NONLINEAR AUTOREGRESSIVE WITH EXOGENOUS INPUT MODEL BASED CONTROL STRATEGY FOR BATCH ESTERIFICATION REACTOR

by

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LIST OF ABBREVIATIONS

Ac	Lauric acid
A/D	Analog to digital
Al	Citronellol
ANN	Artificial neural networks
ARMAX	Autoregressive moving average with exogenous inputs
ARX	Autoregressive model with exogenous inputs
BF	Best fit
D/A	Digital to analog
DIC	Direct inverse model control
DMC	Dynamic matrix control
E	Enzyme
Es	Citronellyl laurate
FOPTD	First order plus time delay
GMC	Generic model control
GMC-NN	Generic model control based neural network model
IAE	Integral absolute error
IMC	Internal model control
IMC-PID	Internal model control based PID
KH_2PO_4	Disodium hydrogen phosphate
L	Linear model
M-M	Michaelis Menten
MOPI	Malaysian organization of pharmaceutical industries
MPC	Model predictive control
MR	Momentum ratio

MSE	Mean square error
N	Nonlinear model
$Na_2HPO_4.2H_2O$	Sodium dihydrogen phosphate
NAMW	Number-average-molecular-weight
NARMA	Nonlinear autoregressive moving average
NARMAX	Nonlinear autoregressive moving average with exogenous inputs
NARX	Nonlinear autoregressive model with exogenous inputs
NARX-MBC	Nonlinear autoregressive model with exogenous inputs- model based control
NLJ	New Luus-Jaakola
NN	Neural network
NN-IMBC	Neural network inverse model based control
NN-IMC	Neural network based internal model control
NO	Nitric oxide
ODE	Ordinary differential equation
PI	Proportional-integral
PID	Proportional–integral–derivative
PLS	Partial least squares
R^2	Correlation coefficient
RBF	Radial Basis Function
RGA	Relative gain array
SOPTD	Second order plus time delay
SVM-IMC	Support vector machine approximated based IMC
W	Water

LIST OF SYMBOLS

		Unit
a	Dilation of the mother wavelet	-
A	Heat exchange area	m^2
A_{Ac}	Pre-exponential factor for lauric acid	L mol/min
A_{Al}	Pre-exponential factor for Citronellol	L mol/min
A_i	Pre-exponential factor for inhibition	L mol/min
a_{sk}	Scaling coefficients	-
a_{wk}	Wavelet coefficients	-
b	Translation of the mother wavelet	-
b_{n}	Dilation matrix in wavelet function	-
b_{sk}	Scaling dilation coefficients	-
b_{wk}	Wavelet dilation coefficients	-
C_{Ac}	Concentration of lauric acid	mol L^{-1} g^{-1} of enzyme
C_{Al}	Concentration of Citronellol	mol L^{-1} g^{-1} of enzyme
C_{Es}	Concentration of Citronellyl laurate	mol L^{-1} g^{-1} of enzyme
c_n	Translation vector in wavelet function	-
Cp_{Ac}	Specific heat of lauric acid	J/ mol K
Cp_{Al}	Specific heat of Citronellol	J/ mol K
Cp_{Es}	Specific heat of Citronellyl laurate	J/ mol K
Cp_j	Specific heat of water in jacket	J/ mol K
Cp_w	Specific heat of water	J/ mol K
c_{sk}	Scaling translation coefficients	-
C_w	Concentration of water	mol/L
c_{wk}	Wavelet translation coefficients	-
D(s)	Denominator of transfer function	-

d	Disturbance	-
e	Error	-
E_{Ac}	Activation energy for lauric acid	J/ mol K
E_{Al}	Activation energy for Citronellol	J/ mol K
E_i	Activation energy for inhibition	J/ mol K
$F(.)$	Function	-
f_i	Model data	-
F_j	Jacket flowrate	L/min
$G(s)$	First principle model	-
$G_c(s)$	Controller model	-
$\check{G}_p(s)$	Internal model	-
h_k	Basis function	-
K	Number of wavelet	-
K_{Ac}	Michaelis constant for Citronellol	mol L^{-1} min^{-1} g^{-1} of enzyme
K_{Al}	Michaelis constant for lauric acid	mol L^{-1} min^{-1} g^{-1} of enzyme
K_i	Inhibition constant for lauric acid	mol L^{-1} min^{-1} g^{-1} of enzyme
K_p	Process gain	-
N(s)	Nominator of transfer function	-
n_t	Length of actual data period	-
$\dot{Q}$	Heat flow	kW
R	Gas constant	J/ mol K
r	Positive integer	-
r_{Ac}	Initial rates of reaction for lauric acid	mol min^{-1} g^{-1} of enzyme
r_{max}	Maximum rates of reaction	mol min^{-1} g^{-1} of enzyme

t	Time	min
T_j	Jacket temperature	K
T_{ji}	Jacket temperature inlet	K
T_r	Reactor temperature	K
U	Heat exchange coefficient	W/m^2/K
u	Input	-
V_j	Jacket volume	L
w_k	Weight coefficient	-
x	Memory of the model	-
y	Real process output	-
$\bar{y}$	Means of the experimental data	-
$\tilde{y}$	Model output	-
y_i	Experimental data	-
y_{sp}	Output set point	-

Greek letters

$\delta(t)$	Degree of interaction	-
$\lambda(t)$	Degree of nonlinearity	-
∂u_i	Changes in input	-
∂u_i	Changes in output	-
$\mu(\theta)$	NARX model	-
α	Parameter for kinetic model	-
β	Parameter for kinetic model	-
τ	Time constant	min
τ_c	Low-pass filter time constant	min
ρ_j	Density of water in jacket	g/L
ΔH_{rxn}	Heat of reaction	kJ/mol

STRATEGI KAWALAN BERDASARKAN MODEL AUTO TAK LELURUS DENGAN MASUKAN LUAR UNTUK REAKTOR PENGESTERAN KELOMPOK

ABSTRAK

Reaktor pengesteran kelompok kerap digunakan di dalam industri pemakanan dan farmasi. Kebiasaannya, pengesteran kelompok adalah proses yang kompleks kerana ia melibatkan kelakuan dinamik yang tak lelurus. Ini adalah disebabkan faktor pencirian kinetik dan proses yang tidak stabil. Oleh kerana itu, pemodelan dan kawalan reaktor pengesteran menjadi sukar dan penggunaan strategi kawalan termaju boleh menghasilkan prestasi kawalan yang lebih baik. Di antara semua strategi kawalan termaju, teknik kawalan berdasarkan model lebih banyak mendapat perhatian sepanjang dekad yang lalu. Sehubungan dengan itu, dalam kajian ini, kawalan berdasarkan model auto mundur tak lelurus dengan masukan luar (NARX-MBC) telah dibina bagi mengawal suhu reaktor pengesteran kelompok. Pembangunan strategi kawalan NARX-MBC dibahagikan kepada tiga bahagian penting iaitu proses, model dalaman dan pengawal.

