REAL-TIME OPTIMIZATION FOR ESTERIFICATION OLEIC ACID WITH GLYCEROL USING ADAPTIVE INFRARED SENSORS SELECTION

IWAN AANG SOENANDI

SCHOOL OF POST GRADUATE STUDIES
BOGOR AGRICULTURAL UNIVERSITY
BOGOR
2017

DECLARATION OF ORIGINALITY AND COPYRIGHT TRANSFER

Hereby, I declare that the dissertation entitled Real-Time Optimization for Esterification Oleic Acid With Glycerol Using Adaptive Infrared Sensors Selection is my own work, under the supervision of Dr. Eng.Ir. Taufik Djatna, MSi, Prof. Dr. Ir. Ani Suryani, DEA and Dr.Ir.Irzaman, MSi. It has never previously been published in any university. All of incorporated originated references from other published as well as unpublished papers are stated clearly in the text as well as in the reference list.
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Bogor, February 2017

Iwan Aang Soenandi
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RINGKASAN

IWAN AANG SOENANDI. Optimasi *Real-Time* untuk Esterifikasi Asam Oleat dengan Gliserol Menggunakan Seleksi Sensor Inframerah Secara Adaptif. Dibimbing oleh TAUFIK DJATNA, ANI SURYANI dan IRZAMAN.

Monogliserida sebagai salah satu produk esterifikasi yang antara lain adalah gliserol monooleat diperlukan sebagai bahan baku dengan jumlah yang meningkat sepanjang tahun dalam industri obat-obatan, kosmetik dan perawatan tubuh lainnya. Berdasarkan peningkatan tersebut maka produksi dari gliserol monooleat dengan menggunakan bahan baku asam lemak menengah atau rantai panjang memberikan peluang untuk dioptimasi lebih lanjut.

Untuk mengoptimalkan produksi Gliserol MonoOleat (GMO) dengan proses esterifikasi yang memiliki gangguan dan ketidakpastian dalam proses reaksi yang berlangsung di dalam reaktor akan digunakan metode optimasi secara *real-time (RTO)*. Dari penelaahan pada penelitian sebelumnya, ditemukan bahwa aplikasi RTO dapat digunakan pada proses yang berlangsung untuk mengevaluasi variabel dalam operasi, dan dalam perkembangan selanjutnya beberapa penelitan menerapkan aplikasi RTO untuk proses kimia berdasarkan pengamatan dari komposisi produk.

Berdasarkan motivasi utama penelitian ini, dirumuskan lima tujuan besar yaitu: (1) untuk membuat model simulasi proses esterifikasi secara *real-time* dengan dukungan metode *Self-Optimization* (SO), (2) untuk membangun sistem monitoring secara *real-time* dengan menggunakan metode klasifikasi, (3) untuk mengembangkan model optimasi menggunakan RTO pada proses esterifikasi menggunakan kontrol *cluster* secara adaptif, (4) untuk meningkatkan metode RTO dengan sistem seleksi gradien sensor yang adaptif, dikombinasikan dengan metode *classification* pada proses *batch* dan (5) untuk mendesain model *scaling-up* menggunakan metode RTO untuk proses *batch*. Untuk memenuhi tujuan tersebut, sebagai langkah pertama adalah pembuatan analisis sistem awal dengan diagram *Business Process Modelling and Notation* (BPMN 2.0) untuk mendeskripsikan fungsi SO dari alur kerja yang bekerja sama dan berkorelasi sebagai hasil abstraksi pada fase konseptual. Selanjutnya, pemodelan sistem kontrol, yang dibuat menggunakan simulasi *Stateflow*, untuk mensimulasikan tiga kelompok dari *control state* menggunakan klasifikasi dengan metode SVM dan pada akhirnya divalidasi dalam skala laboratorium untuk pengukuran kinerja pada sistem baru yang dikembangkan ini.

Penelitian ini menggunakan hasil validasi dengan skala laboratorium menggunakan metode RTO dengan seleksi sensor secara adaptif, menunjukkan peningkatan hasil hingga 14%, mengurangi durasi proses sebesar 20 menit, kecepatan rotasi pengaduk efektif adalah 300*rpm* sampai 450*rpm* dan suhu akhir pada rentang 200°C sampai 210°C. Rekomendasi dari penelitian saat ini, maka diperlukan peningkatan mutu bahan komponen dari sensor-sensor, terutama sensor yang tahan digunakan dalam suhu dan tekanan tinggi serta integrasi peralatan untuk kontrol yang lebih baik dan cepat.

Kata kunci: Optimasi *Real-Time*, pemodelan, Simulasi, Sensor Adaptif, *Support Vector Machine (*SVM*)*, *State Control*, pengukuran gradien.

SUMMARY

IWAN AANG SOENANDI. Real-Time Optimization for Oleic Acid Esterification With Glycerol Using Adaptive Infrared Sensors Selection. Supervised by TAUFIK DJATNA, ANI SURYANI and IRZAMAN.

The esterification product such as monoglyceride and diglyceride are representing high needs as modifying agents and showing steadily incremental for consumption in the future, as they have various industrial application. This product as a raw material required in pharmaceutical, cosmetics, and personal care industries. Several constraints occur in the production of derivatives of glycerol by esterification process, particularly on the yield of target production which is unstable and lacking of efficiency.

In order to optimize esterification process with glycerol to produced glycerol monooleate which has to cover various disturbance and uncertainty in real-time mode, a Real-Time Optimization (RTO) with adaptively selection sensors was proposed as a new development to the solution. Developed since 1995, RTO has been used to indicate the continuous re-evaluation of selecting variables in operation; furthermore, RTO was used for the current chemical process based on product composition.

According to its main motivation, five research objectives were formulated are (1) to deploy a simulation model of esterification with real-time simulation to support Self-Optimization (SO), (2) to build real-time monitoring system using classification method, (3) to develop a model of optimization in RTO for esterification process using real-time clustering adaptive control, (4) to improve RTO method with adaptive sensors selection system, supported by classification method measured in a batch process and (5) to design scaling-up model using RTO method in a batch process..

To fulfill those objectives first, a Business Process Modelling and Notation (BPMN 2.0) was built to describe the tasks of SO workflow in collaboration with NCO as an abstraction for conceptual phase. Next, to use the model, its implementation with Stateflow package was deployed to simulate the three states of control using SVM classification. Finally, validation was run for this proposed system.

In validation, the result of RTO with adaptive sensors selection showed an increase of the yield up to 14%, process duration reduction by 20 minutes, with the effective stirrer rotation was 300rpm to 450rpm and final temperature between 200°C to 210°C. This improvement, hopefully this new system will be applied for industrial sector. For scaling up recommendation, it is necessary to improve the quality of the sensor component materials especially for high temperature and pressure. Also needed the improvement of control system integration, to make it faster and more precision.

Keywords: *Real-Time Optimization, Modelling, Simulation, Adaptive Sensors, Support Vector Machine (SVM), State Control, Gradient Measurement*

GLOSSARY

Adaptive	:	readily capable of adapting or of being adapted
Arduino	:	an open-source project that created microcontroller-based kits for building digital devices and interactive objects that can sense and control physical devices
BPMN	:	Business Process Modelling and Notation is a graphical representation for specifying business processes in a business process model.
Computational intelligence	:	the ability of a computer to learn a specific task from data or experimental observation
Esterification	:	conversion of an acid into an ester by combination with a n alcohol and removal of a molecule of water
Event-based simulation	:	simulation that operate by taking events, one at a time, and propagating them through a design until a steady state condition is achieved
FFA	:	Free Fatty Acid, when fatty acids circulating in the plasma (plasma fatty acids) are not in their glycerol ester form (glycerides), they are known as non-esterified fatty acids (NEFAs) or free fatty acids (FFAs)
FTIR	:	Fourier Transform Infrared Spectroscopy is a technique which is used to obtain an infrared spectrum of absorption or emission of a solid, liquid or gas
GCMS	:	Gas Chromatography–Mass Spectrometry is an analytical method that combines the features of gas-chromatography and mass spectrometry to identify different substances within a test sample.
Glycerol	:	also called glycerine or glycerin ($C_3H_8O_3$) is a simple polyol compound. It is a colorless, odorless, viscous liquid that is sweet-tasting and non-toxic.
GMO	:	Glycerol MonoOleate are used as emulsifiers or oiling agents for foods, spin finishes and textiles; antifoaming and antistatic agents for plastics; and lubricants, water treatment, metal working fluids, and dispersing agents. End applications include cosmetics, foods, personal care products, medicine, pesticides, paper making, plastics and paints.
Gradient	:	the degree of inclination, or the rate of ascent or descent
Infrared	:	producing or using rays of light that cannot be seen and that are longer than rays that produce red light
K-means	:	cluster analysis in data mining that aims to partition n observations into k clusters in which each observation belongs to the cluster with the nearest mean, serving as a prototype of the cluster

Laplace transform	:	an integral transform named after its discoverer Pierre-Simon Laplace. It takes a function of a positive real variable *t* (often time) to a function of a complex variable *s* (frequency).
Measurement	:	the act or process of measuring something
Microcontroller	:	is a small computer on a single integrated circuit containing a processor core, memory, and programmable input/output peripherals
Modelling	:	graphical, mathematical (symbolic), physical, or verbal representation or simplified version of a concept, phenomenon, relationship, structure, system, or an aspect of the real world
Oleic Acid	:	a colorless, odorless, liquid, water insoluble , unsaturated acid $C_{18}H_{34}O_2$ obtained from animal tallow and natural vegetable oil
Optimization	:	an act, process, or methodology of making something (as a design, system, or decision) as fully perfect, functional, or effective as possible
PID Controller	:	a proportional–integral–derivative (PID) controller is a control loop feedback mechanism (controller) commonly used in industrial control systems.
Pillar K-means	:	an algorithm that has a new approach to optimizing the designation of initial centroids for K-means clustering
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RTS	:	Real Time Simulation refers to a computer model of a physical system that can execute at the same rate as actual "wall clock" time. In other words, the computer model runs at the same rate as the actual physical system
Scale-up	:	the migration of a process from the lab-scale to the pilot plant-scale or commercial scale
Selection	:	the act of choosing something from a group
Sensor	:	a device that detects and responds to some type of input from the physical environment

Simulation	:	imitation or representation, as of a potential situation or in experimental testing
State	:	the system's modes of operation, represent the logic for switching between modes using transitions and junctions
Stateflow®	:	an environment for modeling and simulating combinatorial and sequential decision logic based on state machines and flow charts
SO	:	Self-Optimization is a process in which settings are autonomously and continuously adapted for optimizing the system
SVM	:	Support vector machine are supervised learning models with associated learning algorithms that analyze data for classification and regression analysis
SQL	:	Structured Query Language is is a special-purpose domain-specific language used in programming and designed for managing data held in a relational database management system

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IWAN AANG SOENANDI

Dissertation

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PREFACE

I would like to thank Almighty God for all His gifts so that this research was finally successfully completed. The theme chosen for the research, which conducted, starting June 2014 up to May 2016 is Real-Time Optimization using sensor supported with computational intelligence, with entitled Real-Time Optimization for Esterification Oleic Acid With Glycerol Using Adaptive Infrared Sensors Selection.

Especially, I would like to express my sincere gratitude to Dr Eng Ir Taufik Djatna as Supervisor for his support and encouragement during my study in Bogor Agricultural University, to Prof Dr Ir Ani Suryani as First Co-Supervisor for her advice and supervision during the dissertation work, Dr Ir Irzaman as Second Co-Supervisor for his support during the dissertation work, to Prof Dr Ir Machfud, MS as head of Graduate and Post Graduate Program in Agroindustrial Technology Study Program, to Prof Dr Eng Khairurrijal and Dr Dwi Setyaningsih STP, MSi as external examiners for their advices during examination. Additionally, I would like to thank Ministry of Research, Technology and Higher Education of the Republic of Indonesia as a sponsorship of my doctoral study and research.

I would like to say many thanks to my family, my father Henky Bambang Soenandi, my mother Netty Suryadi, and my wife Karina for their true and endless love for never failing patience and encouragement. I would like to thank all lecturers and staff of Agroindustrial Technology Department, all of my colleagues, especially my Phd student mates in Agro-industrial Technology particularly Rahmawati, Gunawan and Azrifirwan, colleagues in Computer Laboratory of Agro-industrial Technology Department particularly Hadi, Yoga, Denny, Rino and Imam, all of my student colleagues in Agro-industrial Technology 2013 and 2014, and also special thanks to my colleagues in Krida Wacana Christian University such as Meriastuti Ginting, Oki Sunardi, Budi Marpaung, Johansah Liman and Budi Harsono for their support. It has been a pleasure to work and share with you all.

Hopefully, this dissertation is useful.

Bogor, February 2017

Iwan Aang Soenandi

BIBLIOGRAPHY

Iwan Aang Soenandi was born in Bandung on July 23rd, 1977 as first son from Bambang Soenandi and Netty Suryadi. The author received his Bachelor degree in Mechanical Engineering from Atma Jaya Catholic University Jakarta and Master Degree in Industrial Engineering from University of Indonesia in 2000 and 2002, respectively. Next, followed by entering Doctoral Programs in Post Graduate School of Bogor Agricultural University, majoring Agro-industrial technology in September 2013 with sponsored by Ministry of Research, Technology and Higher Education of the Republic of Indonesia on BPPDN 2013 program. Currently, he works as lecturer and researcher at Industrial Engineering Department Faculty of Engineering and Computer Science in Krida Wacana Christian University Jakarta. His several current research interest, related to this dissertation partly, such as discrete simulation, real-time optimization and adaptive control has been published and presented in several international conferences. First, an article with title "A real time simulation model of production of glycerol esterification with Self-Optimization" was presented and published on ICACSIS 2014 (indexed in IEEE and Scopus). Second, an article with title "Optimization process of glycerol esterification using real-time adaptive control" was presented and published on ICACSIS 2015 (indexed in IEEE and Scopus). Third, an article with title "Real time optimization using gradient adaptive selection and classification from sensor measurement for esterification glycerol" has passed the second review stages in International Journal of Intelligent Computing and Cybernetics (Emerald Publishing) that indexed and abstracted in: Scopus, CSP, EI Compendex, INSPEC, MathSciNet, QUALIS, Read Cube Discover, Zentralblatt MATH.

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1 INTRODUCTION

Background

Esterification product such as monoglyceride and diglyceride are representing high demand as modifying agents and showing steadily incremental demand for consumption in the future, as they have various industrial application, particularly as the main raw material in pharmaceutical, cosmetics and personal care, as well as in ink manufacturing (Pagliaro and Rossi 2008; Prasetyo *et al.* 2012; Fernandez *et al.* 2005). Another use for derivative products of glycerol is for mixing substances to produce OBM (Oil Based Mud) and WBM (Water Based Mud) in oil mining industries in Indonesia. Production of Monoglyceride from esterification of glycerol and synthesis of middle or long-chain fatty acids offers promising industrial opportunities (Mostafa 2013). Then, to optimize the esterification reaction, it is essential to conduct it in real-time mode, which has to cover various disturbance and uncertainty. In recent years, spectroscopic methods using infrared has gained popularity to be chosen for industrial process control in real-time, especially for esterification process (Blanco *et al.* 2004).

In esterification, as previously researched (Hui 1996) there is some influence of process variables, such as temperature and time requirement in reactor that affected their efficiency. In esterification process has several constraints, such as inconstant yield of production target and lack of efficiency as there is a synthesizing reaction that requires high-energy use in the reactor to achieve desired temperature and time for the process (Mostafa 2013).

As the stage in these dissertation, in order to optimize the esterification process in real-time mode, this research started with challenges are to model a Self-Optimization in real-time simulation of esterification, to build a real-time classifying system of esterification, to develop an optimized model for esterification process using real-time clustering adaptive control, to improve esterification process with real time adaptive sensors selection system, and to design a scale-up esterification process in proposed model for batch production.

For the purpose to solve those objectives, we used RTO concept that was developed based on SO, which is defined in Skogestad (2000) as an achievement for an acceptable loss with constant set point values for the controlled variables, without has to re-optimize when has disturbances. This method was combined with Necessary Condition Optimum (NCO) (Srinivasan and Bonvin 2007). However, according to Ye *et al.* (2012), it is necessary to measure all of the NCO components in real-time using sensors. For this research, we claimed of this new optimization method, supported with measurement using sensors with several types in medium wavelengths in IR spectrum named mid-IR and a real-time data acquisition system supported with computational intelligence was new. This new method also useful for chemical industry application that has dynamic condition such as in esterification process.

To deploy a requirement model of the system, analysis and design of the dynamic condition of glycerol esterification production in detail was built on Business Process Model and Notation version 2.0 (Lin *et al.* 2002). This new model implemented as support by computational intelligence methods such as Support Vector Machine (SVM) for classification (Vapnik 2000) and Pillar k-means for adaptive clustering (Barakbah 2010). Next, to determine the state group in control the proposed system consists of several variables such as temperature and stirring speed of the reactor. Finally, by finding optimization parameters that have to be fulfilled, which consisted of process time and yield, focuses on the production of Glycerol Monooleate (GMO) with 130 minutes observation time.

Problem Statement

It has always been a big challenge for chemical industries to develop the production system as regard to its complexity. In order to optimize esterification process in real-time mode, RTO was chosen as a development method which involves control variable in its process. Thus, there were several problems related to the optimization of esterification based on RTO, as follows:

1. How to model a self-optimization in real-time simulation of esterification?
2. How to build a real-time classifying system of esterification?
3. How to develop an optimized model for esterification process using real-time clustering adaptive control?
4. How to improve esterification process with real time adaptive sensors selection system?
5. How to design a scale-up esterification process in proposed model for batch production?

Objectives

Based on the problem statement above, there are five main objectives formulated in this research as follows:

1. to model a self-optimization in real-time simulation of esterification,
2. to build a real-time classifying system of esterification,
3. to develop an optimized model for esterification process using real-time clustering adaptive control,
4. to improve esterification process with real time adaptive sensors selection system,
5. to design a scale-up esterification process in proposed model for batch production.

Benefits

The most accomplished result in integration method RTO with adaptive selection for future implementation was contributed to support a prediction of production volume with real-time simulation, improving identification of esterification stage in real time mode, deploying states control model precisely with adaptive selection sensor and operating more efficient in a batch process.

Boundaries

In order to focus on the solution as the mentioned issue, we are doing all the experimental studies in laboratory scale in batch system. We also used some assumption in this research such as the reaction was conducted with raw material of glycerol 80% and 90% purity, mixed with oleic acid in 1:1 ratio and focused on the target production of Glycerol Monooleate. The process was observed in range between 90 to 130 minutes using catalyst Methyl Ester Sulfonic Acid (MESA) 0.5% and the performance of apparatus was in normal and standard condition. The product of esterification was tested with FTIR and GCMS to measure the yield of Glycerol Monooleate.

Novelty of Research

According the combinations of methods that was developed. This dissertation has several novelties such as the application of analysis and design in esterification process using BPMN 2.0 diagram, the model deployment for real-time measurement using sensor in esterification process and the development for adaptive real-time monitoring and control combined with computational intelligence to support Real-Time Optimization.

2 METHODS

Simulation and Laboratory Experiments

In simulation stages, a group of synthetic data for quality target that found from literature was used to simulate the process of esterification to measure the performance of the system using real-time discrete simulation and found the related parameter of quality using feature selection method.

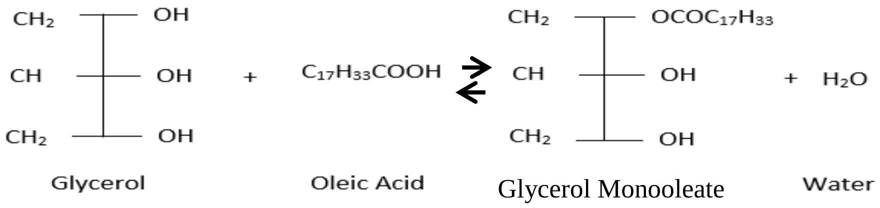


Figure 1 Esterification reaction (Pagliaro and Rossi 2008)

In laboratory scale, the reaction as described in Figure 1 was performed in a four-necked glass reactor with a 1000 ml volume. Glycerol mixed with oleic acid and required amounts of catalyst MESA were used for each experiment. The catalyst amount was based on weight percentage of oleic acid. The reaction time was defined as zero when the temperature reached the set point since negligible conversions were observed. The stirring rate was set at 200rpm up to 500rpm related to the stages of process. The catalysts were added to the reaction mixture when the temperature reached 65 °C

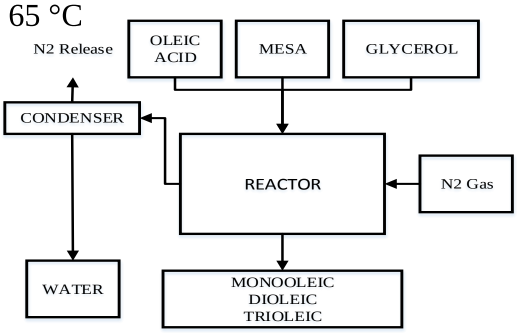


Figure 2 Esterification process diagram (Hui, 1996)

In esterification process as described in Figure 2 has several constraints, such as inconstant yield of production target and lack of efficiency as there is a synthesizing reaction that requires high-energy use in the reactor to achieve desired temperature and time for the process (Mostafa 2013). In this process also has several disturbance that involved in the reaction such as variance of purity level of glycerol and oleic acid as a raw material. Another disturbance was from environment of the system such as outside temperature variance of the reactor. The photo of reactor in laboratory experiments was shown in Appendix 1.

