac{-C_{\text{MG}}}{V}$$

$$\begin{aligned} \frac{\partial f_4}{\partial T} = \text{bu}_{41} = & -C_{\text{TG}} C_{\text{MeOH}} \left(A_{1\text{f}} \exp\left(\frac{-E_{1\text{f}}}{RT}\right) \right) \left(\frac{E_{1\text{f}}}{RT} + 1 \right) \\ & + C_{\text{DG}} C_{\text{FAME}} \left(A_{1\text{r}} \exp\left(\frac{-E_{1\text{r}}}{RT}\right) \right) \left(\frac{E_{1\text{r}}}{RT} + 1 \right) \\ & - C_{\text{DG}} C_{\text{MeOH}} \left(A_{2\text{f}} \exp\left(\frac{-E_{2\text{f}}}{RT}\right) \right) \left(\frac{E_{2\text{f}}}{RT} + 1 \right) \\ & + C_{\text{MG}} C_{\text{FAME}} \left(A_{2\text{r}} \exp\left(\frac{-E_{2\text{r}}}{RT}\right) \right) \left(\frac{E_{2\text{r}}}{RT} + 1 \right) \\ & - C_{\text{MG}} C_{\text{MeOH}} \left(A_{3\text{f}} \exp\left(\frac{-E_{3\text{f}}}{RT}\right) \right) \left(\frac{E_{3\text{f}}}{RT} + 1 \right) \\ & + C_{\text{GL}} C_{\text{FAME}} \left(A_{3\text{r}} \exp\left(\frac{-E_{3\text{r}}}{RT}\right) \right) \left(\frac{E_{3\text{r}}}{RT} + 1 \right) \end{aligned}$$

$$\frac{\partial f_4}{\partial F_{\text{MeOH}}} = \text{bu}_{42} = \frac{C_{\text{MeOH,in}} - C_{\text{MeOH}}}{V}$$

$$\begin{aligned}
\frac{\partial f_5}{\partial T} = bu_{51} = & C_{TG} C_{MeOH} \left(A_1 f \exp\left(\frac{-E_1 f}{RT}\right) \right) \left(\frac{E_1 f}{RT} + 1 \right) \\
& - C_{DG} C_{FAME} \left(A_1 r \exp\left(\frac{-E_1 r}{RT}\right) \right) \left(\frac{E_1 r}{RT} + 1 \right) \\
& + C_{DG} C_{MeOH} \left(A_2 f \exp\left(\frac{-E_2 f}{RT}\right) \right) \left(\frac{E_2 f}{RT} + 1 \right) \\
& - C_{MG} C_{FAME} \left(A_2 r \exp\left(\frac{-E_2 r}{RT}\right) \right) \left(\frac{E_2 r}{RT} + 1 \right) \\
& + C_{MG} C_{MeOH} \left(A_3 f \exp\left(\frac{-E_3 f}{RT}\right) \right) \left(\frac{E_3 f}{RT} + 1 \right) \\
& - C_{GL} C_{FAME} \left(A_3 r \exp\left(\frac{-E_3 r}{RT}\right) \right) \left(\frac{E_3 r}{RT} + 1 \right)
\end{aligned}$$

$$\frac{\partial f_5}{\partial F_{MeOH}} = bu_{52} = \frac{-C_{FAME}}{V}$$

$$\begin{aligned}
\frac{\partial f_6}{\partial T} = bu_{61} = & C_{MG} C_{MeOH} \left(A_3 f \exp\left(\frac{-E_3 f}{RT}\right) \right) \left(\frac{E_3 f}{RT} + 1 \right) \\
& - C_{GL} C_{FAME} \left(A_3 r \exp\left(\frac{-E_3 r}{RT}\right) \right) \left(\frac{E_3 r}{RT} + 1 \right)
\end{aligned}$$

$$\frac{\partial f_6}{\partial F_{MeOH}} = bu_{62} = \frac{-C_{GL}}{V}$$

State-space matrix

$$A = \begin{bmatrix} a_{11} & a_{12} & a_{13} & a_{14} & a_{15} & a_{16} \\ a_{21} & a_{22} & a_{23} & a_{24} & a_{25} & a_{26} \\ a_{31} & a_{32} & a_{33} & a_{34} & a_{35} & a_{36} \\ a_{41} & a_{42} & a_{34} & a_{44} & a_{45} & a_{46} \\ a_{51} & a_{52} & a_{35} & a_{54} & a_{55} & a_{56} \\ a_{61} & a_{62} & a_{36} & a_{64} & a_{65} & a_{66} \end{bmatrix}$$

$$B_U = \begin{bmatrix} b_{11} & b_{12} \\ b_{21} & b_{22} \\ b_{31} & b_{32} \\ b_{41} & b_{42} \\ b_{51} & b_{52} \\ b_{61} & b_{62} \end{bmatrix}$$

APPENDIX C
NOMENCLATURE

NOMENCLATURE

Abbreviations

$C_{\text{MeOH,in}}$	Initial concentration of methanol
CSTR	Continuous stirred tank reactor
$C_{\text{TG,in}}$	Initial concentration of triglyceride
CV	Controlled variable
DG	Diglyceride
FAME	Fatty acid methyl ester
F_{MeOH}	Methanol inlet flowrate
F_{oil}	Oil inlet flowrate
FR	Molar ratio
GL	Glycerol
IAE	Integral absolute error
MeOH	Methanol
MG	Monoglyceride
MV	Manipulated variable
P	Reactor pressure
RGA	Relative gain array
SVA	Singular value analysis
SVD	Singular value decomposition
TG	Triglyceride
T_{R}	Reactor temperature
V	Reactor volumn

APPENDIX C
International Proceeding

International Proceeding

Nanthinee Rakseethong, Winyusak Sritongkul, Withun Sonkhamwong, Veerayut Lersbamrungsuk, “Structural controllability evaluation of transesterification reactor and methanol recovery column in alkali-biodiesel production plants” TIChE International Conference 2011, Songkhla, Thailand, 10-11 Nov, 2011 (Oral presentation)

Nanthinee Rakseethong, Pinlada Tagoma, Alongkorn Makmeesup, Veerayut Lersbamrungsuk, “Design and control of alkali-catalyzed transesterification reactor” TIChE International Conference 2012, Nakornratchasima, Thailand, 25-26 Oct, 2012 (Oral presentation)

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