Proses pengesteran kelompok telah diwakili dengan model prinsip pertama. Ia melibatkan persamaan-persamaan kinetik, imbangan jisim dan tenaga. Keputusan yang didapati melalui kajian kinetik menunjukkan proses pengesteran Citronellyl laurate mematuhi mekanisma urutan bi-bi dengan R^2=0.982. Kemudian, ia telah diselesaikan dengan menggunakan kaedah Runge-Kutta urutan keempat yang boleh didapati di dalam MATLAB. Ia juga disahkan dengan data eksperimen dengan nilai MSE=$1.2x10^{-2}$. Ia juga digunakan dalam kajian kelakuan dinamik dan menunjukkan

bahawa penghasilan ester bertambah dengan bertambahnya suhu dan kuantiti masukan pemangkin.

Di dalam pembangunan model dalaman, proses pengesteran kelompok telah diklasifikasikan sebagai proses tak lelurus dan mempunyai interaksi yang nyata antara pembolehubah. Sehubungan dengan itu, semua pembolehubah masukan dan luaran dipersembahkan dengan menggunakan model tak lelurus iaitu model NARX. Berdasarkan kajian interaksi, suhu reaktor telah dipilih sebagai pembolehubah kawalan, kadar aliran air di dalam jaket sebagai pembolehubah olahan dan suhu air di dalam jaket sebagai gangguan. Struktur dan pengesahan model NARX telah dilakukan dengan menggunakan kotak perkakasan bagi sistem pengenalpastian dalam MATLAB. Model NARX yang mengandungi tiga masukan terdahulu, tiga luaran terdahulu dan 85 unit nombor untuk jaringan gelombang telah dipilih sebagai model terbaik berdasarkan nilai BF=91.58 %. Untuk melengkapkan pembinaan NARX-MBC, model pengawal telah dipersembahkan dengan balikan fungsi pindahan urutan pertama dan parameter untuk model tersebut telah dikenalpasti dengan menggunakan kaedah Cohen-Coon.

Prestasi untuk pengawal NARX-MBC telah dinilai di dalam mengesan titik set, perubahan titik set dan perubahan bebanan dengan nilai IAE masing-masing 1.928×10^{-2}, 1.747×10^{-2} and 1.568×10^{-2}, yang mana nilai-nilai ini lebih kecil daripada pengawal PID dengan nilai IAE masing-masing 5.173×10^{-2}, 6.405×10^{-2} and 8.745×10^{-2}. Di dalam ujian ketegapan, NARX-MBC dikenalpasti lebih tegap dengan purata nilai IAE=2.214×10^{-2} berbanding dengan pengawal PID dengan nilai IAE=7.901×10^{-2}. Kesimpulannya, NARX-MBC yang telah dibina sangat berkesan

dan lebih baik daripada pengawal PID dalam mengawal reaktor pengesteran kelompok.

NONLINEAR AUTOREGRESSIVE WITH EXOGENOUS INPUT MODEL BASED CONTROL STRATEGY FOR BATCH ESTERIFICATION REACTOR

ABSTRACT

Batch esterification reactors are frequently used in the food and pharmaceutical industries. In general, batch esterification is a complex process since the system involves a nonlinear dynamic behavior due to its kinetic characteristics and unsteady state process. Due to these reasons, the modeling and control of the batch esterification reactor is difficult and the application of advanced control strategies often lead to a better control performance. Among all the advanced control strategies, model-based control techniques have been largely extended and have gained prominence during the past decades. In this work, a Nonlinear AutoRegressive with eXogenous inputs-Model Based Control (NARX-MBC) strategy was developed to control the reactor temperature of the batch esterification reactor. The development of the NARX-MBC strategy is divided into three important sections i.e. process, internal model, and controller.

The batch esterification process was represented by the first principle model which involved kinetic, mass and energy balance equations. The results obtained from the kinetic studies indicated that the Citronellyl laurate esterification process follows an ordered bi-bi mechanism with R^2=0.982. Then, the first principle model equations are solved using the 4^{th} order Runge-Kutta method which is available in the MATLAB and has been validated with experimental data with Min Squared Error (MSE) =$1.2x10^{-2}$. The validated first principle model was used to study the dynamic behavior of the esterification process and it was found that the ester conversion increased with increasing reactor temperature and amount of catalyst loading.

In the internal model development, the relationship of all input and output variables were represented by a nonlinear model (i.e. NARX model) since the batch esterification process has been classified as a nonlinear process and has significant interactions between the process variables. Based on the interaction study, reactor temperature was selected as the controlled variable, jacket flowrate as the manipulated variable and jacket temperature as disturbance. The structure and validation for the NARX model were carried out using the system identification toolbox in the MATLAB. The NARX model that consisted of three past inputs, three past outputs and 85 number of wavenet units was selected as the best model based on its performance with Best Fit (BF) value = 91.58 %. As a completion of the NARX-MBC development, the controller model was represented by the inverse of the first order transfer function and the model parameters were identified using the Cohen-Coon method.