The yield of esterification process as focused in this research was measured by the transmittance of infrared sensors with several wavelength based on Planck theorem that the transmittance level in each frequency has related in energy measurement of molecules. This sensor was selected within several mid IR wavelength spectrum which has high difference of transmittance level (peak) and the measurement was calibrated by compared the data transmittance using Fourier

Transform Infrared Spectroscopy (FTIR) and Gas Chromatography–Mass Spectrometry (GCMS) (Kartnaller *et al.* 2016).

Research Framework

The research framework was designed in a parallel form to solve the problem which begins from a real-time simulation model of self-optimization and then developing in real-time monitoring. Finally, optimization in real-time with an adaptive sensor selection to determine the control state. In this research, microcontroller ATmega8535 (Atmel 2005) and Arduino (Ivrea 2005) were used as a hardware of data acquisition. For the tools such as BPMN 2.0 (SAP 2013), Sigmaplot 12.5 (Cranes 2011), Arena (Rockwell 2011), Orange Python (Biolab 2015) and Stateflow (Mathworks 2014) were used in software for process modeling, create exact graphs, discrete simulation and data processing.

In this research, generally, experimental data from laboratory scale practices of reaction were used to generate several real-time optimization models of esterification production using clustering, classification, and adaptive selection sensor. The detail of general research framework and method in each chapter are presented in Figure 3.

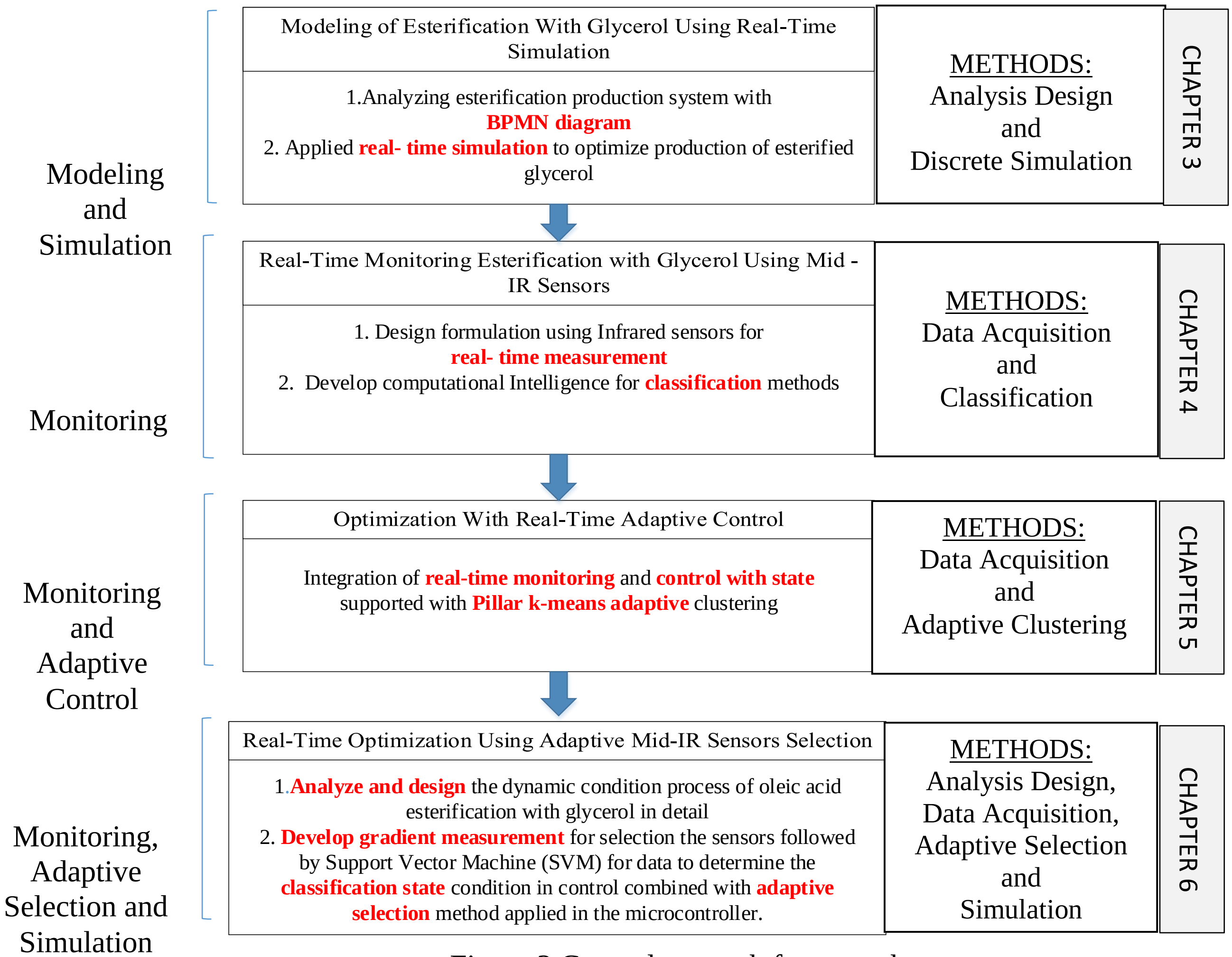


Figure 3 General research framework

Location of Research and Time

This research was started from June 2014 up to May 2016. The sensors and detectors were tested and assembled in Laboratory of Electronic Material and Physics in Department of Physics Bogor Agricultural University. For data analysis and computational process was run in Laboratory of Computational Research Department of Agroindustrial Technology Bogor Agricultural University. We run several reactions and variation conditions for esterification process that were tested in laboratories of Surfactant Bioenergy Research Center (SBRC) at Bogor Agricultural University.

Method of Collecting Data

Data collection was done based on the needs of the developed model. In this research types of data can be either primary data or secondary data. Primary data was obtained from mid-IR sensors that works in real-time and laboratory assay test such as Fourier Transform Infra-Red (FTIR) and Gas Chromatography–Mass Spectrometry (GCMS). Secondary data in this study were collected from books, reports, and scientific publications and other sources of information.

3 A REAL-TIME SIMULATION MODEL OF PRODUCTION OF GLYCEROL ESTERIFICATION WITH SELF-OPTIMIZATION

Abstract

The quality and capacity of Glycerol Monooleate (GMO) production are the factors that need to monitor and control. In this model of Real-Time Simulation (RTS), we deployed BPMN diagram to analyze and design the requirement and used data acquisition system with the real-time sensor. We used real-time data acquisition by optical sensors to acquire several quality parameters such as viscosity, pH, purity and density which was interfaced to an SQL database. The model of self-optimization for quality surveillance was based on previous work. As a result of RTS, showed that the evaluation of the system performance was worked to optimize a range of parameter esterification glycerol with oleic acid in producing Glycerol Monooleate (GMO) such as optimized process time to 2.888 hours and volume at 28L for each batch. Compared to the traditional works was set to a fixed value 24L output only and 3.4 hours process time, that was increased about 16% volume of production for each batch and quality parameters that has closely related are density, viscosity and purity. In future work, it is recommended to accelerate the data processing including to re-structure the real-time monitoring sensor system.

Introduction

This chapter presents a newer concept of a Real-Time Simulation (RTS) compared to traditionally RTS that based on offline simulation and develop a model of the Glycerol Monooleate(GMO) production with Self-Optimization (SO) as a support for RTS. The integration of advanced data acquisition for optimization and RTS offers considerable potential for innovations in the field of conventional system. As compared to previous research by Sanchez in 1995, there is dynamically condition that related to performance which is offline simulation has problem in time delayed to catch that situation to get optimization for product quality and volume. To increase the system performance that affected by many internal or external factors in the process, such as quality of end product could use self-optimization (Schmitt and Beaujean 2010). Using Self-optimization (SO) on process level, for example to calculate machine and process parameters according to changing outer conditions (Wagels and Schmitt 2012). In GMO process production there exist many possible scenarios how to solve these problems, but which of these scenarios is the best or the most optimal one? Is it possible to imagine how a change in the subsystem affects the entire system? As one a product of agroindustrial sector, this chapter was focus on glycerol as by-product of biodiesel, that the production of biodiesel is still increased, because of the government policy that regulates this GMO components for mixed material for biodiesel had affected in increasing glycerol production rate. As a raw material, glycerol is an oil soluble food additive is also used as an ingredient in the production of chewing gum and ice cream (Soares *et al.* 2011). To produce esterified glycerol that using raw material from crude glycerol, there are many small medium manufacturing that used conventional process and has many customer requirements to make Glycerol Monooleate (GMO).

One of motivational point in optimization in GMO production is the quality of product may vary from the customer requirement. That condition would affect the unstable volume of production as this requirement observed and controlled manually. Our research for the optimization and simulation in a real-time mode to monitor the flow of production and find the best production parameter with SO.

The objective of these chapter is to construct a model that applied the RTS method supported with SO in implementation for Quality Control (QC). This method was explained with BPMN diagrams, using sensors and data acquisition system and finding the parameter of production that closely related to the quality target of the product and the volume of production GMO.

Materials and Methods

Production of Glycerol Monooleate

The complexity of GMO production using reactor which is charged with materials glycerol that was explained in Chapter 1 at introduction, and reactor that put in a heater apparatus with working temperature maximum at 260°C were used.

Business Process Modelling

This stage is important because with this modeling we can analyze all the activities taken in production, especially in esterification process. We referred a Business Process (BP) as a collection of related and structured activities undertaken by one or more organizations or process in order to pursue some particular goal. Within an organization, a BP results in the provisioning of services or in the production of goods for internal or external stakeholders. Moreover, BPs are often interrelated since the execution of a BP often results in the activation of related BPs within the same or other organizations (Grooskopf *et al.* 2009). The primary goal of BPMN is to provide a standard notation readily understandable by all business stakeholders (Ryan *et al.* 2009), this modeling notation method has become an highly attractive topic both for industries and for the academy such as: the BPMN 2.0 (Stephen and Conrad 2011).

Quality Control (QC)

In testing sample of GMO with FTIR, Fourier transform infrared spectroscopy (FTIR) is a technique which is used to obtain an infrared spectrum of absorption or emission of a solid, liquid or gas. An FTIR spectrometer simultaneously collects high spectral resolution data over a wide spectral range (Brault 1996). This confers a significant advantage over a dispersive spectrometer which measures intensity over a narrow range of wavelengths at a time. We set up QC system using mid-IR optical source and a detector for glycerol ester peaks for identification to meet the quality needed as the level of transmittance for the requirement in GMO production, as detailed specification of this requirement was provided in Business Process and Model Notation (BPMN) diagram as described in experimental results.

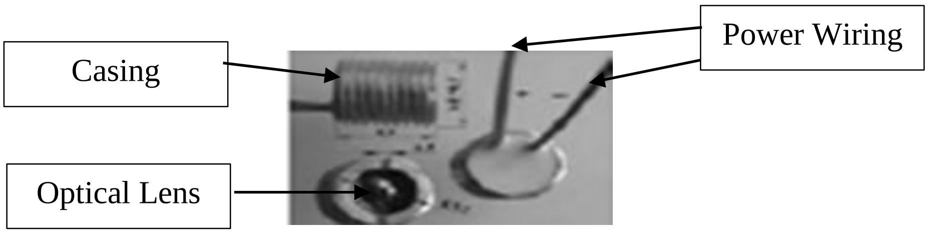


Figure 4 Mid-IR optical source

Self-Optimization

In order to design a esterification process with optimum parameters. A combination of SO and simulation with real-time data acquisition method was proposed with IR sensors as shown in Figure 4, this illustration refer to specification of mid-IR sensor from Boston Electronics (Boston 2014) such as casing, optical lens and power wiring. The behavior of this optimization algorithms is random, so we had to perform many optimization experiments to identify the pure nature of. By considering the number of simulation experiments we can divide the number of simulation experiments as simulation experiment that simulation run of simulation model, Optimization experiment that performed with concrete optimization method setting to find the optimum of objective function and series that replication of optimization experiments with concrete optimization method setting. In this chapter the simulation and optimization experiment was used to find the optimum of the objective function.

We specified the same conditions to satisfy each criteria in optimization such as termination criteria, search space for the global optimum. Optimization experiment method has the same parameters as another optimization method.

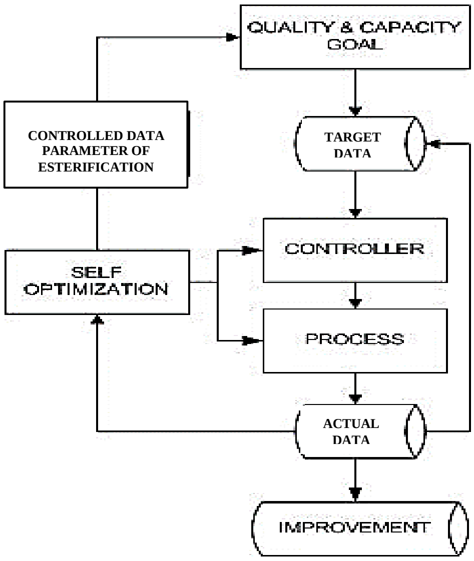


Figure 5 Self-optimization conceptual diagram
(modified from Adelt 2009)

The optimization algorithms doing it by automatically as diagram in Figure 5 that described the SO was operate as controller and process to set the controlled parameter adaptation of esterification to achieve the requirements of quality goal

Self-optimizing systems are defined as the interaction of contained elements and the recurring execution of the actions (Adelt *et al.* 2009), that has continuous analysis of the current situation, in this case is parameter selected of pure glycerol, determination of quality targets of GMO, production capacity, and adaptation of the system's behavior to achieve these targets like temperature setting and the stirring speed of reactor.
In this chapter we concerned for the SO formulation in general to find the minimization of process time with selected parameter as in Equation 1

$$\min\ (\mathbf{t}) \leftrightarrow (\mathbf{P}) \qquad (1)$$

Where t$\in T$ T is time needed in process and P is process with selected parameter of quality. For the more detailed formulation we described in Equation 3.

The development of a simulation of an actual system involves the creation of a conceptual model of the actual system to be simulated, which may be based on a set of rules, or a set of mathematical equations, or some other method of defining the state of the simulation and the way in which it changes with time. A simulation based on a discrete model establishes an initial state of the system and a future event queue with event timings. Thus, an event-based simulation in which time advances from event to event in a single software thread has been the basis of many popular discrete simulation languages, but, as parallel computing options increase, process -based simulation using parallel processors and multiple software threads has become the most popular approach.

Real-Time Simulation

Real time simulation is based on real-time systems that differ from traditional data processing systems, they are constrained by certain nonfunctional requirements (e.g. dependability and timing constraints or requirements). An efficient simulation of the real-time system requires a model that satisfies both simulation objectives and timing constraints (Bergero and Kofman 2010). There were several previous research developed in a structure and architecture for automatic simulation model generation. Theirs very detailed simulation models intended to be used for real-time simulation based shop floor control (Lee and Fishwick 2001). They identified two essential stages to be automated for automatic simulation model generation: System specification and the associated model construction (Law and Kelton 2000). In this work, a proposed methodology for building an Arena simulation model from a resource model as seen in Figure 6. This was made possible, the Arena simulation software is supported by Visual Basic Application (VBA) coding,

With this application which enables application integration and automation we deploy the simulation model. Then, undertook the development of efficiently model real-time systems to satisfy given simulation objectives and to achieve arbitrary timing requirements and used SQL for running the simulation.

According as the method of this research, we developed the simulation of GMO production that performed the RTS concept with discrete event computer simulation program. The model deployed in ARENA Version 12 (Rockwell Ltd 2011) with the schema in Figure 6 that has block of module such as create entities, VBA coding, esterification process, quality control and database that built input measurement with mid-IR sensor. The data was collected to database after the system of process has reached steady state condition

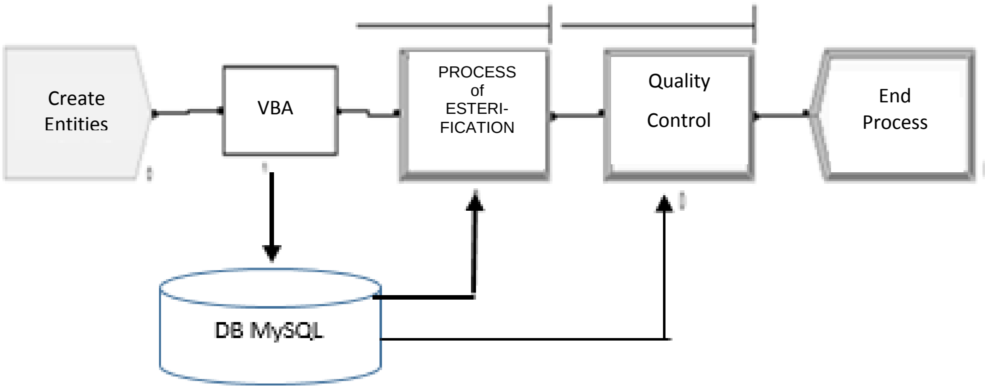


Figure 6 Real-time simulation model of GMO production using discrete simulation

As seen in above simulation diagram. To provide data transmission smoothness, a new system has several block function such as front end, database, microcontroller and sensors to operate the system. We designed the system block for processing signal from control panel that arrange the flow of data from sensors to interface with an SQL connection for database building. In this work we deployed MySQL (Oracle 2014) as data access and repository media by using microcontroller.

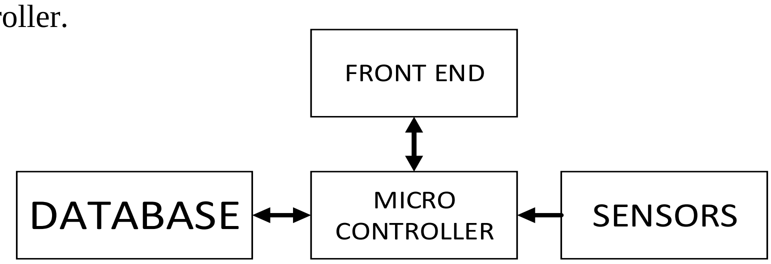


Figure 7 System block diagram for real-time data acquisition

In this chapter as shown in Figure 7, we implemented a real time simulation that need to support by sensors block to do the simulation with little time delay with theirs respectively as special function of microcontroller and front end to built the database in real time that needed in simulation.

Next, we provided relevant tables designed in SQL engine and it is related to simulation as in Table 1.

Table 1 List of data type for SQL database

Field	Data Type	Related to simulation
Time	Time	Parameter
Sensor 1.2 µm	Numeric	Esterification
Sensor 3.4 µm	Numeric	Esterification
Sensor 5.5 µm	Numeric	Esterification
pH Sensor	Numeric	Quality
Viscosity Sensor	Numeric	Quality
Temperature	Numeric	Variable
Stirring speed	Numeric	Variable

In this new concept, analog signal from sensors was converted to digital as interfaced to database using microcontroller in less than 2 seconds for each data stream and RTS works based on simulation running in discrete simulation software using the data from database.

Feature Selection

Feature selection was used in this research because there is many attributes to measure for controlled the quality. To select the parameter that closely related we used Relief (Reliable Elimination of Features) method. This experiment used two class which are good and not good measurement. The result of weights of design elements of Relief a weight of the ith feature is then updated, the mathematical model was stated by Kira and Rendell in 1992 is:

$$W_i = W_i + (x_i - NM_i(x))^2 - (x_i - NH_i(x))^2 \qquad (2)$$

where W_i= Weighted vector of ith attributes, x_i= ith feature vector, $NM^i(x)$= ith the closest different-class instance called as near miss where the later class (ith+1)in the record is differed from the from the ith record, $NH^i(x)$= ith the closest same-class instance called as near hit where the later class (ith+1) is similar from the ith record. That formulation we used is to find the parameter closely related to determined quality of GMO (Equation 2). We used them to reduce unimportant features that result for a more efficient computation and sensor task.

Optimization formula

We concerned for is the SO formulation for optimization that implemented Relief algorithm for attribute selection and search the minimization of time process to increase the volume of production, which is topR→R<<N, N is number of all attribute. We defined formulated completely as:

$$\underset{t \to T}{\arg min} f(t) = \{t | t \in T \wedge R \leq N \wedge topR \ll N \wedge f(t)_R < f(t)\} \qquad (3)$$

where T={t_1,t_2,......t_N}

Next, in this research we found the minimum of process time by doing several of replications in real time discrete simulation. To increase the precision of simulation result, 20 times of replications was run in the real time simulation.

Verification

For the verification stages we set the logical boundaries as acceptance test and bias allowable for the parameter of time which is between 2.5 to 3.5 hours and for volume of production which is between 23-30L.

Experimental Results

Parameter Selection

In order to propose some updates to improve the reliability of the probability approximation and make it robust to incomplete data, and generalizing it to two-class quality problems which are yes and no pass. We collected the data as needed for Relief method the features for attributes A (a_1,a_2,….a_k) from synthetic 30 datasets as listed in Appendix 5 completely. And some sample was tested by the set of quality condition of GMO as shown partially in Table 2.

Table 2 List of attribute of parameter for quality control

% Water (x_1)	Viscosity (poise) (x_2)	Free Glycerin (%) (x_3)	pH (x_4)	Purity (%) (x_5)	Rel. Density (x_6)	Quality Pass
0.5	0.0548	3	4	72	1.01	NO
0.1	0.0543	1	4	63	0.98	NO
0.4	0.0549	3	3	88	1	YES
0.4	0.0543	2	3	71	1.01	NO
0.2	0.0526	3	4	83	0.96	YES
0.2	0.0519	2	4	62	1.02	NO
0.1	0.0513	3	5	78	0.96	YES

Source: (Pardi 2005;Prasetyo 2012)

The quality that pass or not pass was based on research from Pardi (2005) and Prasetyo (2012) that we define in Table 2 in condition as

$$Quality_{pass} = \begin{cases} Yes(x_1 < 0.5; x_2 > 0.050; x_3 \leq 3; 3 \leq x_4 \leq 5; x_5 > 77; 0.96 \leq x_6 \leq 1 \\ No(x_1 < 0.5; x_2 < 0.050; x_3 \geq 3; 6 \leq x_4 \leq 7; x_5 < 77; 1.01 \leq x_6 \leq 1.5 \end{cases} \quad (4)$$

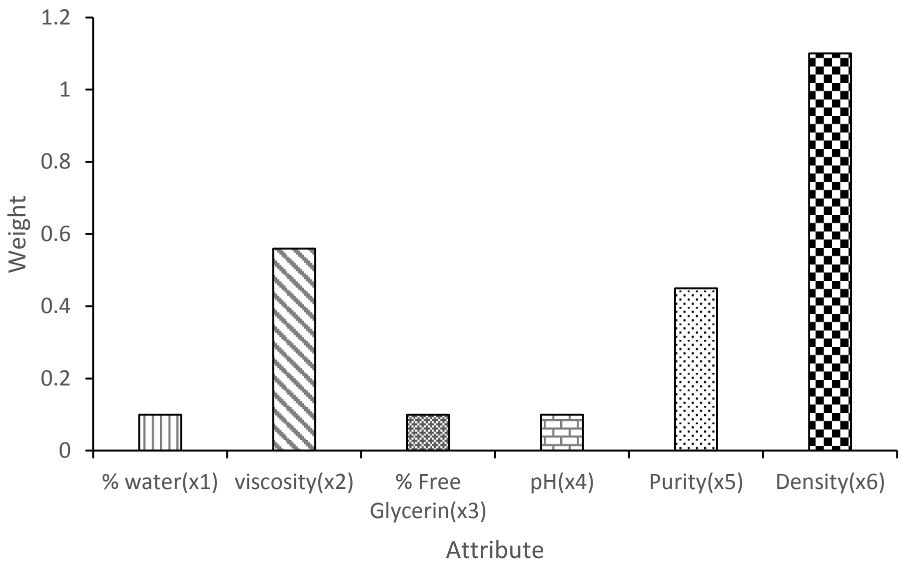
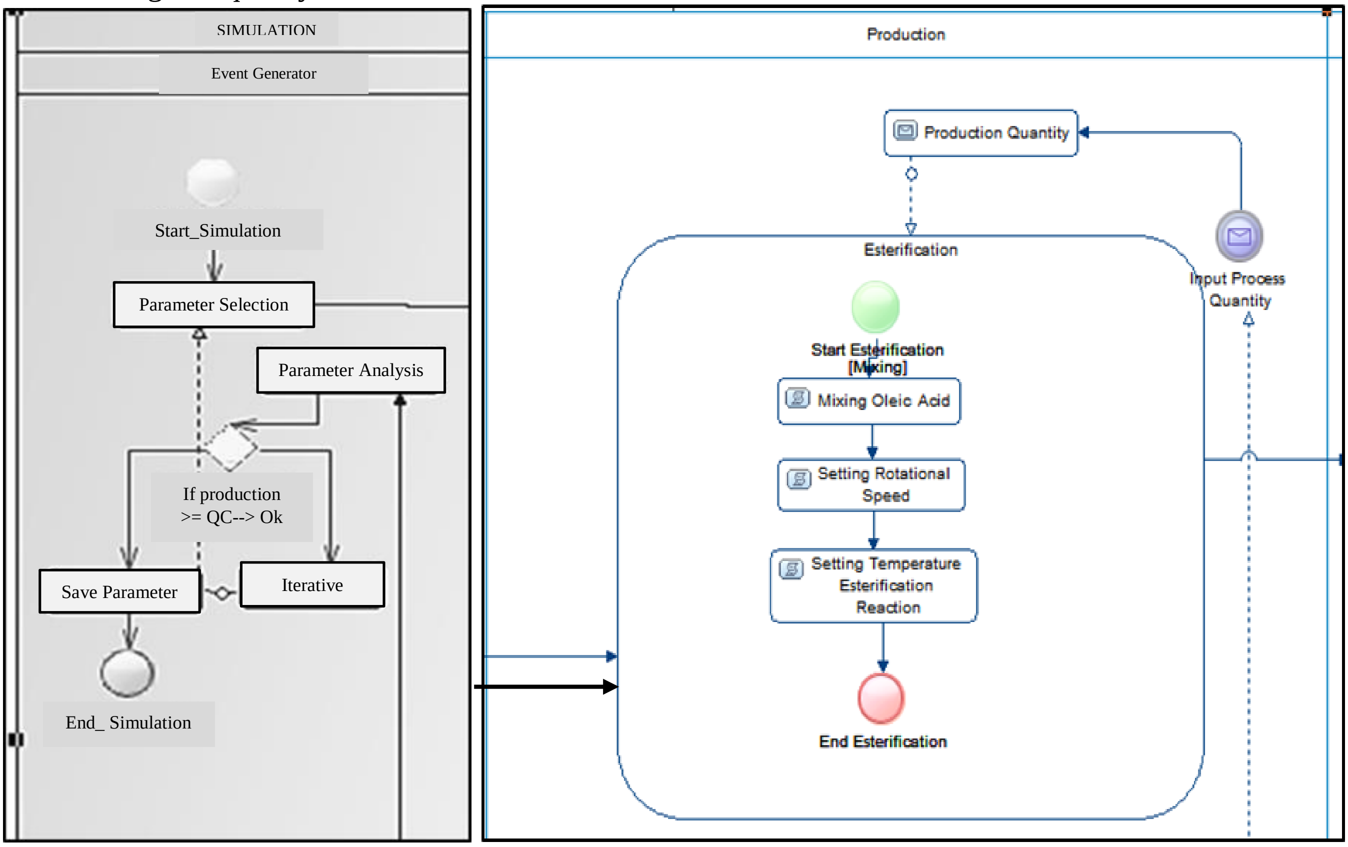


Figure 8 Attribute weight for esterification parameter from training of feature selection based on 30 synthetic data

As an output in Figure 8 has shown the attribute weight W_i for each parameter. The selection of attribute weighted by using Relief method found that the parameters with dominant weight in sequent are density, viscosity and purity.

Business Process Modeling of Esterification with Glycerol

Based on the research from Lin *et al.* (2002) was stated that BPM supports Business Process (BP) experts that providing methods, techniques, and software to model, implement, execute and to optimize BPs which involve humans, software applications, documents and other sources of information. As a widespread adoption of the Business Processing Modelling and Notation (BPMN) also helps unify the expression of basic business process concepts. In this chapter the complexity decomposition of esterification process control was illustrated by using diagram of BPMN 2.0 (SAP 2013) as in Figure 9A as a swimlane of simulation event generator and 9B as swimlane of production. As this analysis, we explained the low-level tasking in the system and simulation that can improve the efficiency and effectiveness of the system (Gunasekaran and Kubo 2002) in order to produce the targeted quality of GMO.



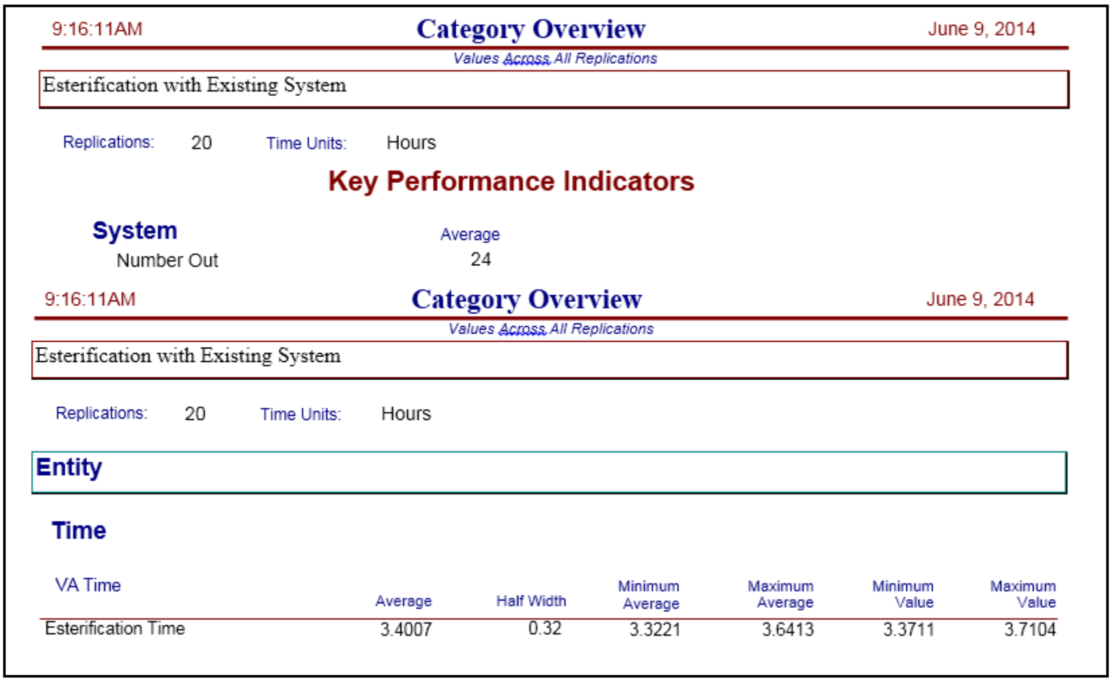
(A) (B)

Figure 9 BPMN of esterification process detailed for (A) swimlane of simulation event generator and (B) swimlane of production

In addition in Figure 9A we defined a parameter analysis block which is required to simulate the esterification process and parameter selection as related by the purity of glycerol as raw material obtained in variations between 80 to 90% due to the presence of impurities such as remaining catalyst, water, soaps, salts and esters formed during the reaction (Min and Lee 2011). For the parameter of density and viscosity has several variations that need to analyze (Pardi 2005). Parameter analysis was set in condition related to pass in QC. In production swimlane as a connection from event generator that appointed where esterification process has several task such as mixing, setting of stirrer speed and setting temperature that was illustrated in Figure 9B. Finally, with BPMN diagrams we was deployed all the process of production by reaction of esterification that involving many parameters affected quality of the end product clearly. as well as advanced process concepts (e.g., exception handling, transaction compensation).

Output of Real Time Discrete Simulation

For data acquisition, a data measurement was interfaced with MySQL and inputs from optical sensors that collected after achieving steady state condition in process production. Then, in Figure 10 the performance in existing system was run in 20 replications and shown the result in Key Performance Indicator (KPI) as number out as represented the volume of production in 24L and average esterification time in 3.4007 hours.



9:16:11AM **Category Overview** June 9, 2014

Values Across All Replications

Esterification with Existing System

Replications: 20 Time Units: Hours

Key Performance Indicators

System	Average
Number Out	24

9:16:11AM **Category Overview** June 9, 2014

Values Across All Replications

Esterification with Existing System

Replications: 20 Time Units: Hours

Entity

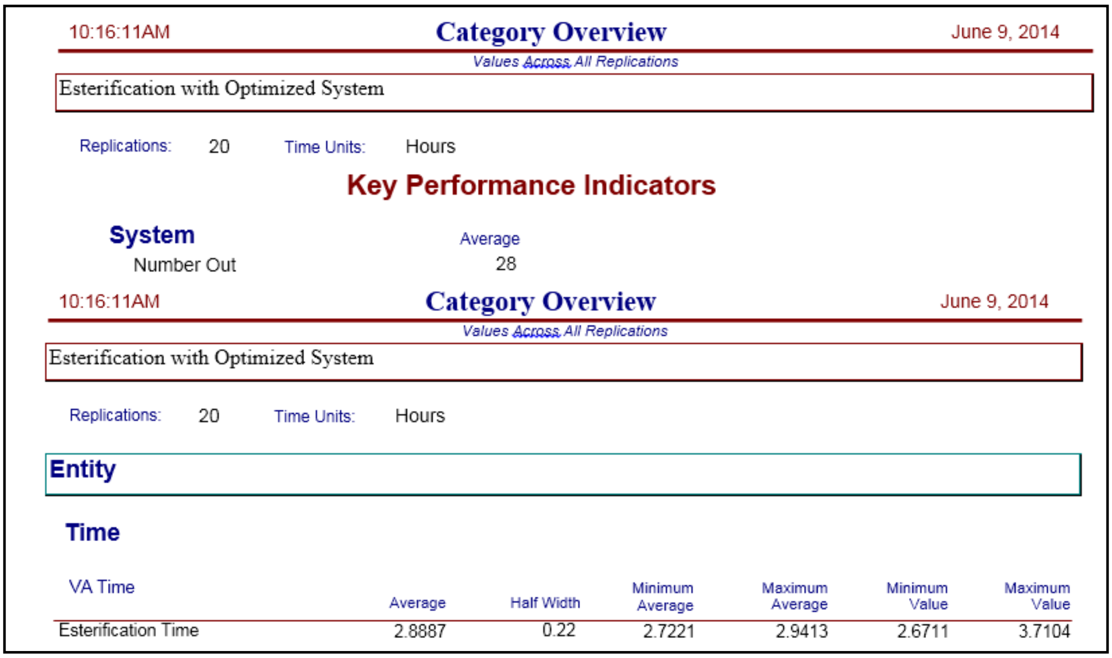
Time

VA Time	Average	Half Width	Minimum Average	Maximum Average	Minimum Value	Maximum Value
Esterification Time	3.4007	0.32	3.3221	3.6413	3.3711	3.7104

Figure 10 Output of simulation result for 6 parameters of QC

From Figure 10 as an output of real time simulation result also described another parameter such as time unit, half width, minimum average, maximum average, minimum value and maximum value. Those output was important to measure the performance of the system.

In addition in Figure 11 we showed the optimized proposed system using SO compared to existing system, that the KPI represented volume of production was increased to 28L and decreased in average esterification process time to 2.887 hours with the differences 0.6 hours as equal to 36 minutes. And also described also described another parameter such as time unit, half width, minimum average, maximum average, minimum value and maximum value. Those output was important to measure the performance of the system.



10:16:11AM **Category Overview** June 9, 2014

Values Across All Replications

Esterification with Optimized System

Replications: 20 Time Units: Hours

Key Performance Indicators

System

	Average
Number Out	28

10:16:11AM **Category Overview** June 9, 2014

Values Across All Replications

Esterification with Optimized System

Replications: 20 Time Units: Hours

Entity

Time

VA Time	Average	Half Width	Minimum Average	Maximum Average	Minimum Value	Maximum Value
Esterification Time	2.8887	0.22	2.7221	2.9413	2.6711	3.7104

Figure 11 Output of simulation result for 3 parameters of QC

With real-time simulation we identified the effect of each process in esterification and quality control. Next, the simulation was run in 20 replications and compared to existing system that operated by knowledge to set the variable of process and monitored manually by operator as described, we found in the simulation result from optimized system has increased in number out parameter in Key Performance Indicators.

Table 3 Comparison of existing and real-time simulation with SO

Parameters	Existing	SO
Esterification time (hours)	3.4000	2.888
Volume (L)	24	28
Quality Parameters	All	density, viscosity and purity

As we compared in Table 3, to find the optimal output of SO for the best parameter in production of esterification glycerol that previously selected with Relief to find the related quality parameters such as density, viscosity and purity, the esterification time was reduced from 3.4 hours to 2.888 hours, volume of production was increased from 24L to 28L, and quality parameters that closely related are density, viscosity and quality.

Discussion

In esterification process, to select the parameter that closely related to self-optimization model of GMO production we used Relief method that compared the attribute weighted from each parameter by collected the data from requirements of GMO specification. In comparison to the previous research (Pardi 2005, Prasetyo 2012) with many parameters was used. We found several parameters that affected the quality of GMO such as density, viscosity and purity with R equal to 3. We have not run the experiments for R (selected attribute) more than 3 because as in training result of weighted attribute the weight number for the others than 3 that we have selected their weight are very small. As implementation of SO those three parameters were identified with sensors.

In comparison to previous research from Wagels and Schmitt (2012), that SO had increased the productivity of shaft production within range of 4.64-7.23%. This current real-time discrete simulation was to evaluate the implementation of proposed system. As the result in simulation report in Figure 10 and Figure 11, we found that a difference in time for processing, output of production had been proven and increased the volume of production up to 16% or from 24L to 28L as a simulation overview output results.

According to the result of this chapter, the difference was related to the parameter that involved in QC for esterification. The advantages of this chapter by optimizing these three parameters which was defined with SO. And by real-time discrete simulation, we can simulate the efficiency improvement of esterification process and increase the volume of production compared to the existing system as shown in Table 3.

As a result of successfully implementation SO in esterification process, in the next chapter presented the development of real-time optimization based on real-time monitoring system and computational intelligence. This new optimization also used data acquisition and sensors system to increase measurement of the parameter related to esterification process.

Conclusion

The modeling of a real-time simulation model from process of glycerol esterification with self-optimization was contributed to support the production system of GMO. The analysis of the model was described for lower level process detailed in BPMN 2.0 diagram. With those diagrams, we found the relationship and interaction between stakeholders and clearly used for the simulation to improve the performance system. Verification of the model ensured implementation of the conceptual model. As a result of zero error checking and the RTS run as well. The experiments showed that the evaluation of the proposed parameter such as process time to 2.88 hours and volume at 28L, that was increased about 16% for each batch and quality parameters that has closely related are density, viscosity and purity. Compared with the previous works set to 24L output only with all quality parameters were used.

In future work, it is recommended to accelerate the whole process including to re-structure the real-time data acquisition system. According this research, there is has some problems in monitoring the data to determine the status of the process in real time, for the future research it is interesting to applied the computational intelligence method such as classification to determine the status of process stages more clearly.

4 REAL TIME MONITORING GLYCEROL ESTERIFICATION PROCESS WITH MID-IR SENSORS USING SUPPORT VECTOR MACHINE CLASSIFICATION

Abstract

The commercial synthesis of fatty acid esters of glycerol has important aspect, as it plays role in other derivative production varieties. This research aims to construct the monitoring system for faster identification of esterification status and increase the efficiency of energy used for production used computational intelligence approach. The monitoring systems are based on the measurement parameters from two sources mid-IR 3.4 µm and 5.5 µm and two detectors that connected using the data acquisition system with ATmega8535 that connected to computer database via USB 2.0 and classifying the status of esterification using the Support Vector Machine (SVM) classifier. The purpose of SVM method is to classify the variations of parameter inputs from the mid-IR sensors in real time monitoring that connected with a microcontroller. In this research, three esterification statuses as initialization, work on process and finishing were divided for process monitoring in the bioreactor. The construction of classification based on SVM deployed in Orange software. In the application of esterification monitoring, the influence of various parameters such as temperature set in the reactor has relation to the process time needed. By monitoring this system statuses in every minute, we obtained one of the optimum process that was set in 210^{o}C is 2 hours.

Introduction

As explained in Chapter 2 of this dissertation, the commercial synthesis of fatty acid esters of glycerol was carried out by two different way namely direct esterification of fatty acid with the glycerol and catalysis by a homogenous acid (Isabel *et al.* 2003). In Indonesia, synthesis by direct esterification of fatty acid is widely used in the esterification process of glycerol because this process is simple and feasible in the batch production system (Pardi 2005). Several factors affecting the conversion efficiency of esterification process are a molar ratio of reactant, amount and type of catalyst, reaction temperature and stirring speed in reactor, and duration of the process (Mostafa and Maher 2013).

In the previous chapter has shown the major factors that should be controlled are reaction time, temperature and stirring speed, ideally supported by real time monitoring. The real-time monitoring also has a development in application as development by using microcontroller platform in low cost data collection and laboratory experiments which are designed and constructed using open source hardware and software (Barber *et al.* 2013; Ali *et al.* 2016) and also application in batch bioresource production as related to monitor the parameter of process (Lu *et al.* 2016).