The performances of the NARX-MBC controller were evaluated based on its set point tracking, set point change and 1oad change with IAE values equal to $1.928x10^{-2}$, $1.747x10^{-2}$ and $1.568x10^{-2}$ respectively which is smaller than the PID controller with IAE values equal to $5.173x10^{-2}$, $6.405x10^{-2}$ and $8.745x10^{-2}$ respectively. In the robustness tests, the NARX-MBC was verified to be more robust with average IAE= $2.214x10^{-2}$ compared to the PID controller with average IAE=$7.901x10^{-2}$. As a conclusion, the NARX-MBC developed performed well and was more superior than the PID controller in controlling the batch esterification process.

CHAPTER 1

INTRODUCTION

1.1 Research Background

Esterification is one of the important processes in chemical industries especially for the food and pharmaceutical industry. In industry, most of the ester productions are produced in a batch reactor because such a reactor is quite flexible and can be adapted to small volumes of production (Barbosa-Póvoa, 2007). In this work, this batch reactor is referred to as a batch esterification reactor. Ester products have wide applications in areas such as pesticides and herbicides, metal extraction agents, synthetic lubricants, polymerization aids for acrylic acid esters, insect attractants and repellants, industrial fragrances, odorants and cosmetic components, pharmaceutical applications, and photographic applications (Serri *et al.*, 2006).

Controlling the batch esterification reactor is very important in order to ensure the successful operation of many food and pharmaceutical industries. A proper control strategy can lead to maximum production rates which can be achieved at minimum energy consumption (Liu and Macchietto, 1995). The difficulties in controlling a batch esterification reactor are similar with other types of batch reactor systems such as the polymerization process (Shafiee *et al.*, 2008), the crystallization process (Paengjuntuek *et al.*, 2008) and the fermentation process (Nagy, 2007) where nonlinearity becomes a main issue. This nonlinearity exists due to the nature of the process behavior such as reaction temperature, reaction time and pressure and in most cases, is caused by the complexity of the process kinetics (Le Lann *et al.*, 1999). However, a majority of industries still use Proportional Integral Derivative

(PID) controller to control batch reactor process (Chen and Huang, 2004) because it is a relatively simple structure that can be easily understood and implemented (Kristiansson and Lennartson, 2006).

The application of a conventional PID controller for the batch reactor, however, does have some fundamental limitations. The conventional PID controller cannot cover a wide range of operating conditions and can poorly cope with highly nonlinear process dynamics. To overcome this problem, many advanced control techniques have been proposed to control batch reactors such as cascade feedforward-feedback control, iterative learning control and model based control (Nagy *et al.*, 2007; Konakom *et al.*, 2008). Among these advanced techniques, the model based control strategy has received a great deal of interest during the past decade because of its ability to give better control performance when compared to other advanced control strategies (M'sahli *et al*, 2002). Some of the advantages of the model based control when compared to other advanced control techniques can be listed as follows (Li *et al.*, 1989):

- It can be used for step and impulse response data which can easily be obtained.
- It gives satisfactory performance even with time delays and high nonlinearities.
- It can be used in multivariable format.
- It is robust in most cases.
- The implementation of such a technique is simple.
- It can be used to control various processes, whether simple or complex ones.

Among the model based control strategies available are the Internal Model Control (Garcia and Morari, 1982), the Generic Model Control (Cott and Macchietto,

1989) and the Model Predictive Control (Garcia *et al.*, 1989). Among the model based control strategies, the Internal Model Control (IMC) strategy is among the most promising since it is relatively easy to implement and can handle multivariable systems (Patwardhan and Madhavan, 1998). The Internal Model Control (IMC) is a type of controller that used model of the process in order to derive both controller structure and parameters (Ogunnaike & Ray, 1994). Many researchers have implemented a linear model to be embedded in the IMC structures (Garcia and Morari, 1982; Toivonen *et al.*, 2003; Alabi and Taiwo, 2005; Selvi *et al.*, 2007; Vilanova, 2008). However, such an implementation can cause inefficient performance if the process is highly nonlinear. Economou *et al.* (1986) developed a nonlinear model based on the IMC and they achieved better control performance. This shows that a suitable nonlinear model is an obvious requirement for a successful IMC application (Vieira *et al.*, 2003).

1.2 Problem Statement

In the early production of esters, they were traditionally produced by extraction from natural sources and were too expensive for commercial use. Therefore, to overcome this problem many researchers (Wang and Linko, 1995; Shintre *et al.*, 2002; Zaidi *et al.*, 2002; Altiokka and Citak, 2003; Romero *et al.*, 2005; Serri *et al.*, 2006) focused on the improvement in the reaction of the esterification process such as the use of different types of catalysts and solvents. However, maximum production of the esterification process can be achieved if the reaction can be kept at optimum conditions rather than changing the catalyst or solvent, which is expensive (Yadav and Lathi, 2003).

Therefore, the control of the batch esterification reactor is vital to ensure high yields of ester production. Batch esterification reactors are traditionally controlled using linear controllers such as conventional PID controllers. However, batch process is characterized by a limited time duration and non-stationary operating conditions thus, nonlinearities may have an important impact on the control problem (Hua *et al.,* 2004).

As a result, PID controllers can lead to poor control performance. Thus, an advanced control strategy is needed to improve the control performance for the batch reactor. To the best of the author's knowledge, there has been no development of an efficient model based control strategy for the batch esterification reactor. A model based control strategy such as the IMC which has received a great deal of interest during the past decade, thus has a high potential to be implemented in a batch esterification reactor (Patwardhan and Madhavan, 1998).

The quality of the IMC controller can be judged based on regulatory, servo behavior and the ability to deal with the robustness of the process system. One of the important issues in IMC is the development of reliable models to represent the real process and the controller. Linear models which have received attention from many researchers in the past were found to have weaknesses especially when dealing with the nonlinear process. Due to this, the development of a reliable nonlinear model to ensure the best performance of batch esterification is a must.

1.3 Research Objectives

The objectives of this study are:

i. To develop and validate a first principle model of batch the Citronellyl laurate esterification process based on an ordered bi-bi kinetics mechanism.

ii. To study the nonlinear behavior of the Citronellyl laurate esterification process.

iii. To develop and validate a Nonlinear Auto Regressive with eXogenous input (NARX) model for the Citronellyl laurate esterification process.

iv. To develop and to evaluate the performance and robustness of a NARX-model-based-control (NARX-MBC) strategy for the control batch esterification process.

v. To compare the performance of the NARX-MBC with a PID controller.