A current method for determination of esterification product is sampling, which needs high cost and time. As in this chapter focused on determine the esterification status in real time, direct and close online monitoring of the product and critical components are highly desirable by monitoring the esterification process that related to control variable as the temperature and process time needed. Determination of the esterification reaction is highly desirable for esterified glycerol product in order to increase the efficiency energy and the cost of production. In this chapter, optical measurement techniques that are promising candidates in spectral region i.e. multi-channel NDIR (non-diffractive infrared) absorption or IR spectroscopy was used. The later method is also used in laboratories, thus allowing better correlation of online data and laboratory results with data acquisition interfaced by the database to the computer. SVM method used to find the correlation is used for calibration parameter in online measurement sensors, especially to identify the esterification status. The position of sensors from Sinelli *et al.* in 2004. demonstrated a set-up with detector array combined with the gradient filter to avoid the need for movable parts was inspired and applied. Wiesent *et al.* (2011) presented a system with an infrared source, a fluid cell consisting of two sapphire windows and a quadruple infrared detector equipped with different filter windows for analysis of phosphate ester.

As the objective of this chapter is to develop a system for the identification of esterification status using mid-IR sources, thermopile detectors, and data acquisition system using classification method, as well as to find the optimize variable related to temperature set in the reactor and the duration of the esterification process. In first section, we briefly explain the materials and methods. Next, we briefly described the calibration. Furthermore, explaination about the concept of classifying esterification status with SVM. Finally, we discussed the conclusion.

Materials and Methods

Materials

In this research, for the raw material and reaction we were used as in Chapter 2. Therefore, the internal mass transfer resistances were considered negligible in this study. The pseudo-homogeneous kinetic model was used to propose the reaction mechanism. The reaction rate equation was defined from Ilgen (2014) as

$$-r_A=k_1C_AC_B-k_{-1}C_CC_D \tag{5}$$

where C_A, C_B, C_C and C_D present the concentrations of oleic acid, glycerol, glycerol monooleate and water, respectively, and k_1 and k_{-1} are the forward and reverse reaction rate constants, respectively. Since the excessive presence of glycerol, the reaction was considered as reversible and the concentration of glycerol was considered as constant. The reaction rate equation was simplified to pseudo-homogeneous first order equation:

$$-r_C=\frac{dC_C}{d_t}=kC_C \tag{6}$$

where $k=k_1C_C$.

When C_A was expressed as a function of conversion (*X)* which *X*=[0,1] and taking natural logarithm, the following equation was obtained:

$$-\ln(1-X)=kt \tag{7}$$

The $-\ln(1-X)$ is related measurement of reaction conditions in conversion. The kinetic data were well fitted with the pseudo-homogeneous first order equation with high regression coefficients (R2 = 0.99). The pseudo-first order rate with respect to the oleic acid for esterification was also proposed by other researchers (e.g Dokic *et al.* 2013).

Equipment

Esterification reactions were carried out in a laboratory-built apparatus as explained in previous chapter. The esterification reaction was carried out under closed system, and temperature of the reactor was controlled using a heater plate (controlled by an internal thermostat) is shown in Figure 12 and monitored by using IR sensors (Soenandi and Djatna 2014) that integrate with microcontroller ATmega8535 and connected with serial to USB cable with prolific chipset to run data acquisition. All the reactants (oleic acids, pure glycerol, and catalyst) were pondered and poured into the reactor. Then, the temperature was increased by adjusting the thermostat. The magnetic stirrer was allowed to operate after 5-10 min (to heat up the mixture). After passing the targeted reaction time, the reactor was removed from the hot plate. The reaction mixture was cooled down to the ambient temperature by immersing it into the water bath. Samples were withdrawn from the reaction mixture for further analysis.

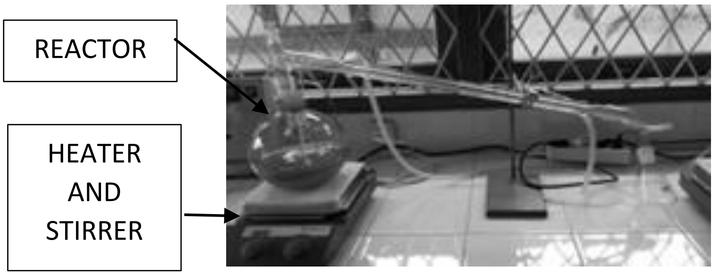


Figure 12 Part of apparatus set in laboratory experiment

Variation in Temperature and Process Time

We run several reactions test and varied condition, which were tested in the laboratory of Surfactant Bioenergy Research Center (SBRC) at IPB Baranangsiang Bogor. In order to get varied conditions of the temperature and reaction time. we monitor the proces in temperature 120^{o}C, 180 ^{o}C and 210^{o}C. On the other hand, the process time was measured in 40, 78 and 120 minutes out of environmental room temperature consideration.

Sensor System

In order to support the monitoring process, a commercial infrared mid-IR was set as source as the original growth of narrow gap semiconductor alloys onto n+-InAs substrate, optical coupling through the use of chalcogenide glasses and Si lenses with an antireflection coating (Boston Electronic 2014).

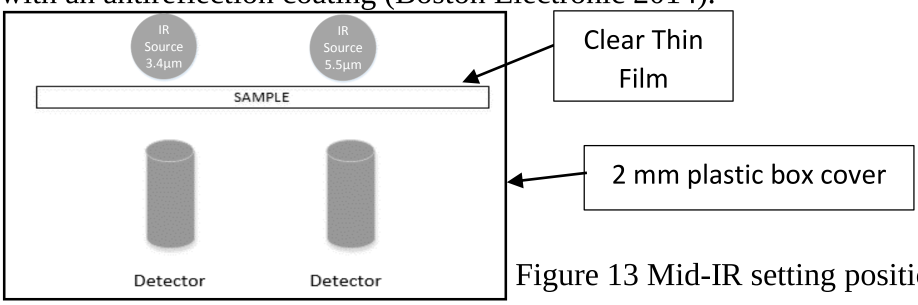


Figure 13 Mid-IR setting position

Three types mid-IR sensors of 3.4 micron LED-34SR full thread body, 5.5 micron LED-55SR full thread body and 7 micron OPLED 70 full thread body, also the thermopile detectors from Heimann HTIA Dx-Tx used for detecting the sample as reach steady state and to record signal amplitude with a good signal-to-noise ratio and position layout of the sensor and detector that covered with 2 mm plastic box to isulate the detector from environment rays that cause noises is in Figure 13. The sensor has Cockcroft–Walton (CW) where as a voltage multiplier converts AC or pulsing DC electrical power from lower voltage level to a higher DC voltage level.
To measure the process in the reactor, we used sensor 3.4μm that has specification as peak wavelength, pulse power and Cockcroft–Walton (CW) voltage in Table 4

Table 4 Specification of mid-IR LED34Sr

Properties	Unit	Value
Peak wavelength	μm	3,4±0.05
Pulse Power	mW	0.25-0.35
CW Voltage	V	Drive Current 0.2A 0.26-0.29

And for sensor 5.5μm and 7 μm specification as peak wavelength, pulse power and drive current of Cockcroft–Walton (CW) voltage in Table 5 until Table 6

Table 5 Specification of mid-IR LED55Sr

Properties	Unit	Value
Peak wavelength	μm	5,4-5.5
Pulse Power	μW	5-7
CW Voltage	V	Drive Current 0.2A 1.5÷2.5

Table 6 Specification of mid-IR OPLED70

Properties	Unit	Value
Peak wavelength	μm	6,5-7.0
Pulse Power	μW	5-7
CW Voltage	V	Drive Current 0.2A 1.5-2.5

Data Acquisition

To ensure the data parameters get collected from the sensors, the system was interfaced with a database such as MySQL.

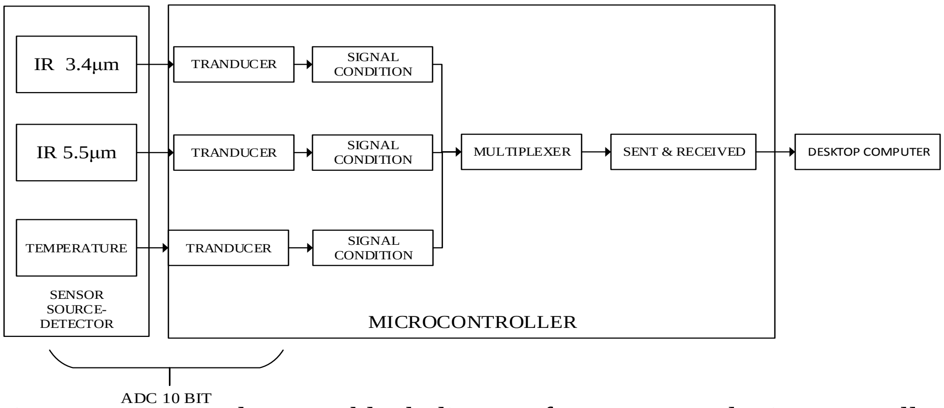


Figure 14 Proposed system block diagram for sensors and microcontroller

As shown in Figure 14, we built the data acquisition system by using three type of sensors as 3.4µm, 5.5 µm and temperature connected to microcontroller ATmega8535 with 10 bit Analog to Digital Converter (ADC) for convert analog signal to digital. Next, attached with Universal Serial Bus (USB) 2.0 prolific to connect the data streaming in real time to the desktop computer. Next, the calibration of the sensors was executed and the output was processed with SVM method in a personal computer (Kwok 1999).

Calibration

To ensure the precision esterification measurement, this system calibrated by 3 step method: testing with the blank sample, full closed sample (using a sheet of paper) and compared the spectrum FTIR of esterified glycerol with parameter 210^{o}C and 120 minutes process as in Appendix 11 FTIR test comparison for non esterified and esterified .

Table 7 Blank sample for sensors calibration

Time (Second)	Digital Output Number		Transmittance Calibration	
	5.5µm	3.4µm	5.5µm	3.4µm
1	205	258	100	100
2	209	260	100	100
3	208	258	100	100
4	210	257	100	100
5	207	258	100	100
6	208	256	100	100
7	209	255	100	100
8	207	259	100	100
9	208	257	100	100
10	206	258	100	100
Average	208	258	100	100

Table 8 Full closed sample for sensors calibration

Time (Second)	Digital Output Number		Transmittance Calibration	
	5.5µm	3.4µm	5.5µm	3.4µm
1	178	203	0	0
2	178	203	0	0
3	182	201	0	0
4	181	202	0	0
5	182	203	0	0
6	182	201	0	0
7	182	201	0	0
8	181	202	0	0
9	178	201	0	0
10	179	201	0	0
Average	180	202	0	0

After we get the average value for the two condition (blank sample and closed sample/paper sheet) to describe transmittance upper and lower limit value in 10 seconds from the sensor as shown in Table 7 and Table 8. We collected the transmittance in 10 seconds, because in 10 seconds we assumed that all the fluctuations has well measured, for the next step to find the interpolation for this identification system we used formula as in Equation 8.

$$\text{Transmittance (\%)} = \frac{x_n - \frac{\sum x_{ic}}{n_{ic}}}{\frac{\sum x_{ib}}{n_{ib}} - \frac{\sum x_{ic}}{n_{ic}}} \times 100 \qquad (8)$$

where:

x_n = bit number of digital output;

$\frac{\sum x_{ic}}{n_{ic}}$ = average bit number used in closed sample;

$\frac{\sum x_{ib}}{n_{ib}}$ = average bit number used in blank sample.

Sensor Selection

In this work, to analyze the sensors that closely related to esterification forming parameter, we used feature selection Reliable Elimination of Features (Relief) algorithm as explained in detail in Chapter 3 page 13. We have collected

30 datasets and measured with sensors to get three classification as represented of three groups of esterification status

Support Vector Machine (SVM)

Support Vector Machine (SVM) was first heard in 1992, introduced by Boser, Guyon, and Vapnik. Support vector machines (SVMs) are a set of related supervised learning methods used for classification and regression (Vapnik *et al*. 1997). SVM is a useful technique for data classification and supported of real-time data processing. For this type of SVM (Vapnik *et al*. 1997), several formulations of training involves the minimization of the error function as shown in Equation 9

$$\frac{1}{2}\omega^T\omega + C\sum_{i=1}^{N}\zeta_i \tag{9}$$

subject to the constraints as:

$$y_i(\omega^T\phi(x_i) + b) \geq 1 - \zeta_i \tag{10}$$

$$\text{and } \zeta_i \geq 0, i = 1,\ldots\ldots, N \tag{11}$$

where C is the capacity constant, ω is the vector of coefficients, b is a constant, and ζ_i in Equation 11 represents parameters for handling non-separable data. Then, the index i labels the N training cases. Note that $y \in \pm 1$ represents the class labels and x={x_i | i=1,…,n} x_i represent the input of IR sensors used for n is time length in measurement space. For the process to find cluster the kernel ϕ is used to transform data from the input (independent) to the feature space. It should be noted that the larger the C, the more the error is penalized. Thus, C should be chosen with care to avoid over-fitting for the cluster. A classification task like esterification status process usually involves with training and testing data, which consist of some data instances. Each instance in the training set contains one target values and several attributes, which tested in the laboratory using sample process in each, attribute. As an output, the goal of SVM is to produce a model, which predicts target value of data instances in the testing set which is given only the attributes (Cristianini and Taylor 2000), in this research, the output of target value is esterification status.

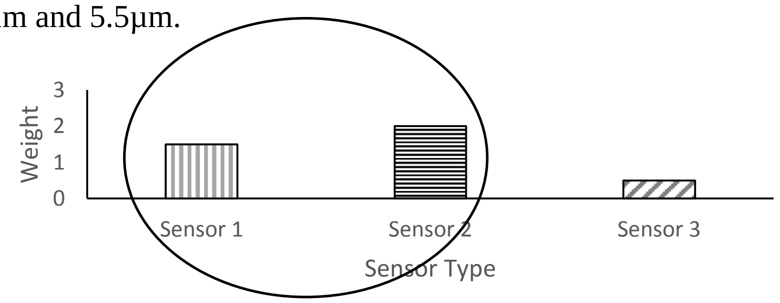
ROC Performance Classifier

A receiver operating characteristic (ROC), or ROC curve, is a graphical plot that illustrates the performance of a binary classifier system as its discrimination threshold is varied. The curve is created by plotting the true positive rate (TPR) against the false positive rate (FPR) at various threshold settings. The true-positive rate is also known as sensitivity, or recall in machine learning. The false-positive rate is also known as the fall-out and can be calculated as (1 - specificity) (Hernandez and Orallo 2013). In test learner evaluation there are three parameters such as Classification Accuracy(CA), Sensitivity (Sens) and Specificity (Spec). CA is a percentage of a number of correct predictions from all prediction made. Sens is measuring the proportion of positives that are correctly identified and Spec has measured the proportion of negatives that are correctly identified. Sensitivity, specificity, and accuracy are described in terms of TP, TN, FN and FP.Sensitivity = TP/(TP + FN) = (Number of true positive assessment) /(Number of all positive assessment), Specificity = TN/(TN + FP) = (Number of true negative assessment)/(Number of all negative assessment), Accuracy = (TN + TP)/(TN+TP+FN+FP) = (Number of correct assessments) /Number of all assessments). Finally, ROC curve is a graphic presentation of the relationship between both sensitivity and specificity and it helps to decide the optimal model through determining the best threshold for the diagnostic test.

Experimental Results

Feature Selection for Sensor

As the result of attribute weighted in Relief method with 30 datasets (Appendix 5) that classfied by sensors measurement and FTIR test as shown in Figure 15, the sensors that has closely related with measurement of esterification are 3.4µm and 5.5µm.



Note: Sensor 1(x_1): 3.4µm, Sensor 2(x_2): 5.5µm and Sensor 3(x_3): 7µm

Figure 15 Attribute weight of sensors from Relief method

During laboratory works, we collected experimental process using data acquisition with temperature between 120^{o}C to 210^{o}C in 160 minutes process time. The plot of data acquisition in graphical was described in Figure 16. With this data acquisition system, we can get real time data plotted in interval of three seconds during 160 minutes and classified with SVM method.

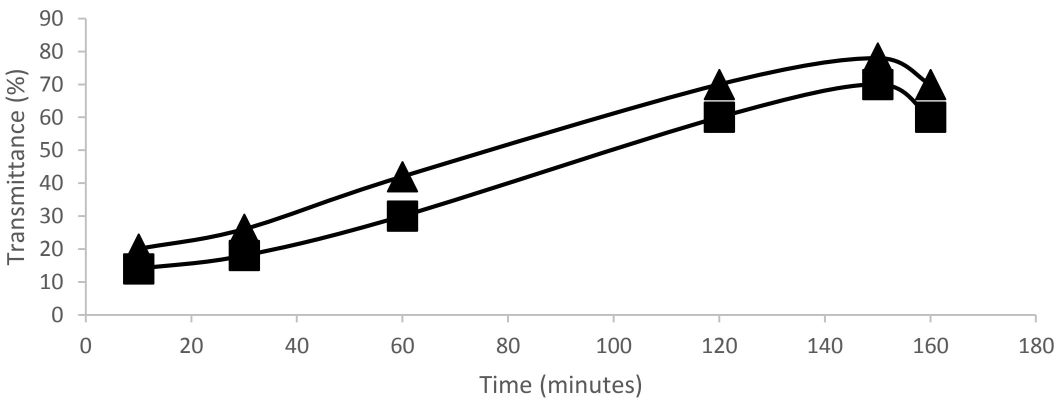


Figure 16 Real-time transmittance measurement from 3.4µm(■) and 5.5µm(▲) sensors

In our research, we used SVM method to classify the input parameter identified by the sensor as a value of transmittance parameter and the accuracy of this model was tested by using ROC analysis.

Esterification Classification with SVM

For the data acquisition in this research, we used online data measurement interfaced with SQL database and identified the parameter inputs by using optical sensors mid-IR that attached in the bioreactor. The input database was collected and clustered into esterification status in real-time with SVM method. Next, with computer data mining software application Orange version 2.7 (Biolab 2015) to deploy a knowledge flow model for the classification step as in Figure 17.

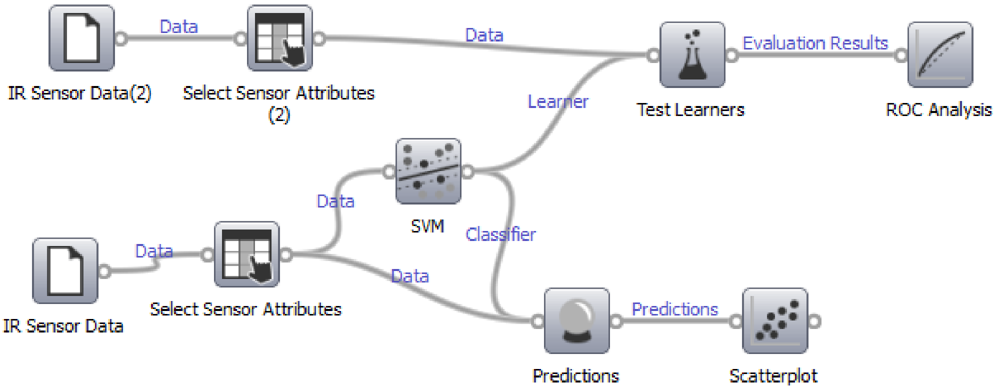


Figure 17 A knowledge flow scheme of SVM method

In this block of a knowledge flow was started from data selection for attributes from mid-IR. Next, the block of SVM method was trained with 100 data and tested by processing the data file with 150 samples, 3 attributes (sensor 3.4µm, sensor 5.5µm and temperature) and Classification Discrete-Class with 3 values (initialization, work on process and finishing). This flow run in C-SVM for C=1.00 and Sigmoid Kernel typed was used.

ROC Analysis

In case of measurement of SVM classifier, we used test learners block on 5 number folds sampling Cross-validation. The evaluation result was displayed with Classification Accuracy rate of 95,58%, sensitivity level at 97.81% and specificity rate of 95.54%. Another test method this classification was tested by ROC curve with a graphical plot on each predicted class. As in this research there are three esterification status as initialization (predicted class 1), work on process (predicted class 2), and finishing (predicted class 3). These graphs plotted between FP (False Positive) rate and TP (True Positive) rate that illustrate the performance of a classifier system as its discrimination threshold is varied. The curve generated by Orange Software data mining (Biolab 2015) which was shown by plotting the true positive rate against the false positive rate at various threshold settings. The closer the points on the ROC curve to the diagonal, the less accurate the test is. As a some example result of classification that shown in Table 9 and more complete data listed in Appendix 7 for data record using two sensors in real time.

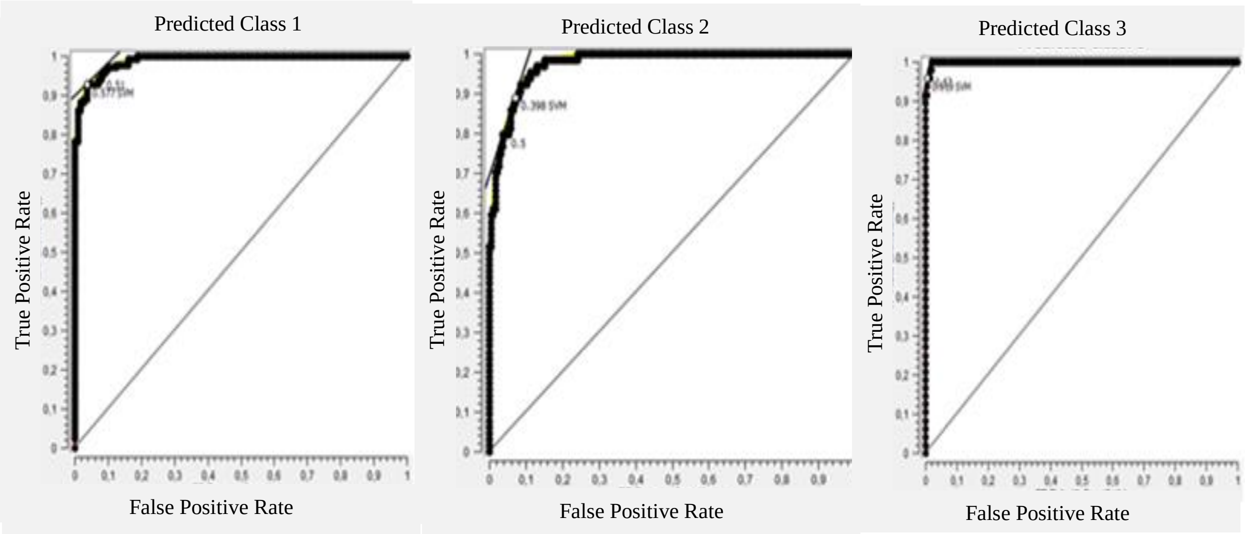


Figure 18 Classification curves of ROC analysis

From the three predicted classes in Figure 18, we found that the classifying performance of SVM was excellent as the interpretation of ROC curves of 3 predicted class is similar to a single point in the ROC space, the closer the point on the ROC curve to the ideal coordinate in upper left, the more accurate the test is

Table 9 Example of identification by sensors in transmittance measurement with SVM classifications

Sensor 1(X_1) (Transmitance)	Sensor 2(X_2) (Transmitance)	Time (minutes)	Temperature (^oC)	Esterification Status from Classification With SVM(ξ_i)
20	34	40	120	Initialization
46	50	78	180	Work On Process
75	84	120	210	Finishing

In Table 9 we have example of result that in measurement from Sensor 1 (3.4µm) has measured the transmittance level of 20 and Sensor 2 (5.5µm) has measured the transmittance level of 34 in process time 40 minutes and temperature was 120 ^oC, by SVM this set of data sample was classified in esterification status of initialization. Next, we have another example of result that in measurement from Sensor 1 (3.4µm) has measured the transmittance level of 46 and Sensor 2 (5.5µm) has measured the transmittance level of 50 in process time 78 minutes and temperature was 180^oC, by SVM this set of data sample was classified in esterification status of work on process. And finally for the last set of data was classified in esterification status of finishing.

In this result has revealed that by classification of the esterification we can monitor the esterification time more precisely and yields of esterification also increased up to a higher conversion.

Discussion

In this chapter the real-time monitoring of esterification process with sensors was supported with classification method is presented to monitor the phase of esterification process because to identify the esterification process is needed. It is important to develop a system for the identification of esterification status using LED mid-IR sources, thermopile detectors, and data acquisition, as well as to find the variable condition related to temperature in the reactor and the duration of the esterification process in real time condition.

In comparison to the previous research from Sinelli *et al*. (2004) that we also set a sensor position in paralel. As in step for real time monitoring was started from the output value from sensors in analog signal that converted in data acquisition system to defined the state of esterification by classification. In comparison from Lu *et al.* (2016) that not using classification for monitoring, in this chapter the real time monitoring was used a classification method after the database was built, and this classification method was deployed in 3 states to simplify the identification of esterification stages. In comparison to Cristianini (2000) that have ratio 1:10 for calibration and data sets, we used 10 sample data for calibration and 300 data training sets.

We obtained the result as shown in Figure 18 for the ROC curved with 3 predicted class using C=1.00 was good in classification because the best possible prediction method would have yield a point in the upper left corner or coordinate (0,1) of the ROC space, representing 100% sensitivity (no false negatives) and 100% specificity (no false positives) and as shown the points are near the upper left corner. The parameter as temperature and time in the process had major effects on the conversion of the esterification. The obtained results showed that by increasing the reaction temperature, the reaction conversion also increased rapidly and after 2 hours, the esterification reached a well-formed esterification status.

Therefore, to test the classification, several various of esterification reaction that was carried out within a temperature range between 120°C to 210°C (maximum heater temperature) was used. The results revealed that by increasing the esterification time, the esterification yields also increased up to a maximum conversion. Finally, to determine the time needed for esterification process exactly, besides considering the maximum yields of esterification, it was also necessary to take the time required to reach the reaction temperature into account as shown in Table 9. The length for heating time to reach the reaction temperature was certainly longer and the energy consumption was surely greater for higher reaction temperature. Consequently, a faster reaction at a set temperature was desirable.

In the next chapter, we proposed a new development to increase the performance of optimization that applied in control system with an adaptive capability to accomodate the various disturbance in esterification process.

Conclusion

The system of real-time monitoring glycerol esterification process with mid-IR sensors classifying with SVM was contributed to support the identification esterification status in every minute and to get information for the time needed for the esterification process. This esterification status achieved a good performance when classifying into 3 states as initialization, work on process and finishing. This classification was trained and tested in Orange Software for data mining using SVM method whereas the performance of the classifier was tested using ROC analysis. In applied for esterification optimization, the influence of variable condition, such as the temperature set in the reactor, had a relation to the process time needed. By using this monitoring system based on the measurement and classification of esterification forming using SVM from two inputs of mid-IR 3.4 and 5.5 µm sensors, we obtained the optimum process condition was set in 210^{o}C and the time needed for the process was 120 minutes. By analyzing data collection which build from esterification process, a precise measurement and classifying still needs some improvements to cope the dynamic condition in the process. For future development an adaptive method to determine the status was needed to be implemented in control system and find the optimum number of esterification status and control state.

5 OPTIMIZATION MODEL OF GLYCEROL ESTERIFICATION PROCESS USING REAL-TIME ADAPTIVE CONTROL

Abstract

The synthesis reaction used in esterification needs high energy consumption and a precise processing time to get the best yield of the target. Based on the problem a model was formulated to optimize glycerol esterification process by minimizing the time needed for the process and maximizing the yield of Monoglycerides. This optimization has gained importance for boosting the esterification industry and improving the production efficiency. Optimization through adaptive monitoring and control has provided significant advances in the process efficiency, a lower energy consumption, and a better product quality. This paper presents the optimization with a computational intelligence in real-time and adaptive control (RTAC), as compared to the conventional (non-controlled) methods to monitor and control of glycerol esterification processes. The identification of esterification status based on temperature and time are evaluated to strengthen the optimization. An adaptive method as feature selection to select wavelength mid-IR sensors at specified intervals was carried out with Relief algorithm and Adaptive Pillar k-means clustering method to set the parameter control was proposed in this paper. Many combinations were evaluated from real time condition process, to achieve the best optimization results. The experimental results demonstrate that real-time adaptive control can be achieved by using three clusters, which are heating up, stabilizing and finishing. In RTAC, each cluster has its own parameter to set the control point by the servo motor that was attached at magnetic stirrer-heater. By using optimization parameter for each cluster, esterification process time can be shortened up to 20 minutes with a higher yield 10% or more, lower range stirrer rotation between 300 rpm to 450 rpm and a lower final temperature between 200^0C to 210^0C.

Introduction

Currently, monoglycerides and diglycerides are important substances in processes where involving emulsification occur. As in previous chapter, one of the key challenges in the process industry is to find the best operation method for the plant under different conditions such as feed compositions, production rates, energy availability, feed and product compositions that change dynamically like in esterification process (Isabel *et al.* 2003). In industrial chemical process synthesis by direct esterification is widely used to get mono-glycerides and di-glycerides because this process is simple and feasible in the batch production system (Cramer *et al.* 2007).

The task for optimizing process especially in esterification is usually tackled using a supervisory control technique, monitored manually to check the parameter of the process, this technique needs attention and still has loss of efficiency. One such technique that has received considerable attention in the process industry is the Real-Time Optimization (RTO) (Adetola *et al.* 2009). Real-Time Optimization

(RTO), which refers to the online economic optimization of a process plant, is a widely employed technology to meet this challenge.

The limitation of RTO is that it is not adaptive to the type of the raw material (feed composition) that affected reaction process of esterification. In this paper, we address this issue by combining RTO with an Adaptive Controller (AC) for glycerol esterification reaction consisting a real-time identifier and a minimum variance regulator for the identification, done by augmenting the state with the unknown parameters of the process was previously proposed by Wieslander and Wittenmark in 1971.

The main problems in adaptive control are to identify the present system parameters and to choose the appropriate control strategy. Early application of adaptive control was presented by Astrom in 1989 and have already used in the control of a permanent synchronous motor for digital adaptive velocity to maintain invariant velocity control over the motor in the presence of varying mechanical parameters.

There are several chemical processes and reaction to produce mono glycerides. We focused on the chemical reaction process in selective synthesis of monoglycerides by esterification glycerol with fatty acids (Soenandi and Djatna 2014), which has a complex reaction because of the immiscibility of reagents and the formation of glycerol dioleate and glycerol trioleate as composition of products. This reaction of the process esterification has characteristics that require time and energy for heating to make the esterification process occur, this process will be optimized by reducing the process time (minimization) and maximizing the yield of glycerol monooleate by monitoring and controlling (Soenandi *et al.* 2015).

Process monitoring is the manipulation of sensor measurements (e.g. force, vision, temperature, the rate of transmittance) needed for determining the state or condition of the processes. Automatic monitoring algorithms utilize selected sensor measurements that, along with inputs, determine the process state. The states of complex processes are monitored by a sophisticated signal processing of sensor measurements. Process control is the manipulation of process variables to regulate the processes (Chryssolouris *et al*. 1992). In traditional methods, the operators perform on-line and off-line process control by adjusting the temperature button and always giving attention to the rotational speeds of stirrer reactor to suppress over-temperature, and watching the time needed for the process with a traditional stopwatch.

In this chapter we defined the objectives are to select the wavelength sensors that are related to esterification status, to find the optimal number of data cluster for parameter control and to evaluate the system performance using a new proposed method. As a method, we describe Real-Time Adaptive Control (RTAC) as used to optimize the number of cluster from real-time data streaming using sensors, during the process of esterification in a bioreactor and clustered it to determine the set point parameter as decision variables of temperature and the rotational speed of stirrer. To achieve those tasks simultaneously, RTAC system is supported by real-time optimization methods using Relief (Reliable Elimination of Features) from Kira and Rendell (1992) as already disscussed in Chapter 2 page 10 and then Pillar k-means algorithm was deployed based on by Barabakh and Helen (2005) work, combined with real-time data acquisition from optical mid-IR sensors to set the best parameter control for the esterification process.

To optimize esterification process, the main challenge is how to determine the time needed for the process to get the best yield for the product. By using sensors to identify the composition of product mixed in mono-glycerides and di-glycerides. We state our optimization problems using parameters of temperature in and rotational stirrer speed to optimize process time and yield of product. To simplify the problem, we assumed the purity of reactant, ratio of charged reactant, catalyst concentrations as constant variables.

In RTAC, there are many tasks to do, initially byrom preparing the sensors and data acquisition system and processing real-time data, finding the best parameter for the process that is being affected by the condition of the external disturbance like reactor's surrounding temperature, the impurity of raw material and instability of the electric current. On the other side, noise problem from the input sensor is also crucial. Calibration is also important because of the variability condition from the bioreactor and the sensitivity of the sensors used for setting parameter of the controller.

The scope of high-performance computing is rapidly expanding from single parallel systems to get clusters of heterogeneous sequential and parallel systems. Moreover, as applications become more complex, more irregular, with a data-dependent execution behavior, and more dynamic, with time-varying resource demands.

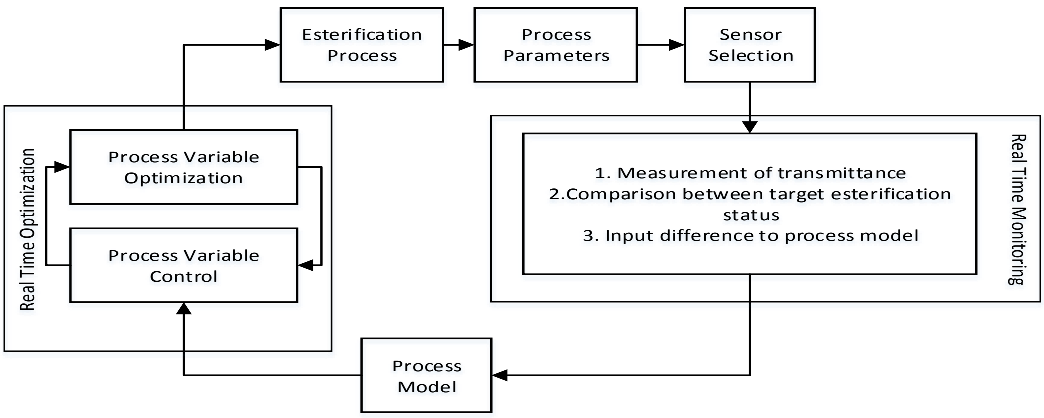


Figure 19 Monitoring and optimization in real time concept

In this chapter, we want to solve those optimization problems with an algorithm to adapt the changes in esterification process by clustering. In Figure 19 was shown the Real-time optimization is the process of extracting interesting and previously unknown or unexpected knowledge from a large amount of process variable optimization and process variable control with support of real time monitoring such as measurement of transmittance, comparison between esterification status and input difference to obtain process variable optimization and control that developed in process model. Its correctness depends on not only to logical correctness but also time constraints. Various methods in the area of data mining have been developed in order to effectively and correctly discover knowledge.

Materials and Methods

First, we formulated the optimization of esterification glycerol with oleic acid as objective functions are to minimize process time and maximize the yield of monoglycerides as target of the product. In mathematical formulations, they are expressed in Equation 12 until Equation 17. The boundaries were set in capability range of existing reactor operation. The Real-Time Adaptive Control problem to solve can be stated as:

Objective functions: $\min_{\boldsymbol{T},\boldsymbol{R}} t(\mathbf{T}, \mathbf{C}, \mathbf{R}, S)$ (12)

$\max_{\boldsymbol{T},\boldsymbol{R}} y(\mathbf{T}, \mathbf{C}, \mathbf{R}, S)$ (13)

with decision variables : $\mathbf{T}$ = temperature (°C),

$\mathbf{C}$ = target of composition (%),

$\mathbf{R}$ = rotational stirrer (rpm), (14)

subject to : $\mathbf{C}=f_p(\mathbf{T})$

$g(\mathbf{T},\mathbf{C},\mathbf{R},S) \leq 0$ (15)

boundaries : $120\ ^0\mathrm{C} \leq \mathbf{T} \leq 230\ ^0\mathrm{C}$

$200 \leq \mathbf{R} \leq 500$ rpm

$S=\{1,2,...,n\}$ (17)

where t is the process time needed, y is the yield of esterification (monoglycerides), f_p is the equations set that represents the real process behavior, S is the cluster set from output and g is the process constraints set.

This section describes the procedure and step from monitoring the reaction in reactor to get the solution for optimization in real time esterification process by controlled the parameter using the cluster. The components of this process detailed in this section are material and equipment, mid-IR sensors, dataset, feature selection, and clustering algorithm.

For the esterification process as described in Chapter 1 in Introduction on page 1 were referred and all the reactants has already discussed in Chapter 3 page 18. In order to get varied conditions of the temperature and reaction time the temperature was controlled at 180-230°C and the process time was varied between 100-130 minutes.

Selected mid-IR sensors were deployed in the real-time data acquisition system to identify the esterification condition process. Infrared energy is emitted or absorbed by molecules when they change their rotational-vibrational movements. Infrared energy excites vibrational modes in a molecule through a change in the dipole moment, making it a useful frequency/wavelength range for the study of these energy states for molecules of the proper symmetry. Infrared spectroscopy examines absorption and transmission of photons in the infrared energy range (Edelman *et al.* 2001). The LED mid-IR sensors with these center wavelengths: 3.4 µm, 5.5 µm and 7.0 µm were used in this chapter. Each comes with the optically immersed specification. LEDs fabricated from III-V hetero structures grown onto Indium Arsenide (InAs) substrates type from Boston Electronics (Boston 2014) were used in this chapter for the identification of esterification condition in the process. These sensors were connected to analog inputs using a microcontroller from Arduino (Ivrea 2005) for streaming data in real time using a USB 2.0 serial port with BAUD rate of 9600bps.

The samples was identified used a sensor as described in Chapter 2 page 6 with connected to Arduino Uno version 2.0 as shown in Appendix 2 and the real-time data were collected and plotted in a graph with spreadsheet Microsoft Excel 2013 (Figure 20) and interfaced with pyserial for computational data mining with Python Integrated Development Environment (PIDE). As long as the esterification process, real-time data streaming with duration of 160 minutes, step in minutes incremental recorded to the database. The input data were the percentage transmittance level of IR rays from wavelengths of 3.4, 5.5 and 7 µm, temperature in the reactor, rotation speed of stirrer in reactor and time of esterification process, while the output data were the esterification process.

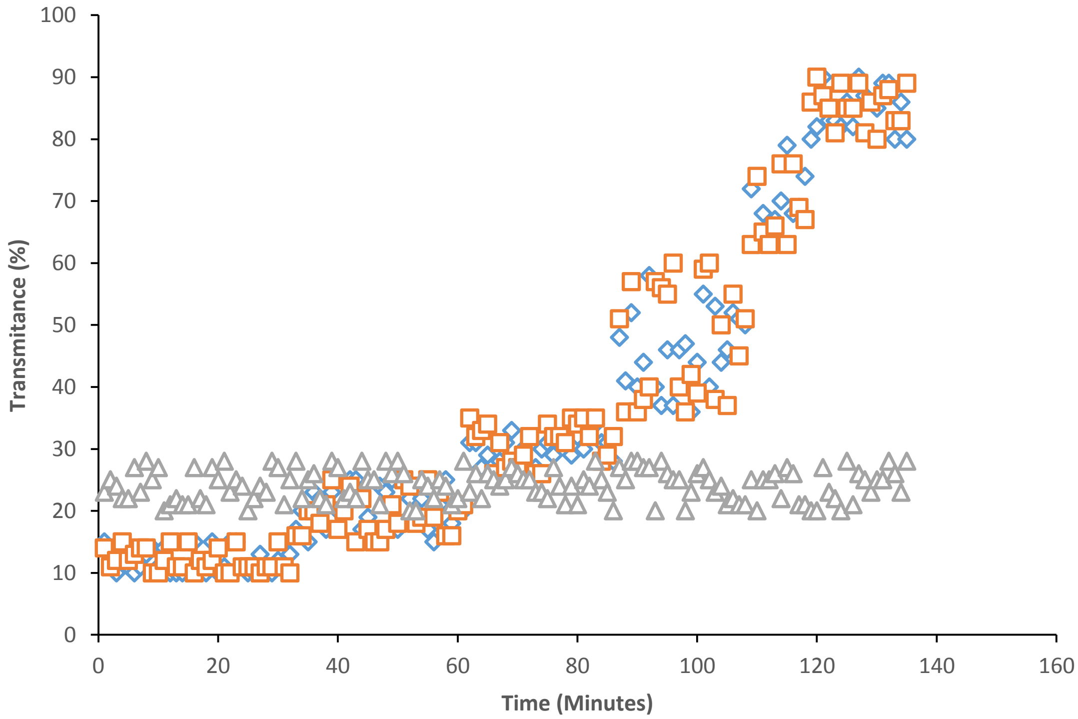


Figure 20 Real-time data from mid-IR sensor as 3.4µm(◊), 5.4 µm(□) and 7µm(Δ)

In Figure 20 we have plotted the real time data measurement from three types of mid-IR sensors that has different minimum and maximum value of measurement for each distribution plot of sensors, this differences makes the measurement of yield was less precise. According this plot, that 7µm sensor has the smallest span of distribution.

Sensor Selection Related to Esterification Status

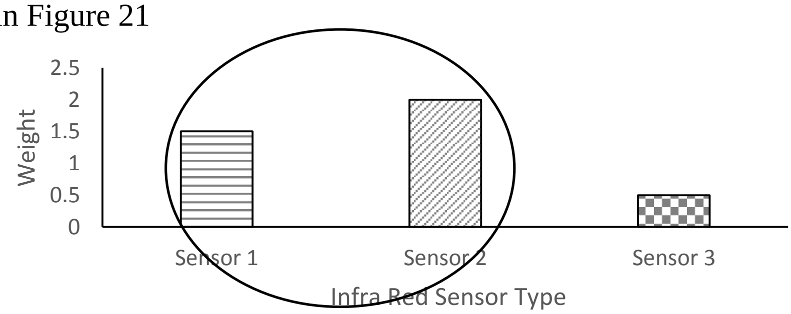
In this work, to select the sensors that are related to esterification status, we used the Relief algorithm which can deal with multiclass problems. The improved algorithm is more robust and also able to deal with incomplete and noisy data developed by Kira and Rendell (1992) as already discussed in Chapter 3 page 18. For validation purposes we used the algorithm in three-class that changed esterification status as in Table 10 from an assay in the laboratory as FTIR (Fourier Transform Infra-Red) and GCMS (Gas Chromatography–Mass Spectrometry) with process time between 90 until 120 minutes. By experiment in laboratory we have collected 8 datasets from the processing glycerol esterification with a range of temperature from 120^0C to 230^0C and tested the esterification status in a certified laboratory as in Table 10. By comparing the value between them, we chose sensor 1 (3.4 µm), sensor 2 (5.5 µm) and eliminated sensor 3 (7.0 µm) because it has the lowest weight value as shown in Figure 21. This means that sensor 3 was not suitable for identification of esterification status.