1.4 Scope of Study

In this work, the batch esterification of Citronellyl Laurate via immobilized *Candida rugosa* lipase is chosen to represent the batch esterification reaction (Serri *et al.*, 2006). Citronellyl Laurate is synthesized from Citronellol and Lauric acid using immobilized *Candida rugosa* lipase. Esterification is carried out using lipase immobilized in batch reactor. Figure 1.1 shows a schematic diagram that represents the esterification of Citronellyl laurate. In the batch esterification system, there are various parameters that affect the ester rate of reaction such as different catalysts, solvents, speed of agitation, catalyst loading, temperature, mole ratio, molecular

sieve, pH and water content (Yadav and Lathi, 2005). In this study, only reactor temperature is chosen as a controlled variable as it is easy to measure as compared to the other parameters.

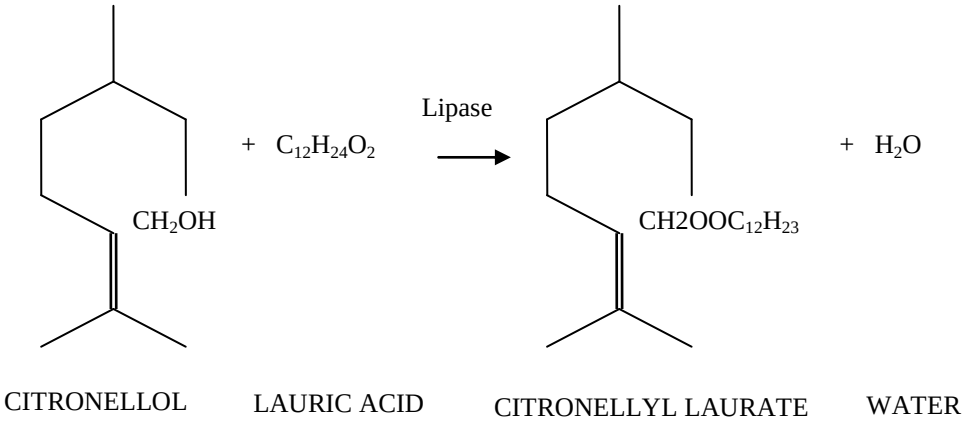

Figure 1.1: Schematic representation of the esterification of Citronellyl laurate

A mathematical model for the Citronellyl laurate batch esterification reactor is developed based on a first principle model which consists of mass balance, energy balance and kinetics equations. Experiments are carried out in order to estimate the kinetic parameters for the esterification of Citronellyl laurate and the results are validated using experimental data. The kinetic scheme for this process is considered as an ordered bi-bi mechanism. Then, the validated kinetic model is implemented in the first principle model and simulation studies are carried out in the MATLAB environment. The validated first principle is then used to study the effects of the operating parameters and to study the nonlinear behavior of the process.

The nonlinear empirical model development and its application in the IMC design is the crucial part in this work. Thus, a Nonlinear Auto Regressive with eXogenous input (NARX) model is developed. The data for the NARX model

identification and validation is generated from the validated first principle model. The input and output data for the NARX model is based on the controlled, manipulated and disturbance variables for the batch esterification process. Then, the identified NARX model is embedded as an internal model in the IMC controller.

The IMC consists of three main components: real process, internal model and controller. In this work, the real process and internal model are represented by the first principle model and the NARX model respectively. Meanwhile, the controller is developed using the inverse transfer function of the process model. This control scheme is known as NARX-MBC. The performances of the NARX-MBC are evaluated through set point tracking, set point change and disturbance rejection. The robustness of the NARX-MBC is studied by using four tests i.e. increase of heat transfer coefficient, increase of heat of reaction, decrease of inhibition activation energy and simultaneous change of all the mentioned parameters. Finally, the performance of NARX-MBC is compared to a conventional PID controller.

1.5 Organization of the Thesis

In this thesis, there are a total of five chapters and each chapter gives specific information about the research project.

Chapter 1 gives a general background about the batch esterification process and an overview of process control applications especially in model based control in the batch esterification process. The problem statement, the objectives and the scope of study of this research project are also stated clearly in this chapter.

Chapter 2 provides the literature review related to the modeling and control for the batch esterification reactor. It also elaborates the information concerning the batch esterification reactor, the advanced control strategy and the IMC. The background information about the first principle and the empirical model related to this project are also presented. Finally, current applications of the IMC applications are discussed together with the design characteristics and their advantages.

Chapter 3 describes the procedure to develop a process model and an effective control system for batch esterification. The first part of this chapter presents information on the experimental batch reactor setup, the experimental measurements and the procedures for dynamic behavior study and kinetic study. The second part describes the methodology to develop the first principle model and the input-output model for the Citronellyl laurate esterification process. In the last part of this chapter, the designs and the control performances of both controller (NARX-MBC and PID) procedures are described.

Chapter 4 presents the results and discussion of this research. The first part of this chapter contains the results and discussion for the kinetics study, the validation of the first principle model, the effect of catalyst loading and reactor temperature to the ester conversion profile and the nonlinear behavior of the process. The results for the NARX model development and validation are discussed in next section followed by the results of the implementation of the NARX model in the IMC design. The final part of this chapter presents the comparison of the NARX-MBC and PID controller performances in set-point tracking, set-point changes, load changes and robustness tests.

Chapter 5 gives the conclusions of the results obtained in the present study. In addition, some recommendations for future studies are also highlighted in this chapter.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

Over the last decade, the Malaysian food and pharmaceutical market has been growing between 8 % to 10 % annually. A significant development in the industry in the last decade cause the strong emphasis placed by local manufacturers on research and development, particularly in areas of product innovation and process improvement (MOPI, 2009). One of the important processes in the food and pharmaceutical market is the esterification process which is mainly used for flavors and fragrance components in products (Güvenç *et al.*, 2002). In research development, many researchers focused on the improvement of the process by modifying the enzymes (e.g. mutation and cloning) and used different types of organic solvents. However, most of the enzymes have limited and selective tolerance to particular organic solvents (Salleh *et al.*, 2002). One of the effective ways to improve the process is by controlling the operating conditions of the process at their optimum values. Optimum operating conditions can influence the reaction rate working at maximum conditions with minimum energy requirement. Therefore, process control is one of the alternative ways in process improvement (Lipták, 2003).

This chapter reviews background information on the various approaches and methods that are currently used to model and control the batch esterification reactor. The overview on the application of the batch esterification reactor model and control in this study is mostly based on the typical batch reactor systems since they have the same modes of operation with nonlinear and unsteady state process.

2.2 Esterification

Ester is one of the most important elements whose compounds are formed from the reaction between alcohols and acids. Esters are widely used for flavorings; many are 'nature-identical', that is, synthetic versions of the esters present in the fruit. Fruit flavors are very complex though, often arising from many different compounds, some of which are present in small quantities. Their lack of hydrogen-bond-donating ability means that ester molecules cannot hydrogen-bond to each other which, in general, makes esters more volatile than a carboxylic acid of a similar molecular weight. This property makes them very useful in organic analytical chemistry (Garcia *et al.*, 1996, 1999 & 2000; Yadav and Lathi, 2003).

Citronellyl laurate ester is a type of bioproduct that is usually produced using batch reactor process. Citronellyl laurate are fragrances and flavors that are widely used in the food, cosmetics and pharmaceutical industries (Serri *et al.*, 2006). The mechanism of the esterification reaction mainly relies on the rates of chemical reactions and kinetics behaviors which depend on operating conditions such as reactant concentration and temperature. Thus, understanding chemical kinetics is important in providing essential knowledge of the esterification process mechanism since it can be confirmed only after a detailed kinetics investigation has been performed.

The characteristics of the esterification process are usually complex and the kinetics information are limited (Bernard and Gouze, 2004). In the past, Yadav and Lathi (2004) and Serri *et al.* (2006) reported that the kinetics model of the Citronellyl laurate esterification process is represented by an ordered bi-bi mechanism with the

inhibition of lauric acid. However, instead of using *Candida Antartica* (Yadav and Lathi, 2003), Serri *et al.*, (2006) used *Candida rugosa* (self-immoblized by the absorption method onto Amberlite MB-1 resin) as a catalyst to the process reaction due to its low price and their economic reused.

2.3 Batch Esterification Reactor

Traditionally, many flavor compounds in the market are still produced via extraction from plant and animal sources but the high cost and low quantities of the product achieved make these techniques inadequate for industrial applications (Longo and Sanroman, 2006). As a result, the industrial production of these kinds of compounds is carried out by chemical synthesis where inorganic solvents and enzymes are used in the reaction synthesis. However, the chemical synthesis is not environmental friendly and leads to the development of biotechnological enzymatic synthesis in which organic lipase and solvents are used. The biotechnological enzymatic synthesis has clear environmental advantages since inorganic acids and solvents are avoided and the enzymes can be reused thus, minimizing the reaction residues (Romero *et al.*, 2007).

Many of the esterification processes that use lipase enzymatic synthesis are usually manufactured in batch reactor system. Moreover, the continuous production of these products is not feasible or economically attractive since it is difficult to handle the immobilized lipase that is involved as the catalyst in the reaction (Bouhenchir *et al.*, 2006). The mode of the operation of batch esterification reactor is similar with other batch reactor processes where there is no inflow or outflow of reactants or products while the reaction is being carried out. The reaction will

continue for a certain period and finally at the end of the process the resultant mixture is discharged.

Figure 2.1 shows a schematic diagram of a batch reactor. A typical batch reactor consists of a tank with an agitation and recirculating heating-cooling jacket fluid to maintain temperature within a desirable range. The use of batch reactors offers many advantages. Firstly, a batch reactor is quite flexible; it can be stopped between batches and it is easy to change the different compositions in the different batches to produce products with different specifications. Secondly, they can achive high reactant conversion and also limit the amount of waste produced during a failed reaction. Finally, only heat transfer rate and agitation are the most common causes of scale-up difficulties compared with continuous process which the volume of reactor contents changes with time (Arpornwichanop *et al.*, 2005). Due to this reason, simulating the continuous process with computer model is difficult.

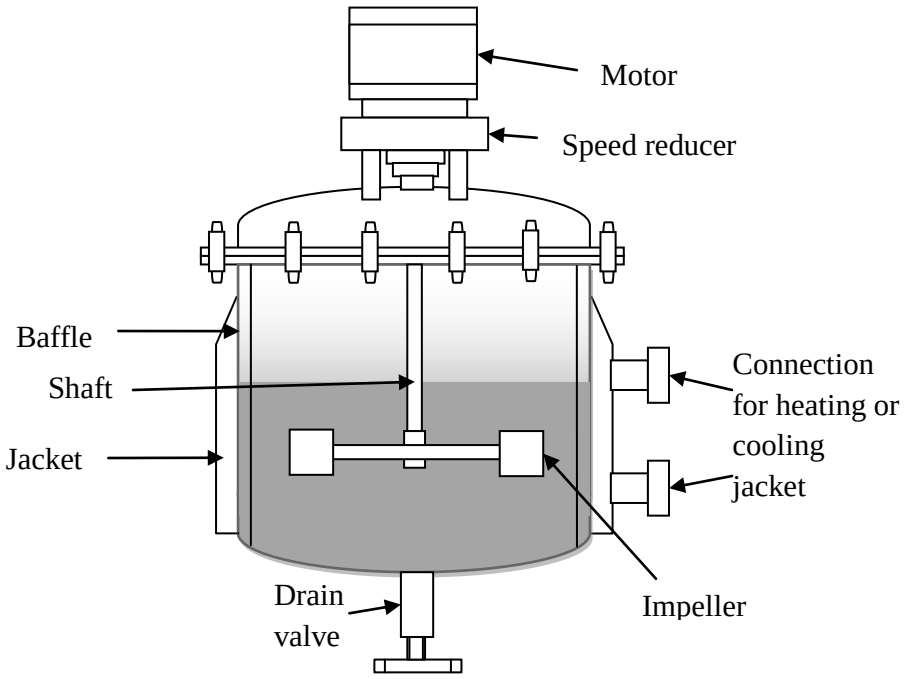

Figure 2.1: A schematic diagram of a batch reactor (Coker, 2001)

In some cases, the concept of a batch reactor assumes that the reaction is instantaneously charged and perfectly homogenized in the reactor (Coker, 2001). However, this concept is not valid with complex processes where the reaction systems that are normally not entirely known; have strong non-linear dynamics and parameters varying with time (Galvan and Zaldivar, 1997). In the batch esterification system, there are various parameters that affect the ester rate of reaction such as different catalysts, solvents, speed of agitation, catalyst loading, temperature, mole ratio, molecular sieve and addition of water (Yadav and Lathi, 2005). In the batch esterification reactor, the parameters that can be controlled in order to operate the system at the optimum condition are speed of agitation, temperature, catalyst loading and mole ratio.