Table 10 Example of esterification status dataset

Transmittance Detection (%)			Esterification
Sensor 1 (x_1)	Sensor 2 (x_2)	Sensor 3 (x_3)	Status
11	13	20	A
12	15	24	A
11	13	21	A
20	21	24	B
35	40	30	B
48	45	32	B
65	60	30	C
73	70	35	C

A is status of initialization; B is status of work on process; C is finishing

As the result of in Relief method using data partially shown in Table 10 and 30 datasets (Appendix 6) completely, that classfied by attribute of sensors measurement, process time and defined the class of esterification status from FTIR as shown in Figure 21



Note: Sensor 1(x_1): 5.5µm, Sensor 2(x_2): 3.4 µm and Sensor 3(x_3): 7µm

Figure 21 Weight average from Relief method of each sensor type in 90-120 minutes period

Pillar K-means Algorithm

In case of to cluster the measurement data from sensor, pillar algorithm to support the real time aspect of this research was used, as an improvement from k-means algorithm. First, the input for this algorithm was get from real-time data acquisition from sensors that selected with Relief as an attribute as x={x_i | i=1,…,n} as in this research we used n=2 and randomly set the pillars to find initial centroids c={c_i | i=1,…,k}. Next, in processing the algorithm considers that pillars placement should be located as far as possible from each other to obtain the centroid. After pillar was located, then it uses DM for storing the accumulated distance metric and D for storing the distance metric for each iteration, where DM={dm_i | i=1,…,n} and D={d_i | i=1,…,n}. To analyze the closeness of the final centroids of the clustering result to the centroids of the real-time data sets from Barakbah and Helen (2005) it can be defined as:

$$\min \sum_{i=1}^{k} (\|c_i - r_i\| \tag{18}$$

where c_i is i_{th} final centroid of clustering result and r_i is i_{th} real centroid of data set.

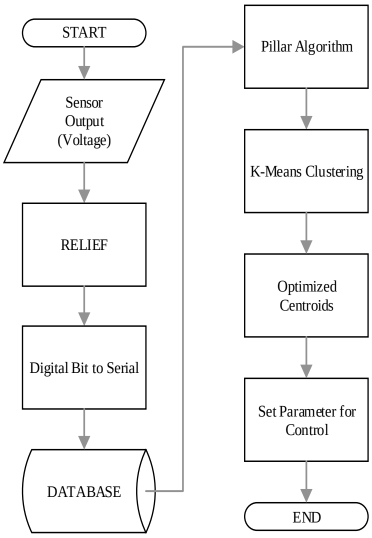


Figure 22 Flowchart of initial centroid optimization of Pillar k-means method

To get the right number of data cluster from two sensors input from defined centroid, this chapter we applied the k-means clustering for grouping by Pillar algorithm after we implemented Relief method as a flowchart in Figure 22 and measured of the Silhouette value. The Pillar algorithm is very robust and superior for initial centroids optimization for k-means by positioning all centroids far separately among them in the data distribution (Barakbah and Kiyoki 2010). The thought process of determining a set of pillars locations in order to make a stable house or building inspires this algorithm. By distributing the pillars as far as possible from each other within the pressure distribution of a roof, the pillars can withstand the roof’s pressure and to stabilize a house or building. It considers the pillars, which should be located as far as possible from each other to withstand the pressure distribution of a roof, a number of centroids among the gravity weight of data distribution in the vector space.

Therefore, this algorithm steps conducted for data sensor segmentation using Pillar k-means clustering are read the dataset as a matrix and after the matrix was built this algorithm was calculated the data size in means arrayed. Next, find a number of clusters with decided initial furthest centroid of the cluster using the Pillar k-means algorithm and create a group of data. Finally, we get control output from data segmentation.

Silhouette score

In this research, to determine the optimal number of the cluster from sensors, we used the Silhouette score that refers to a method of interpretation and validation of consistency within clusters of data.

Silhouette refers to a method of interpretation and validation of consistency within clusters of data. The technique provides a simple graphical representation of how well each object lies within its cluster. The technique provides a succinct graphical representation of how well each object lies within its cluster (Rousseeuw 1987). The silhouette value is a measure of how similar an object is to its own cluster (cohesion) compared to other clusters (separation). The silhouette ranges from -1 to 1, where a high value indicates that the object is well matched to its own cluster and poorly matched to neighboring clusters. If most objects have a high value, then the clustering configuration is appropriate The Silhouette method also relates compactness to separation, it is based on the mean score for every point in the dataset. This difference is then divided by a normalizing term, which is the greater of the two averages, as the formulation from Rousseeuw (1987) for clustering as in Equation 19 and Silhouette score as in Equation 20 are

$$\max\left(\begin{matrix} Arg \\ b(i)-a(i) \end{matrix} \frac{1}{N}\sum_{i=0}^{N} d\big(i, m(i)\big)\right) \tag{19}$$

$$S_i = \frac{b(i)-a(i)}{\max(a(i), b(i))} \tag{20}$$

where N is the number of points in the data set, d(i,m(i)) is the dissimilarity of object i to the nearest representative object, denoted by m(i), a(i) is the average dissimilarity of i with all other data within the same cluster, b(i) is the lowest average dissimilarity of i to any other cluster where S_i is a Silhouette score of i^{th} cluster.

Integration of Sensor Device And Algorithms

Integration of sensor devices and the algorithms (Relief and Pillar k-means) was intended to measure the transmittance level from sensor as an variable input which is related to esterification process precisely to set the parameter for control.

The data was read and collected by the computer using USB 2.0 with a 1-minute interval time sampling to build the database. The esterification process needs a long time to complete (approximately 120-180 minutes) to produce high yield and good quality of product. By using a computational intelligence we still need time to process the data before we can get the information needed especially in dynamic reactions.

In order to solve this problem, we used the data sampling size of 1 minute, and then we applied the Pillar algorithm, these architecture computational models were designed on a computer with CPU processor Intel Celeron 1007U and a memory of 2GB with Software Python version 2.7.10 (Python 2015). Based on the data transmittance read by the sensor, we then used Relief method in spreadsheet Microsoft Excel 2013 with Macro programming to select the sensor that is suitable for the esterification, and sent the signal that was detected by the sensor selected to be clustered with the Pillar k-means in Python 2.7.10.

After clustering the data, the parameters of the process, which are temperature and rotational speed of the stirrer, were used to set the position levels of the PID control in Arduino Uno Microcontroller (Margolis 2011). We used a proportional-integral-derivative controller (PID controller) because it has a control loop feedback mechanism (controller) widely used in industrial control systems. PID control is often combined with logic, sequential functions, selectors, and simple function blocks to build complicated automation systems used for energy production, transportation, and manufacturing. In this research, a PID controller calculates an error value as the difference between a measured process variable and the desired set point and used the values to adjust the level or position of rotary resistor switch at the magnetic stirrer and heater, using a servo motor.

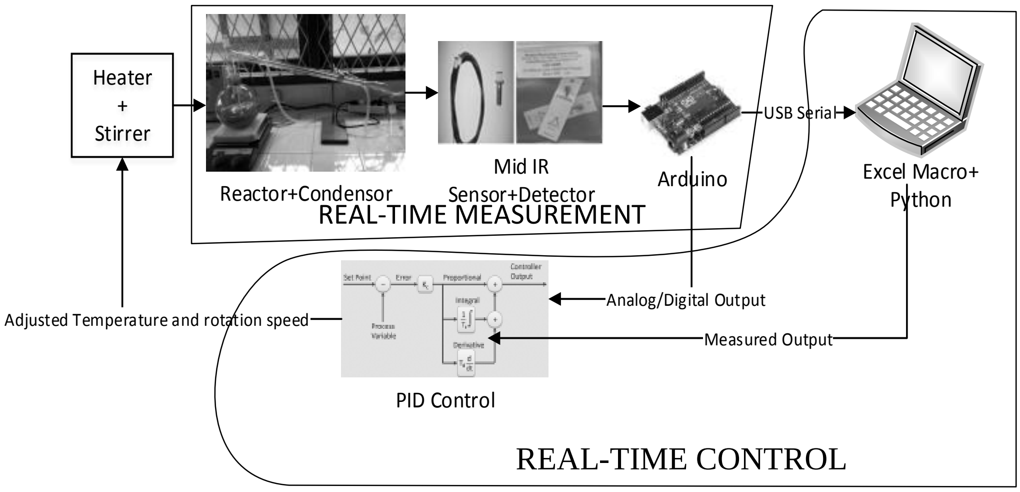


Figure 23 Peripheral block diagram of the integration real-time control system

In Figure 23 has shown the integration of real-time measurement block as implementation in Arduino (Ivrea 2005) with sensors and real-time control block that run in python coding (Python 2015) in a computer as completely shown in Appendix 3. This proposed of control mechanism was very useful in adaptive real-time process monitoring and control in esterification using heater and stirrer, Arduino and computer to develop an RTO (Adetola *et al.* 2009).

Experimental Results

From laboratory scale equipment and integration between sensor and algorithm that explained in this section, we conducted a performance comparison between the traditional control and the proposed method of adaptive optimization control with one sequence of batch learning for the computational intelligence to find the optimum cluster with Silhouette score and set the optimization control. In traditional control, we used monitoring by a human to control the set temperature at 230^{0}C and process time as 120 minutes.

Table 11 Selected mid-IR sensor

Minutes	Sensor 3.4µm (x_1)	Sensor 5.5µm(x_2)	Sensor 7µm(x_3)
0	ON	OFF	ON
30	OFF	ON	ON
60	OFF	ON	ON
90	ON	ON	OFF
120	ON	ON	OFF

For the adaptive proposed method, as Relief algorithm to select the sensor wavelength that closely related to esterification process as a result of comparison between weight average score from Relief in batch learning process for 0, 30, 60, 90, 120 minutes (Table 11) and real-time adaptive control method with the Pillar k-means algorithm used in this paper, we set α=0.25 and β=0.58 for the detection the outliers as used in Barakbah (2010) and the data points with silhouette score as in Table 12 to Table 15.

To find the best of silhouette score, first we run the clustering algorithm in K=5 (5 clusters) to get the data point for each cluster

Table 12 Cluster set for K=5 with Silhouette score of 0.536

Cluster	Data points of time (Minutes)
0	87,89,90,91,92,94,95,96,97,98,99,100,101,102,103,104,105,106,107,108,109,110,111,112,113,114,115,116,117,118,119,120,121,122,123,124,125,126,127,128,129,130,131,132,133,134,135
1	6,7,8,9,10,11,12,13,14,15,16,17,18,19,20
2	1,2,3,4,5
3	21,22,23,24,25,26,27,28,29,30,31,32,33,34,35,36,37,38,39,40,41,42,43,44,46
4	45,47,48,49,50,51,52,53,54,55,56,57,58,59,60,61,62,63,64,65,66,67,68,69,70,71,72,73,74,75,76,77,78,79,80,81,82,83,84,85,86,88,93,

Next we run for the clustering algorithm in K=4 (4 clusters) to get the data point for each cluster and compared to another number of cluster.

Table 13 Cluster set for K=4 with Silhouette score of 0.489

Cluster	Data points of time (Minutes)
0	107,109,110,111,112,113,114,115,116,117,118,119,120,121,122,123, 124,125,126,127,128,129,130,131,132,133,134,135
1	33,34,35,36,37,38,39,40,41,42,43,44,45,46,47,48,49,50,51,52,53,54,5 5,56,58,59,60,61
2	1,2,3,4,5,6,7,8,9,10,11,12,13,14,15,16,17,1819,20,21,22,23,24,25,26, 27,28,29,30,31,32,57
3	62,63,64,65,66,67,68,69,70,71,72,73,74,75,76,77,78,79,80,81,82,83,8 4,85,86,87,88,89,90,91,92,93,94,95,96,97,98,99,100,101,102,103,104 ,105,106,108

Finally, we run for the clustering algorithm in K=3 (3 clusters) for type 1 that has purity 80% and type 2 with 90% purity as in Table 14 and Table 15.

Table 14 Cluster set for K=3 raw material type 1 with Silhouette score of 0.604

Cluster	Data points of time (Minutes)
0	107,109,110,111,112,113,114,115,116,117,118,119,120,121,122,123, 124,125,126,127,128,129,130,131,132,133,134,135
1	54,62,63,64,65,66,67,68,69,70,71,72,73,74,75,76,77,78,79,80,81,82,8 3,84,85,86,87,88,89,90,91,92,93,94,95,96,97,98,99,100,101,102,103, 104,105,106,108
2	1,2,3,4,5,6,7,8,9,10,11,12,13,14,15,16,17,18,19,20,21,22,23,24,25,26, 27,28,29,30,31,32,33,34,35,36,37,38,39,40,41,42,43,44,45,46,47,48,4 9,50,51,52,53,55,56,57,58,59,60,61

From the result, it shows the conditions and characteristic for the control set of the esterification process in the laboratories, the data clustering carried out with K=3 gave a silhouette score of 0.601-0.604, and with K=4 a silhouette score of 0.489.

Table 15 Cluster set for K=3 raw material type 2 with Silhouette score of 0.601

Cluster	Data points of time (Minutes)
0	109,110,111,112,113,114,115,116,117,118,119,120,121,122,123,124,125,126,127,128,129,130,131,132,133,134,135
1	59,60,61,62,63,64,65,66,67,68,69,70,71,72,73,74,75,76,77,78,79,80,81,82,83,84,85,86,87,88,89,90,91,92,93,94,95,96,97,98,99,100,101,102,103,104,105,106,107,108
2	1,2,3,4,5,6,7,8,9,10,11,12,13,14,15,16,17,18,19,20,21,22,23,24,25,26,27,28,29,30,31,32,33,34,35,36,37,38,39,40,41,42,43,44,45,46,47,48,49,50,51,52,53,55,56,57,58

The silhouette score with K=3 with two type of feed composition type 1 with 80% and type 2 with 90% purity was higher than K=4 and K=5, so we decided to use three control of clusters as heating up, stabilizing and finishing for temperature position level and rotational stirrer speed control parameter as in Table 16.

Table 16 Control set with cluster

Cluster	Time (Minutes)	Temperature position level	Stirrer speed position level	Mode
2	1-58	5 (Full)	2	Heating Up
1	59-108	3-4	3-4	Stabilizing
0	109-135	2	4	Finishing

and by comparison the result from existing methods (traditional) that operator was run the process using the set temperature and stirrer as previous research from literature (Pardi 2005), which is not use the control system listed in Table 17 that compared in parameters as process time, yield, stirrer speed and final temperature with variations in raw material purity.

According the experiments results of this research, we found the advantage of this proposed model are in the esterification process finished in a shorter time than with existing methods, an increase in the percentage yield of product, a lower final temperature in the reactor (reduce the consumption of energy), lower range of stirrer rotation using two type of feed composition of glycerol with 80% and 90% purity.

Table 17 Comparison result of existing and adaptive methods

Comparison	Methods		
	Existing (Non-Controlled)	Adaptive used of 80% raw material purity	Adaptive used of 90% raw material purity
Process Time	120 minutes	105 minutes	100 minutes
Yield (%)	75	77	85
Stirrer Speed (rpm)	200-500	300-450	300-450
Final Temperature(°C)	230	210	200

We listed the comparison of output from several methods as shown in Table 17. On the other hand, the disadvantage of this proposed model are this esterification optimization system works well in reaction with oleic acid but need a different sensor for the different reactant, this system still require learning batch and several precise laboratory tests for calibration.

Discussion

In this paper real-time monitoring was supported with adaptive classification method because of the phase of esterification process needs to identify used three IR sensors which has a different wavelength and different maximum and minimum measurement, with Relief method the selection of sensors was well implemented with specified intervals within 30 minutes. This 30 minutes related to control respond as a characterization of the reactor and the heater in this experiments. Next, the output value from each sensor in analog signal was converted in data acquisition system to defined the state of esterification.

For data training we used 10 samples of data for calibration and defined the number of clustered data in three sections because with adaptive k-means that tested by set K=3, 4 and 5, for K=3 we get the highest silhouette score as 0.604 it means the best (suitable) number of data clusters for parameter control are three. We did not test for a number of the data cluster more than five because the esterification process has very long process time and slow of control response in the reactor. To set temperature and rotational speed control, in this paper we set the variable position of servos. As shown in Table 17 the implementation of the new adaptive system has reduced the process time in esterification.

In comparison to the previous research from Chryssolouris *et al.* (1992) that synthesized the state variable estimates determined by the different measurement and corresponding process models through a mechanism based on training such as a neural network and not use clustering method, in this chapter we proposed the adaptive control in cluster with Pillar k-means algorithm using sensors with feature selection.

To identify the dynamic condition in esterification process more precisely, the next chapter we implemented an adaptive sensor selection method, to improve the measurement of esterification identification state in real time.

Conclusion

In this chapter, we have developed a new approach for optimization process of esterification glycerol with oleic acid using Relief feature selection to select the sensors at specified intervals within 30 minutes supported with Pillar k-means data clustering algorithm. This clustering method used to determine the set control parameters like temperature and rotational speed of stirrer in real time adaptive control which has reached optimality using 3 clusters. From the experimental results of this chapter show a new approach for this research is able to identify the conditions of the process, increased product yield 10% or more, reduced process time up to 20 minutes, reduced range of stirrer rotation between 300rpm to 450rpm and reached a lower final temperature between 200^0C to 210^0C. The recommendation for future research is the model will develop using IR sensors with variable wavelength an another adaptive algorithm for sensor selection.

6 REAL-TIME OPTIMIZATION USING GRADIENT ADAPTIVE SELECTION OF INFRARED SENSORS FOR GLYCEROL ESTERIFICATION

Abstract

Recently, derivatives production of glycerol by esterification process, has many constraints such as the yield of target production which is not constant and lack of efficiency. As previous research, the yield and efficiency was improved by using accurate monitoring and control of the process. As a new development, we used a Real Time Optimization (RTO) using gradient adaptive selection sensors and control to optimize esterification process which has to cover various disturbance and uncertainty in real time mode. Thus, the objectives of this chapter are: to analyze the integration of esterification process using Self-Optimization (SO) combined with Necessary Condition Optimum (NCO) supported with laboratory scale mid-IR sensors, to develop a real-time control state with adaptive selection sensors system with computational intelligences and to measure the proposed optimization system indicator in batch process. To achieve those objectives, firstly a Business Process Modelling and Notation (BPMN 2.0) was built to describe the tasks of SO workflow in collaboration with NCO as an abstraction for conceptual phase. Next, using the modeling and its implementation with Stateflow package was deployed to simulate the three states of gradient-based adaptive control combined with Support Vector Machine (SVM) classification. This method was validated by running the esterification process in laboratory scale apparatus. In validation, RTO with adaptive selection sensors showed increased yield up to 14%, reduced the process duration up to 20 minutes, with the effective stirring speed between 300 rpm to 400 rpm and reaction temperature between 200°C to 210°C.

Introduction

The esterification product such as monoglyceride and diglyceride are representing high needs as modifying agents and showing steadily incremental for industrial consumption in the future, as they have various industrial application. This product as a raw material is required in pharmaceutical, cosmetics, and personal care industries, also in ink manufacturing (Pagliaro and Rossi 2008, Fernandez *et al.* 2005, Mostafa *et al.* 2013). Another use for a derivative product of glycerol in Indonesia, particularly in oil mining, this substance used for mixing to produce OBM (Oil Based Mud) and WBM (Water Based Mud). Production of monoglyceride from esterification of glycerol and synthesis of middle or long-chain fatty acids offers promising industrial opportunities (Hui 1996).

In esterification process, as previously researched (Srinivasan *et al.* 2003), there is an influence of process variables as temperature and time requirement affected their efficiency in esterification study. This process has constraints such as inconstant yield of production target and lack of efficiency for it has synthesize reaction that needs high-energy in the reactor to achieve desired temperature and time for the process. As explained in previous chapter, to optimize the esterification reaction to get a glycerol monooleate substance as target of product, it is essential to undergo the process real time mode, which has to cover various disturbance and uncertainty.

Traditionally, this type of process was operated using operator judgment (experienced-based) or non-controlled. By traditional method, it is not possible to exactly determine when it is necessary to supply the energy for heating up the reactor or when the process has completely formed the target product, especially when the new type of raw material with different purity was used. In this chapter, we improved the precision of real-time mode measurement with sensor selection method to accommodate the control of process variables more precisely.