2.4 Batch Reactor Control

In industrial operations, control contributes to safety and environmental impact minimization by maintaining process variables near their desired values. The control system will move the process manipulated variables at its desired conditions in order to satisfy performance criteria (Cott and Macchietto, 1989) such as:

- *Economic*: These can be associated with maintaining process variables at the targets which can minimize operating cost function.
- *Safety and Environmental*: Some process variables must not violate specified bounds such as personnel or equipment safety or environmental regulations.
- *Equipment*: The control system must not drive the process outside the physical limitations of the equipment.
- *Product quality*: Consumer specifications on the product must be satisfied.

When the process parameters change, the controller frequently needs to be retuned in order to maintain satisfactory performance. Retuning the controller is usually time consuming and requires a combination of operational experience and trial-and-error processes (Nikravesh *et al.*, 2000). One of the difficulties in analyzing the dynamic response on batch processes is because of its nonlinearity behavior. To a certain extent, most of the systems in many applications are nonlinear and this makes the nonlinearity study a must rather than the exception (Chow *et al.*, 2001).

Pearson (2003) classified the processes by using a degree of nonlinearity, i.e. mild, intermediate, or strongly nonlinear and the qualitative nature of the process nonlinear behavior. Mild nonlinear processes included those that exhibit asymmetric response to symmetric changes in input and those show input multiplicities i.e., the same output could be generated by different input magnitudes. Intermediate nonlinear processes show an output multiplicative behavior whereas strongly nonlinear processes show chaotic dynamics. This classification is very important since it can be used for the rational selection of controller design strategies (Hernjak *et al.*, 2004).

The nonlinearity of the batch reactor process can be due to the behavior of external processes (e.g. stirring behavior; nonlinear heating-cooling jacket recirculating flow rate) and also from the behavior of internal processes (e.g. kinetics characteristics, excess heat from chemical reaction and unsteady state process). Due to these difficulties, linear controllers or control strategies based on a local linearized model e.g. standard PID controller lead to poor control performance (Liu &

Macchietto, 1995). Hence, advanced control methodologies need to be designed to improve control performance.

2.5 Advanced Control Strategy Applied in Batch Reactor

There are many types of advanced control techniques using either specific algorithms for particular systems or very general methods with a wide application area and a well-developed theory. A classification of these techniques is difficult because many of the algorithms are very similar and only differ slightly in terms of performance criteria, prediction horizon, and constraint handling. There are two main advanced control techniques that have been applied to control batch reactors: knowledge based control and model based control strategy. Knowledge based control strategy is applied by knowledge gained during repeated operations under the control of highly skilled operators and model based control is applied by using the mathematical model of the process (Shioya *et al.*, 1999).

The process of knowledge base requires a sophisticated operational logic which cannot easily fit into the mathematical framework of the traditional control approach (Horiuchi, 2002). Moreover, the knowledge based approach often leads to time delays, which are larger than the system time-constant (Meinberg and Jakobza, 2005). These techniques also require modern nonlinear control methods which are computationally hard and thus are generally only feasible for small-scale systems. As a result, a mathematical model is preferred in order to predict the output of the process and is known as the model based control strategy.

Model-based control techniques have been largely extended and have gained prominence during the past decades. The classification of these techniques is shown in Figure 2.2. The increase of mathematical models in the chemical engineering field was a major driving force behind the development of the model-based control strategy. Advanced computational methods and available supercomputers can now readily identify the important phenomena in a complex chemical process over the entire range of applicable conditions by the exhaustive solution of the detailed model. A better model can replace laboratory or field tests that are difficult or costly to perform or to identify crucial experiments that should be carried out. Agachi *et al.*, (2006) estimated that up to 80% of time and expense in the design and installation of the model based control is attributed to modeling and system identification.

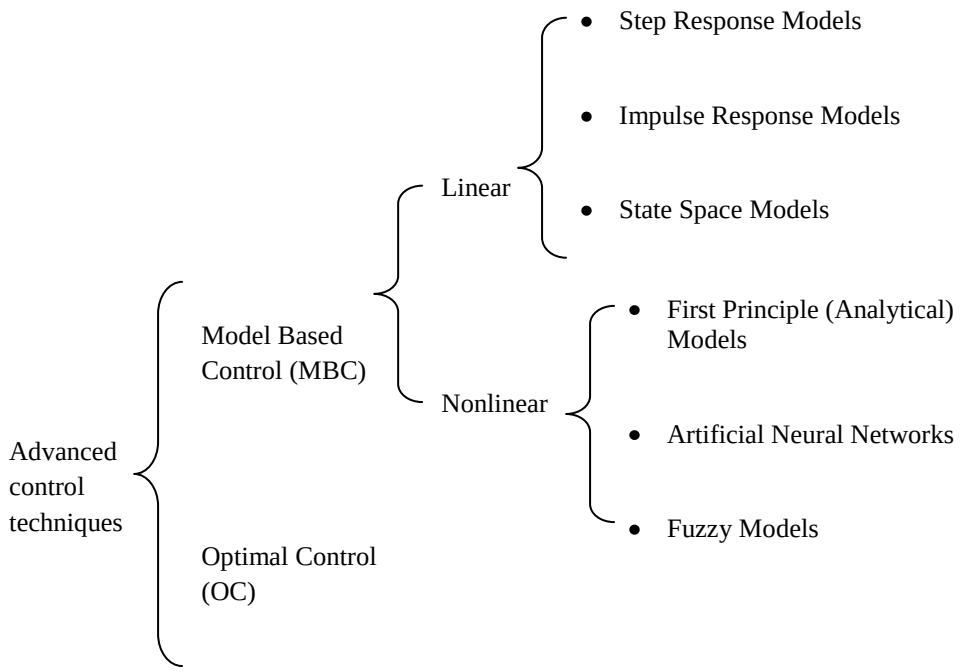

Figure 2.2 Classification of model based control techniques (Agachi *et al.*, 2006)