As related in this chapter, the Real Time Optimization henceforth referred to as RTO is used to indicate the continuous re-evaluation of selecting variables in operation (Chacuat *et al.* 2009) as a part of this research and in the next development of RTO for chemical process based on necessary condition optimum (Srinivasan and Bonvin 2007). With recent advances in digital hardware and optimization software the RTO method can connected to a computer control systems (Bocker *et al.* 2006).

In recent years, spectroscopic methods using infrared has gained popularity to be chosen in industrial process control in real time, especially for esterification process (Blanco *et al.* 2004). A systematic and rational approach such as feature selection was required in order to accommodate different sources of sensors and process fluctuations as a dynamic condition such as data quality that can affect monitoring and classification performance from class imbalance and noisy attributes (Yusta 2009) and adjusting the set point basic control accordingly to adapt the system requirements. Feature selection is generally used in a machine learning when the learning task involves high-dimensional and noisy attribute datasets as parameter as observed in real time sensor application. In this chapter, feature selection with gradient measurement was used to select the type of appropriate sensor that related to measurement in this esterification process. This higher dimensional result was improved the accuracy in monitoring measurement, especially for yield measurement.

In this chapter, we set the combination of RTO with adaptive sensor, supported by computational intelligence; so the objectives of this chapter are to analyze and design the dynamic condition process of glycerol esterification in detailed, in which Business Process Model and Notation version 2.0 was used. Next. gradient measurement used to select the sensors then supported with computational intelligence such as classification by Support Vector Machine (SVM) consecutively set to determine the state condition, was applied in the microcontroller. As validation for esterification process, the investigated system consists of several performance indicators that have to be fulfilled; those are yield, process time, stirring speed and temperature.

Methods

Current Development in Real-Time Optimization

RTO concept was developed based on Self-Optimization (SO), which is defined in Skogestad (2000) as a situation when we achieved an acceptable loss with constant set point values for the controlled variables without the need to re-optimize when disturbances occur. In this chapter, the previous method combined with Necessary Condition Optimum (NCO) (Srinivasan *et al*. 2008). However, according to the research from Ye *et al*. in 2012 stated that it is necessary to measure all of the NCO components in real time. By validation, we claimed this new optimization of combined method, supported with measurement by adaptive selection sensor and real-time data acquisition system, is useful for chemical industry application that has a dynamic condition such as esterification process. In sensor deployments, each sensor collected data at regular time intervals, captured a time series representing the occurrences of dynamic condition and build the database that needs classification approach (Alstad *et al.* 2009). In this chapter, the developed active control variables are obtained in real time with measurement from the sensor with adaptive selection and computed for tracking the NCO to select the state with computational intelligence classification. The real-time task has an integral action to track the selected sensor using the adaptive method and to set the control at necessary set point with the state. The set point was determined as considered to the best performance condition of those apparatus such as a heater and agitation motor.

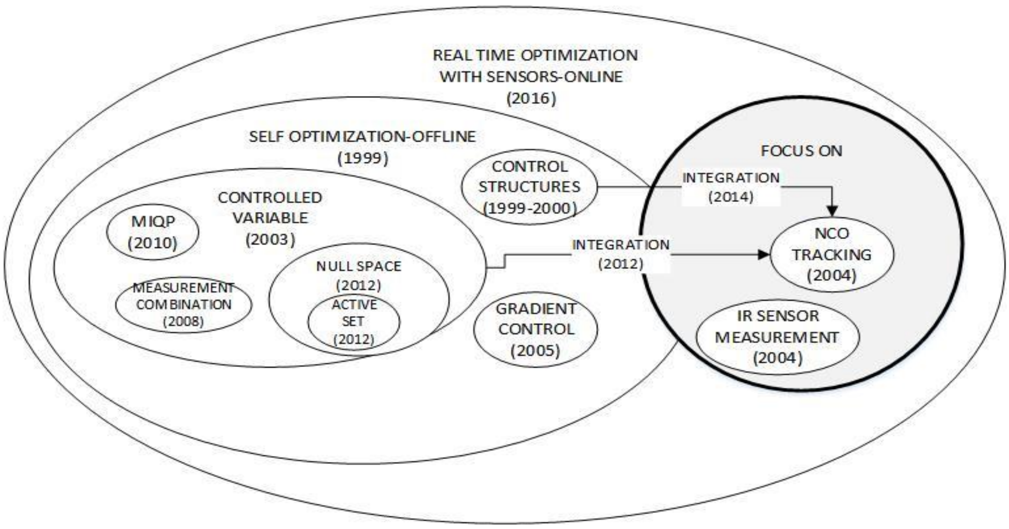


Figure 24 Nested relationship between SO and NCO in RTO

In the Figure 24 as shown the diagram of an integration of SO and NCO method to set in real-time control process that focused on NCO tracking and IR sensor measurement. The concept of SO is a strategic aim to appropriately select the controlled variables (CVs) in control structures so when they are maintained at constant set points, the overall plant operation is optimal or near optimal despite various disturbance with implemented of measurement combination (Francois *et al.* 2005); this concept was related to offline system. To develop a real- time system and improve the performance, the concept of SO was integrated with NCO that has related to the CVs to control the process (Halvorsen *et al*. 2003).

Implementation of RTO Method with Gradient and State Control to Optimize Process Time

Many run-time process variations need to be accounted for, especially in the industrial process with chemical reaction, typically to cope with these uncertainties by adopting a conservative strategy that guarantees constraint satisfaction even in the worst-case situation. This measurement can be used in an optimization framework to compensate the effects of uncertainty in the form of model mismatch or process disturbances. Nowadays, Real-Time Optimization with Necessary Conditions of Optimality (NCO) proposed by Jäschke (Jaschke and Skogestad 2011) and optimal operation is achieved by designing a "smart" control structure. As SOC was combined with NCO tracking (Srinivasan *et al*. 2003) or zone control MPC (Graciano *et al.* 2015), it's become Real Time Optimization that using online model. Among the various options for input adoption, promising approach consists of directly enforcing the necessary conditions of optimality (NCO) that include two parts, the active constraints and the sensitivities (Srinivasan *et al.* 2008). The use of measurements to compensate the effect of uncertainty has recently gained attention in the context of real-time optimization of dynamic systems.

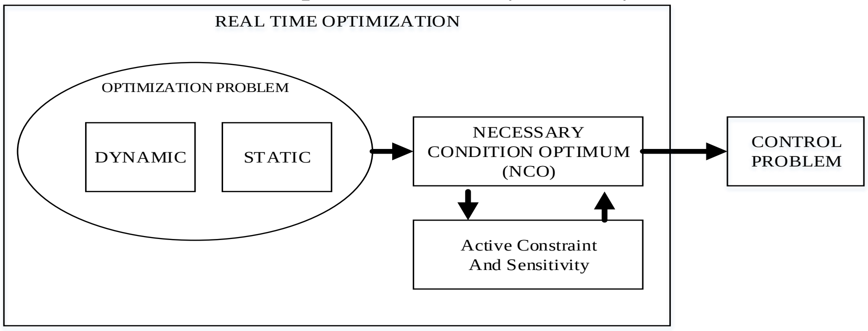


Figure 25 Connection diagram of NCO and control

In Figure 25 the RTO has commonly used approach consists of updating a process model and performing numerical optimization problem using the refined model as is dynamic and static condition. In this chapter, we used the methodology as in NCO, there is a two-level approach that does not require repeating the optimization as named active constraint and sensitivity. At the upper level, the constraints that are active in the optimal solution are identified from optimization of a nominal process model. At the lower level, feedback control is used to enforce the necessary conditions of optimality to define control problem in matched criteria.

To select the appropriate sensor for data processing in real time condition within 3 different wavelengths of IR sensors from the model developed in the previous research (Soenandi *et al.* 2015), and to improve the performance in measurement a gradient method was used to select the appropriate sensors for regression. The gradient of f is defined as the unique vector field whose dot product with any vector v at each point x is the directional derivative of f along v (Korn 2000) as shown in Equation 21, that is:

$$(\nabla f(x)) \cdot \mathbf{v} = D_{\mathbf{v}} f(x) \tag{21}$$

which:
$\boldsymbol{\nabla f(x)}$= vector field
f(x)= vector point
Dv= derivative of directional

In this research, we measured the gradient by comparing sample data points from 2-dimensional field as identified by the sensors. The formulation can be seen in Equation 22 and by limiting the gradient value m_i we set the decision to use the data or pass it,

$$m_i = \frac{t_n - t_{n-1}}{x_n - x_{n-1}} \tag{22}$$

which:
t_n= time interval sampling
x_n= value from sensor

The optimization algorithm uses the process model and the objective function to solve the new optimum state for the process. Afterward, the optimal operation can be formulated in Equation 23 as maximizing a process time function t:
Objective function:

$$\max_{T,R}\, y(\mathbf{T}, \mathbf{R}, \boldsymbol{S_i}, \mathrm{C_n}) \tag{23}$$

With decision variables: **T**: temperature (°C)
R :stirrer rotation (rpm)
S$_i$:i^{th} sensor measurement with selection
C_n: n^{th} control state

Subject to: $C_n = f_i(S_i)$

$$g(\mathbf{T}, \mathbf{R}, \boldsymbol{S_i}, \mathrm{C_n}) \leq 0 \tag{24}$$

where f_i is the measurement set that represents the process from sensor, g is the process constraints set.

In addition, assumed that we had a plant measurement model from the sensor

$$\mathbf{Y} = f^y(\mathrm{u}, \mathbf{X}, \mathbf{D}(\mathrm{t})) \tag{25}$$

Where **Y** is the n-dimensional vector of measurements, and f^y is the function mapping the variables u, **X** and **D(t)** onto the measurement space. As researched by Datskov in 2006, the optimal operating point found by solving Equation 25 that must be realized through a control system. To implement the control system, any number of degrees freedom as changeable variables from the optimal operating point vector must be specified and classified in the identification process stage with Support Vector Machine (SVM) algorithm and developed adaptive control.

Several reactions and variation conditions were tested in Surfactant and Bioenergy Research Center (SBRC) Bogor Agricultural University at Baranangsiang. For RTO of esterification process, real-time data and laboratory data were integrated and merged first, both in steady state manner. In measuring the performance, yield is one of the most important indexes for the esterification process. The yield was calculated for operation evaluations in each batch basis.

Experimental Results

Analysis and Design System

To support the new concept of RTO, in this research BPMN 2.0 diagrams (Sap 2013) was used to analyze and design the system modelling for esterification process by abstraction for conceptual design to describe the section of SO and NCO in detail (Figure 26), with this diagram we can describe more detail for each task of digital apparatus and communication between them, which will be the base structure for programming in Arduino as described in Appendix 8 to run the implementation step such as data acquisition and controlling in the esterification process.

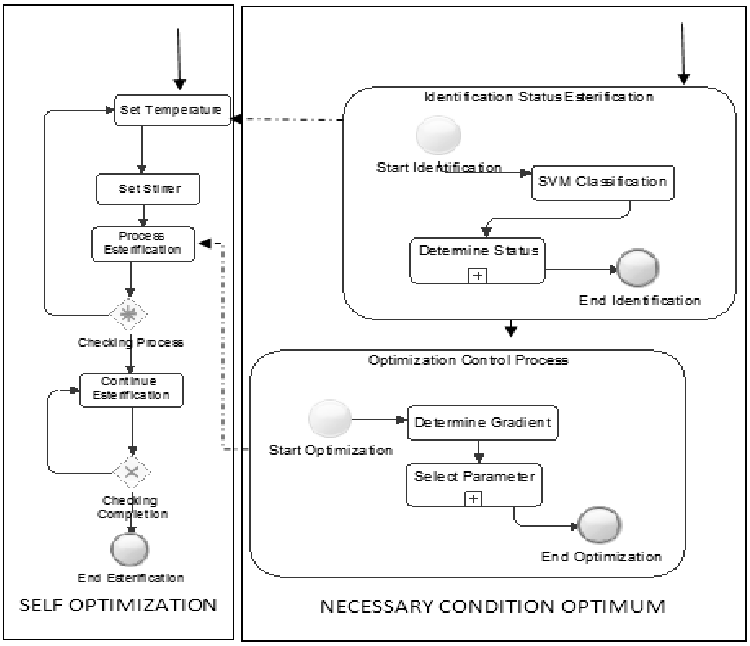


Figure 26 BPMN diagram for SO and NCO

Characterization Tests

This study carried out several characterization tests for calibration the signal from sensors, both for the identification and validation of the process results, such as Fourier Transform Infrared Spectroscopy (FTIR) and Gas Chromatography-Mass Spectrometry (GCMS). The information (transmittance level) from FTIR Test was used for selecting the wavelength for the sensor. The selected wavelengths, regarding the difference of transmittance level between before, during the process and after esterification were: 3400nm, 5500nm and 7000nm in Figure 27. To confirm this selected wavelength, a graph of GCMS test was carried out to ascertain the level of (yield) glycerol resulted from the end of esterification as the production target during the process span, generated by Sigmaplot 13.0 (Crane 2014).

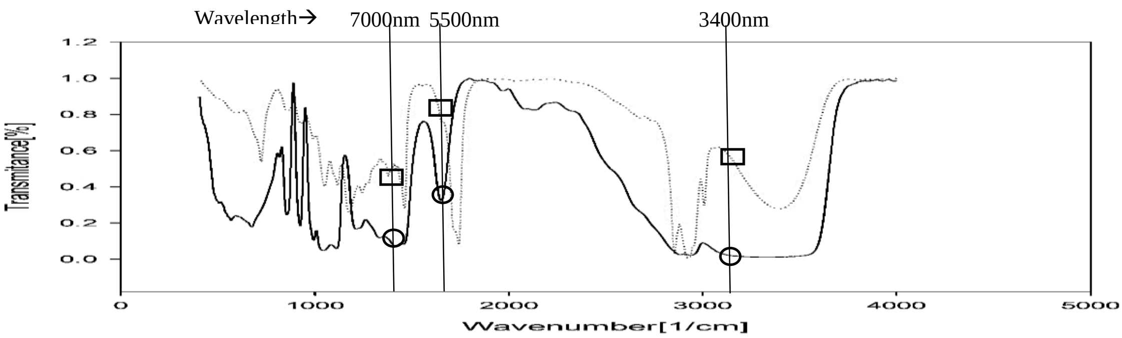


Figure 27 Spectrum FTIR before esterification (○) and after esterification (□)

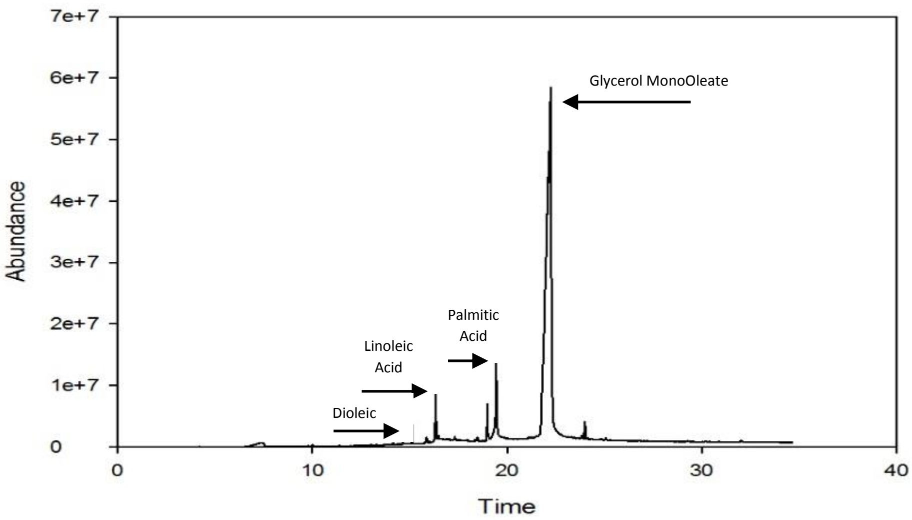


Figure 28 GCMS spectrum for product testing from sample with the treatment of 200°C for 120 minutes

The test of GCMS in sample with variance in temperature and process time compared with a database of WILLEY09TH.L was detected as glycerol monooleate by the reference number of 587486 with molecular weight =356. The yield obtained at 200°C for 100 minutes' process using raw material with 90% purity was 90% as shown in Figure 28 and more detail in Appendix 9 and 10, which was the highest obtainable yield compared to another condition.

State Flow Model

This model assisted in predicting and simulating the behavior of control with state, using Stateflow (MathWorks 2014). In this stage, we wanted to simulate a control logic tool used for modelling reactive systems via state machines and flow charts within a Simulink model with specific applications in mode logic, where each discrete mode of a system represented by a state was deployed as control model as in Figure 29,

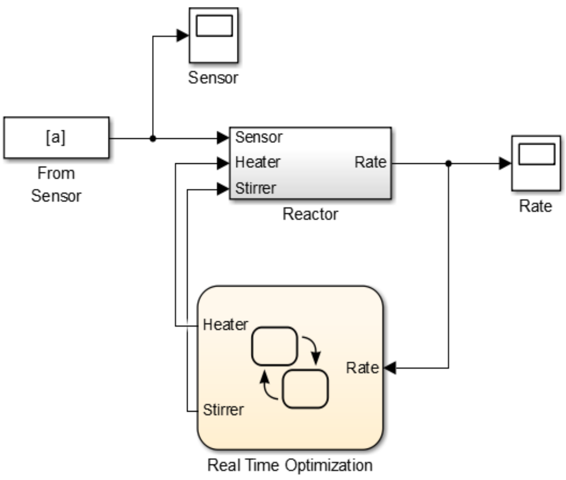


Figure 29 Control model of self-optimization in Stateflow

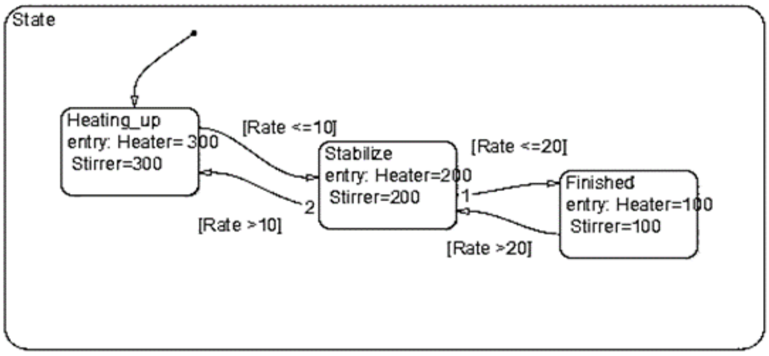


Figure 30 Control state model in Stateflow

In Figure 30 shown the Stateflow model that has three states as heating up, stabilize and finished. Each states has each control variables for heater, stirrer and rate. In this research we have set for the heating up states of heater and stirrer was 300 angle degrees position of switch (zero angle degree in off and 340 angle degrees in full), for stabilize states of heater and stirrer was 200 angle degrees position of switch and for finished states the position switch of heater and stirrer was in 100 angle degrees.

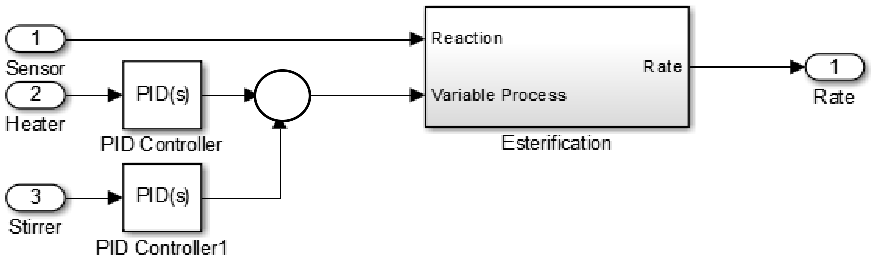


Figure 31 Control model of reactor in Stateflow

In Figure 31 in PID block we set the constant for Stateflow in heater and stirrer block using laplace transform with coefficients for the proportional, integral, and derivative P=1.1, I=1.2 and D=0.5 respectively, as a model of control reactor characterization. In esterification block there is reaction and variable process that need to define as a function. The block of esterification connected to rate that plotted the reaction of control system as a responds of disturbance.

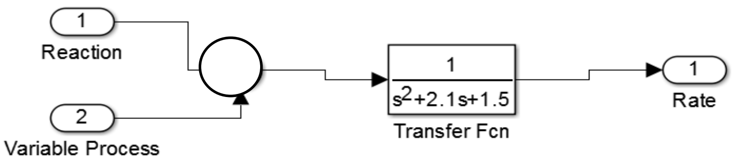


Figure 32 Control model of esterification in Stateflow

In control model of esterification we set the transfer function with numerator coefficient is [1], denominator coefficients are [1 2.1 1.5] and absolute tolerance [auto] as in Figure 32 that we obtained by using Tfest function in Stateflow (MathWorks 2014) with esterification data from laboratory experiments.