There are many types of model based control strategies such as Model Predictive Control (MPC), Generic Model Control (GMC), Dynamic Matrix Control (DMC) and Internal Model Control (IMC). However, the IMC strategy is found to be relatively easy to implement when compared to other model-based control methods. The IMC controller has a simple structure and fewer parameters to be tuned. Moreover, it has significant effectiveness in improving the robustness and control performance of the system with a long time delay (Li *et al.*, 2009). Because of these advantages, the IMC has received a lot of attention from many researchers in the field of process control system.

2.6 Internal Model Control (IMC)

IMC is a control strategy which uses a process mathematical model to design the controller. The IMC structure originated based on the idea proposed by Smith (1959) with the Smith Predictor Control (SPM) which consisted of a plant model as a reference model. Garcia and Morari (1982) formulated and unified the SPM concept within the context of process control applications and came out with the Internal Model Control (IMC) scheme. The basic IMC structure proposed by Garcia and Morari (1982) was based on the linear control strategy. Thereafter, the design procedure for the IMC controller was extended to a nonlinear controller by Economou *et al.* (1986). From that point, the IMC has become one of the most tempting advanced control strategies. More work was carried out to improve the IMC strategy such as the multiloop-IMC structure (Economou and Morari, 1986), the IMC based PID control (Rivera *et al.*, 1986), the adaptive IMC (Narayanan *et al.*, 1997), neural network based IMC (Aziz *et al.*, 2000; Mujtaba *et al.*, 2006; Ekpo and

Mujtaba, 2008), and Support Vector Machine approximate based IMC (SVM-IMC) (Wang and Yuan, 2008).

Figure 2.3 shows the basic block diagram of the IMC structure as proposed by Garcia and Morari (1982). For simulation environments, the real process (*G(s)*) can be represented using the *first principle model*. The main components in the IMC structure consists of the internal model ($\check{G}_p(s)$) to predict the effect of the manipulated variables on the output which can be represented by the *empirical model.* The IMC structure also consists of a control algorithm ($G_c(s)$) to compute future values of the manipulated variable which can be developed by the *inverse model*. Theoretically, the drawbacks of the IMC for the open loop control system are based on its sensitivity to modeling errors ($y - \tilde{y}$) and the inability to deal with external disturbances (*d(s)*) entering the system.

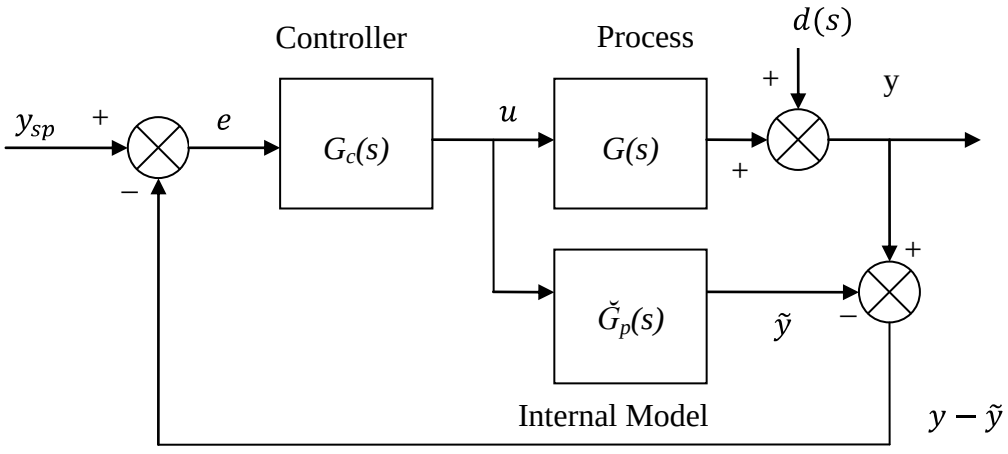

Figure 2.3: Block diagram of IMC structure

The theory of the IMC is based on the encapsulation of the process control system by either implicit or explicit process representation (Garcia and Morari, 1985). This theoretical control performance, whereby the output remains equal to the

input (or set-point) without the use of feedback, illustrates that the perfect control can be achieved if complete knowledge of the process is well known and feedback is only necessary when knowledge about the process is inaccurate or incomplete. These conditions in practice are unachievable in an open-loop system due to the following reasons:

- A mismatch between the actual process and the process model.
- The process model may not be invertible.
- Unknown disturbances within the system.

2.6.1 First Principle Model

As can be seen in Table 2.1, many researchers developed the first principle model of the batch reactor process for general second order reaction where the process is simple and easy to implement. Some researchers considered other types of reaction such as polymerization, pharmaceutical, exothermic and neutralization reaction. Table 2.1 also reveals that the first principle model of the batch reactor can be categorized as reduced and full models. However, the implementation of the full first principle model is more attractive since this model can represent the real process better. This is because the full model takes into considerations the kinetics, mass and energy balance of the system. In contrast, the reduced model that is developed depends on the objective of the process control.

Table 2.1: First principle models applied in batch reactor

No	Authors	Years	Model	Batch Process System
1	Garcia *et al.*	1995	Reduction model	General second order reaction
2	Nagy and Agachi	1997	Full model	Polymerization
3	Galvan and Zaldivar	1997	Full model	N/A

4	Louleh *et al.*	1999	Reduced model	N/A
5	Aziz *et al.*	2000	Full model	General second order reaction
6	Xaumier *et al.*	2000	Full model	N/A
7	Aziz and Mujtaba	2002	Full model	General second order reaction
8	Sampath *et al.*	2002	Reduced model	Polymerization
9	Altiokka and Citak	2003	Reduced model	Esterification
10	Bequette *et al.*	2004	Full model	Pharmaceutical
11	Hua *et al.*	2004	Reduced model	General second order reaction
12	Arpornwichanop *et al.*	2005	Full model	General second order reaction
13	Asteasuain *et al.*	2006	Full model	Polymerization
14	Bouhenchir *et al.*	2006	Full model	Neutralization chemical reaction
15	Baghli and Benyettou	2006	Reduced model	N/A
16	Cho *et al.*	2008	Full model	General second order reaction
17	Paengjuntuek *et al.*	2008	Full model	Crystallization
18	Konakom *et al.*	2008	Reduced model	Exothermic reaction

The full first principle model of batch esterification reactor consists of kinetics, mass and energy balance equations in order to describe the behavior of the associated physical, chemical and other related disciplinary behavior of the batch esterification process. However, many of the previous researchers only came out with the derivation of the kinetics model and the rate of reaction equation (Garcia *et al.*, 1996, 1999 & 2000; Janssen *et al.*, 1999; Miguel *et al.*, 2001; Yadav and Lathi, 2003; Serri *et al.*, 2006; Foresti *et al.*, 2008; Yadav and Borkar, 2008). This kinetics model and the rate of reaction equation are unable to describe the relationship behavior between various operating conditions towards ester production in the batch reactor.

The kinetics model of reaction catalyzed by the enzyme process is usually derived based on the applications of the Michaelis-Menten rate equations. However, based on Foresti *et al.* (2008), this type of model is only valid for reactions where only one substrate is converted into one product and not valid for reactions involving two or more substrates. In this case, the general mechanism for esterification reactions catalyzed by immobilized lipase involves the formation of an acyl-enzyme intermediate, followed by the incorporation of an alcohol molecule to produce a molecule of ester and water (Garcia *et al.*, 1996). As a consequence, the kinetics model for this kind of reactions can be derived by the ordered bi-bi mechanism which rules out the random mechanism (Yadav and Trivedi, 2003).

Some researchers (Janssen *et al.*, 1999; Paiva *et al.*, 2000; Foresti *et al.*, 2008) have suggested using the Ping Pong bi-bi mechanism for this kind of reaction to derive the kinetics model equations. However, based on Yadav and Lathi (2004), the kinetics models of Citronellyl laurate esterification can be derived from ordered bi-bi mechanism with the dead end complex of lauric acid. These assumptions were reported by Serri *et al*. (2006) in which the kinetics constants obtained from ordered bi-bi mechanism is more reliable compared with the kinetics constant obtained from the Ping Pong bi-bi mechanism.

Mass balances can be specified either in the total mass of the system or in the component balances. The mass balance on batch processes can be described based on the differential balances which indicate what is happening in a system at an instance in time. Each term of the balance equation is a rate and has units of the balanced

quantity unit divided by a time unit (Felder and Rousseau, 2001). For the esterification reaction, the rate of reaction equation consists of the rate of acid, alcohol, ester and water concentration which assumes that 1 mole of acid and alcohol produce 1 mole of ester and water (Altiokka and Citak, 2003).

For batch reactor considerations, some assumptions have been made in order to derive the energy balance equations. One assumption is that the reactor and jacket are well-mixed (Bequette *et al.*, 2004). Many researchers have come out with mathematical models for batch reactors by using this assumption (Garcia *et al.*, 1995; Aziz *et al.*, 2000; Aziz and Mujtaba, 2002; Arpornwichanop *et al.*, 2005; Hua *et al.*, 2004). The energy balances around the reactor can be described by using the rate of the reactor and the jacket temperature (Arpornwichanop *et al.*, 2005), which consist of the heat released by a chemical reaction (Xaumier *et al.* 2000) and heat released from the heating or cooling system (Bouhenchir *et al.*, 2000).

2.6.2 Empirical Model

The internal model of the IMC strategies is represented by an empirical model. Empirical models are referred to as black box models because the process being modeled can be likened to an opaque box (Romagnoli and Palazoğlu, 2006). The input and output variables (***u*** and ***y*** respectively) are known but the inner workings of the box are not known. Time-varying variables ***u***(t), ***y***(t), and ***d***(t) are shown in Figure 2.4.

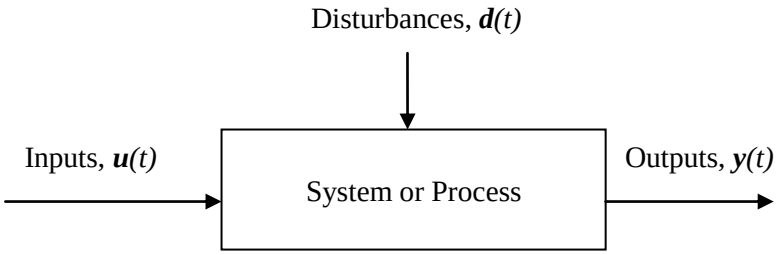

Figure 2.4: A process with inputs *u(t)* and outputs *y(t)*

The capability to construct the mathematical models of a system provides a mechanism for assessing the understanding of the fundamental phenomena in the system by comparing predictions made by the model with experimental data. In the model based control strategy, the best decision on the best control is continually updated using information from this comparison (Rossiter, 2003). Process models may be expressed in many different ways. Such models may be classified into linear and nonlinear models. Although most real processes are nonlinear, for small variations in input variables these processes can be approximated by linear constant coefficient models through a procedure of linearization (Brosilow and Joseph, 2001).

In order to reduce the complexity of the model and to capture important control-relevant system information, the estimated identification model must correctly describe the important dynamics of the system under consideration. This requirement can be satisfied through a careful designed identification experiment design which ensures that the process measurements are informative. The input design problems have been studied by several researchers. Parker *et al.* (2001) classified the input sequence into two types: 'plant friendly' input sequence is a constant sequence while the 'least plant friendly' possible sequence is any sequence that changes at every time step. Godfrey *et al.* (1999) reported that for linear model identification, the input signal amplitude should be small enough to negate the effects