Real Time Data Acquisition

Data acquisition was defined as processing of sampling signals that measure real world physical conditions (usually using sensors) and converting the result into digital numeric values that can be manipulated by a computer. In this research, real-time data acquisition was operated in Arduino Mega 2560, connected via USB 2.0. The database was built in Personal Computer with specification of Core i5 2.2 GHz CPU, 4GB RAM, using Microsoft Excel with add-in program named PLX-DAQ (Parallax-Data Acquisition) to collect the data from sensor model set (S_1, S_2 and S_3) listed in Table 18. Then, the task of selecting the sensor and the adaptive control was sequentially run in. The task of data acquisition from the sensors was collected in one-second interval during 130 minutes of each batch process. Hence, the total data collected in the database was approximately containing 7200 items data for each sensor and the sensors was selected by using gradient value minimum of 1.6 in 10 second interval.

Table 18 List of sensor model

Symbol	Description	Availability	Sampling Period	Number of samples
S_1	The temperature of reactor	Real-time	1 s	7200
S_2	The composition of target yield	Real-time	1 s	7200
S_3	The speed of agitation	Real-time	1 s	7200

Especially for the measurement of composition target yield (S_2), after the sensor mid-IR selection step, we developed regression analysis for estimating the relationships among variables (X_1, X_2 X_3) from sensor input. In this research, we performed regression for converting variables of sensor measurement to measure the yield of the esterification reaction, as for the result can be seen in Table 19. For non-adaptive measurement, we used regression $Y=-33.62390+ 1.04874(X_1) +0.8362(X_2)- 0.29288(X_3)$ from the start to the end of the process. But in adaptive selection sensor, we performed different regression for each state (Table 19). To check the regression of those equation for adaptive selection, we have computed the coefficient of determination from each state and the range was from 0.9280 to 0.9878.

Table 19 Yield regression (S_2) for adaptive sensor selected in each state

State	Sensor Selection			Regression	R^2
	3.4μm (X_1)	5.5μm (X_2)	7μm (X_3)		
1	ON	ON	ON	$Y= -33.62390+ 1.04874(X_1) + 0.8362(X_2)- 0.29288(X_3)$	0.9668
2	OFF	ON	ON	$Y= 59.85565-0.04523(X_2) +0.062339(X_3)$	0.9878
3	ON	ON	OFF	$Y=26.77324+0.039855(X_1) +0.709494(X_2)$	0.9280

For the esterification data measurement, we retrieved from Chapter 5 page 34. The real-time data plot acquisition using 3 wavelength mid-IR sensors within time length 7200 seconds, the value was a digital bit number output related for transmittance level in the esterification process, in each wavelength and determine the track of data with this regression.

The measurement of yield from the regression was resulted by collected data input from sensors. We compared it by setting in a non-adaptive sensors selection (revered to Chapter 5) and adaptive sensor mode using two samples of purity, to find the different yield measurement from each mode, for the next step using in validation.

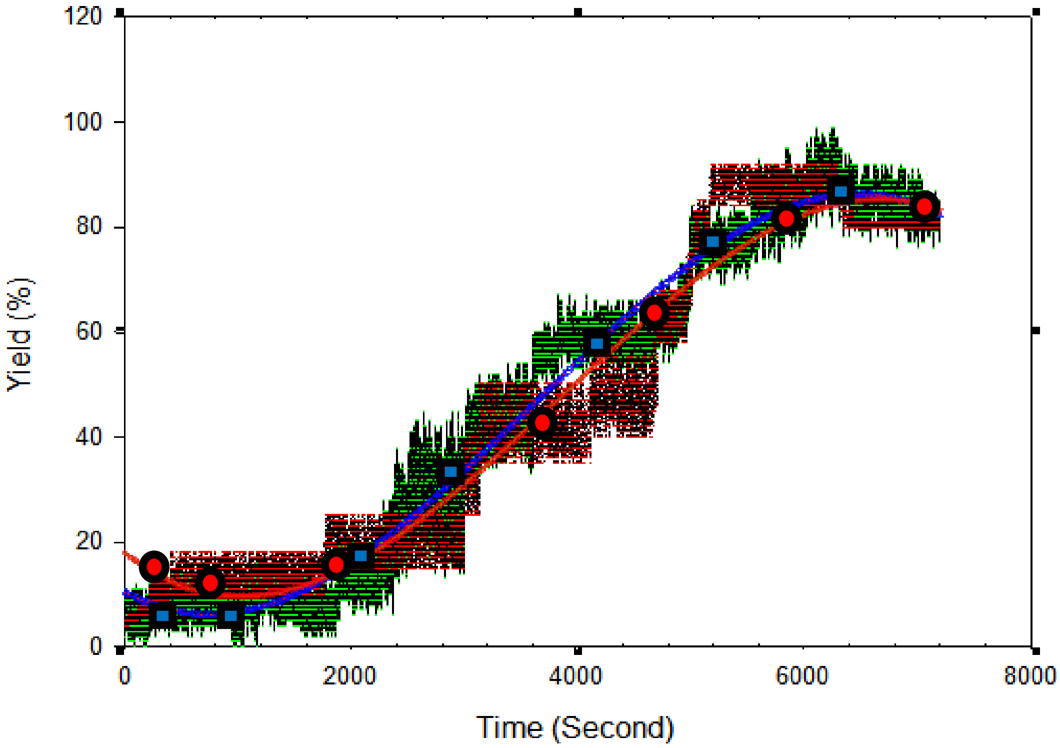


Figure 33 Comparison of yield measurement from mid-IR sensors with adaptive gradient selection (■) and non-adaptive (●)

As shown in Figure 33 to measure the performance of development method, we compared the yield measurement using regression. As seen in the figure, there are slightly difference in yield measurement between adaptive and non-adaptive methods, especially in 500, 1000, 5000 and 6000 second.

Table 20 Comparison yield from non-adaptive and gradient adaptive selection

Sample	Yield (%)		
	Non Adaptive Sensor	Adaptive Sensor with gradient selection	GCMS Test
80% purity glycerol processed in 210°C and 105 minutes	75 Percentage error 5.33%	78 Percentage error 1.28%	79
90% purity glycerol processed in 200°C and 100 minutes	85 Percentage error 4.70%	89 Percentage error 1.12%	90

By comparing the yield measurement from non-adaptive sensor and adaptive sensor selection with gradient, we obtained the smallest percentage error in experiment using 90% purity raw material with treatment of 200°C for 100 minutes as 1.12% as shown in Table 20.

Table 21 Some sampling time for SVM in state control variable

Sampling time (min)	Selecting Sensor			Controlled Variable	
	Sensor 1	Sensor 2	Sensor 3	State Temperature	State Agitation
0	On (10)	On(15)	On(16)	Heating-up	Heating-up
30	On(20)	On(30)	On(40)	Heating-up	Work on process
70	Off	On(45)	On(46)	Work on process	Work on process
88	On(54)	On(60)	Off	Finishing	Finishing
120	On(73)	On(60)	Off	Finishing	Finishing

In this research, Table 21 was shown the result for adaptive gradient selection sensor implementation as on or off for each sensor, in several sampling time

Table 22 Comparison result of existing and adaptive selection sensors

Comparison	Methods		
	Existing (Non-Controlled)	Optimization with real time adaptive control	Real time optimization using sensors adaptive selection of sensors
Process Time	120 minutes	100 minutes	100 minutes
Yield (%)	75	85	89
Stirrer Speed (rpm)	200-500	300-450	300-450
Final Temperature(°C)	230	210	200

In Table 22 we listed the comparison of esterification time process reduction between optimization with real-time adaptive control and real-time optimization using sensors adaptive selection of sensors

Discussion

In this research to improve the yield measurement is an important step to optimize the esterification process precisely. In comparison to previous research from Srinivasan *et al*. (2008) that develop the method for tracking the necessary conditions of optimality with changing set of active constraints using a barrier-penalty function. That method still have a problem when the set of active constraints is unknown or changes due to uncertainty. This problem was solved by implementation of adaptive selection of sensors and cluster set of control.

The implementation of analysis and design by using BPMN 2.0 was successful to describe the requirement of SO and NCO for the dynamic condition in esterification process.

We found the accuracy of yield measurement using adaptive methods with the gradient method was more precise and it had reduced the process time of esterification. The equation of regression was deployed to define the yield measurement as a number. We set the adaptive selection sensors worked in a gradient value between 1.6 and 1.8 with 10 seconds' time interval because in this gradient and time interval the control for heater and strirrer was obtained to increase the yield of glycerol esterification.

In order to measure the yield exactly, we compared the sample of production of adaptive gradient sensor selection with GCMS test; we used only several sample tests in order to simplify the validation and to convince the sensors has been well calibrated. For the comparison of optimization indicator, we have tried the experiments until we obtained 75-89% as highest performance using laboratory scale apparatus, which maximum temperature limit was set at 210ºC to extend the lifetime operation of the heater.

For validation purpose, it is necessary to indicate related information of adaptive selection sensors performance using a gradient with the regression to measure the yield measurement between non-adaptive and adaptive sensor selection using two samples of raw material (Table 20). Result showed that there was an improvement in measurement of yield using the adaptive method. Those result prove the output of SVM in state control, which was listed partially in Table 21 with state, was clustered in proportionally. According to the improvement result in this chapter, for the next chapter, we proposed the scaling up model for higher capacity in industrial implementation.

Table 23 Comparison of optimization indicators

Indicators	Comparison			
	Adaptive Control in Raw Material Purity of 80%	Adaptive Control in Raw Material Purity of 90%	Adaptive Control with Gradient in Raw Material Purity of 80%	Adaptive Control with Gradient in Raw Material Purity of 90%
Process Time (minutes)	105	100	100	98
%Yield	77	85	78	89
Stirring Speed (rpm)	200-500	200-500	300-400	300-400
Maximum Temperature (°C)	230	230	210	200
Average Temperature (°C)	178	177	174	172

Then, in Table 22 we listed the comparison of optimization indicators in real time was summarized as indicators as process time, %yield, agitation speed, max temperature and the average temperature

As a comparison between control system that we get from Chapter 4 without gradient selection of sensor and adaptive control with gradient selection,

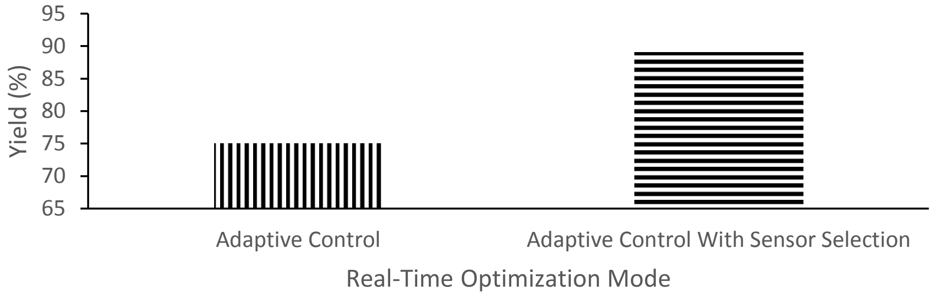


Figure 34 Barchart of real-time optimization mode indicator vs yield

We have summarized the optimization parameter in bar chart for real-time optimization mode as measurement of yield shown in Figure 34 that adaptive control with selection sensor has higher yield of production.

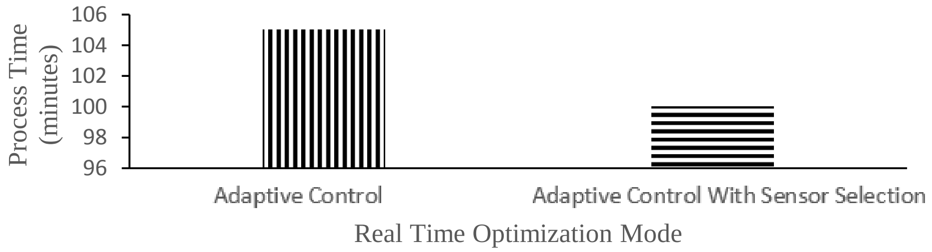


Figure 35 Barchart of real-time optimization mode indicator vs process time

In Figure 35 has shown the optimization parameter as process time that adaptive control with sensor selection as a result the adaptive control with sensor selection has lower process time. It means the performance of adaptive control with sensor selection was faster or better.

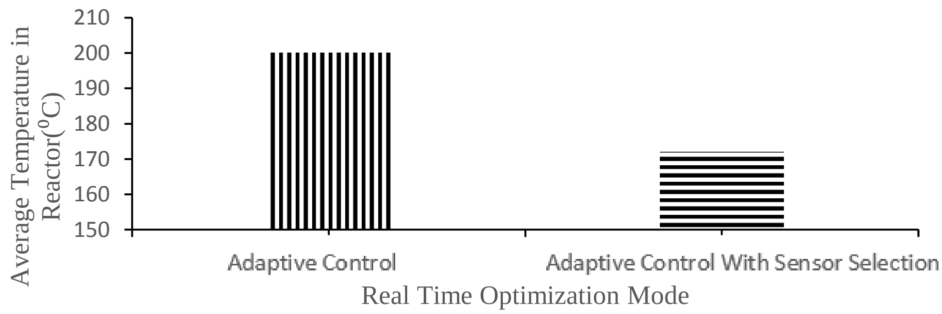


Figure 36 Barchart of real-time optimization mode indicator vs average temperature in reactor

In Figure 36 has shown the average temperature in reactor in real-time optimization mode. The adaptive control with sensor selection has lower average temperature, it means for the adaptive control with sensor selection increase the efficiency on energy consumption.

From all that three parameters, adaptive control with selection sensor successfully achieve improvement in all aspects.

Conclusion

This research presents a new RTO system of esterification oleic acid with glycerol using infrared sensors. As the result of this research, we have presented an abstraction for conceptual model optimization process in real time with BPMN 2.0 to ensure the implementation of RTO. For adaptive selection, mid-IR sensors work using gradient value minimum at 1.6 with 10 second time interval, followed by SVM and state control with three states, has achieved a good performance. For system validation, RTO shows that the responsiveness of control increased product yield up to 14% and reduced the required process duration in range up to 20 minutes, with an effective range of stirrer rotation set in range between 300 to 400 rpm and process temperature at 200°C to 210°C. For scaling up recommendation of the current research, it is necessary to improve the quality of sensor component materials and related control for apparatus integration.

7 A PROPOSAL FOR SCALING UP MODELING FOR A BATCH ESTERIFICATION PROCESS WITH REAL TIME OPTIMIZATION

Abstract

Scale-up process is an essential task in industrial development activities. Scaling up conversion for esterification batch processing is needed to approach the real production stage while it is still necessary for running in optimizing mode especially for the process time in larger scale of pilot plant. This proposal addressed the opportunity and potential of current research achievements such as real time monitoring, real time adaptive control and real time adaptive control with gradient selection for scaling up purposes. The objectives are to propose a formulation for scaling up process that a pseudo first order rate model was previously used and to find a conceptual and physical design for scaling up esterification process into a pilot-plant level. These objectives was achieved by developing the studies on existing laboratory scale in batch esterification process supported with Real Time Optimization (RTO) based on an empirical scale-up methods. This proposal are divided the scale-up as in conceptual and physical design. The conceptual scale-up plan were based on knowledge from system implementation in real-time data to ensure feasibility of temperature and mid-IR as inputs measurement. Next, a physical design model that was constructed with Stateflow, which was supported by data classification Support Vector Machine (SVM) was included in the model by using three types of implemented electrical transformer. These methods makes the proposed model are suitable for scaling up esterification in batch system with real-time data acquisition which was supported with a microcontroller. Finally, the design set of control using three types of activator that represented by transformer in the scale-up plan was proposed.

Introduction

The increasing global competition, push each companies in the processing industries sectors to face an intense, incremental pressure to improve production efficiency and product quality by scaling up the process (Caygill 2006). Esterification product was needed incrementally as high demand for various raw material that used in industrial sector (Pardi 2005). For the cheap oil sources contain high FFA like crude glycerol, the esterification step is usually required in order to convert that raw material to get the derivative product (Bail *et al*. 2012).

As these background of research above, to scale up the esterification process was needed immediately to fulfill the industrial purpose level. The scale-up of chemical processes, particularly those involving batch or semi-batch manufacturer, is a well-known problem in area of engineering development (Trevor 2010). It is not possible to build a plant without supporting calculations, studies, and demonstrations of its functioning at a smaller operating scale (Bisio 1985). For years, scale-up has been a sort of art in which expertise, rules of thumb, trial and error, and particular solutions have been implemented to obtain a proper result at a new operating scale, batch processing is considered to be important in the chemical industry, mainly when low production volumes or a great variety of products within a single process unit are required (Fernandez 2012).

In the previous chapter was described that state of the art in RTO industrial applications for batch system has been investigated by Darby *et al*. (2011) and Mansour and Ellis (2008). They identified the system that was run in batch process, it means the materials are loaded, the process is initiated, and as the reactions are completed, the products are removed. This proposal addressed the opportunity and potential of current research achievements such as real time monitoring, real time adaptive control and real time adaptive control with gradient selection for scaling up purposes. Generally, in this proposal we focused on the scale-up problems that was implemented with an advanced control system using RTO as requirements in higher volume and variable of reaction was controlled in real time.

Based on above motivations and challenges, the objectives in this proposal are to formulate the optimization in scale up process using pseudo-homogeneous model was used to describe the kinetics of oleic acid esterification and to find a conceptual and physical design for scaling up esterification process.

To fulfill the objectives and limitation above, as a first step of our methods, a formulation for the esterification scale-up based on mathematical and computation knowledge with assumption that the esterification reaction was proceeds as a pseudo first order rate that means the catalyst not involved in reaction (Geng *et al.* 2012) and from Chacuat *et al.* (2009) that stated an assumption for both internal and external mass transfer resistances can be omitted for most reactions in the presence of MESA catalysts. Next, implemented the basic principle of scaling-up as previous research from Bentolila (2013) which is using data collection from bench-scale laboratory equipment and exploit the data by design commercial-scale process configuration. From the research by Harmsen (2013) that the empirical scale-up method is often employed for polymerizations and fermentations in mechanically stirred vessels and the reactors are batch or fed-batch operated, in our case in esterification we also implemented this method in identical process. Next, with the flow of knowledge based on basic knowledge and laboratory experiments used to build the optimization model that the principal of optimality was used, the ideas was obtained to implement in pilot or industrial scale (Donati and Paludetto 1997). The control model was developed with knowledge generation in achievement in real time measurement using mid-IR sensors and real time adaptive control with gradient adaptive that deployed the model in Stateflow (Mathworks 2014) supported by clustering with computational intelligence from achievement in real time adaptive control, that finding the best control set for the scale-up process using voltage converter to control the heater within DC phase and AC phase power (Ramirez 2001) using multi clusters. Finally, the design set of control using three types of activator that represented by transformer in the scale-up plan was proposed.

Methods

To design the model of scale-up of esterification process we have several stages as framework, first we have formulation for the RTO as process and parameter for laboratory scale in optimum to implement the principle of optimality for scaling-up, next the conceptual design was proposed and finally the physical design was proposed in detail.

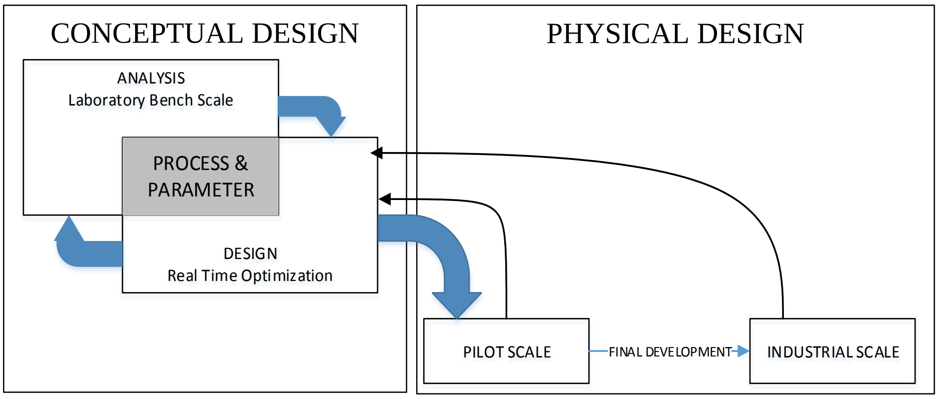


Figure 37 Basic stages in analysis and design for scaling up (Modified from Bentolila 2013)

As shown in Figure 37 the conceptual design block was based on analysis in laboratory scale an design of requirement for RTO which has similar process and parameter. Next, the physical design was proposed as a pilot scale and then for industrial scale for the next development.

Optimization of scale-up formulation

In this research, we defined the optimization for scale up esterification problems was minimizing time which it was necessary for setting the optimal control parameter in scale up to satisfy the requirements of process as a pseudo first order rate in assumption as in Real-time optimization using gradient adaptive selection of infrared sensors for glycerol esterification was formulated in Equation 26.

Objective function: $\min\ t_f(\mathbf{C},\mathbf{T},\mathbf{R})$ (26)

where decision variables are **C**: concentration of Monooleic(%)
T: temperature used (°C)
R: reactor stirrer (rpm)

subject to
$$150°C \le T \le 190°C$$
$$150\ rpm \le R \le 300\ rpm$$

boundary $t \in [0, t_f]$ t: time of esterification

Conceptual Design

In this research, the conceptual design was based on data collection from bench-scale laboratory equipment and exploit the data by design commercial-scale process configuration (Bentolila 2013) as detailed in Figure 38, the process and parameter was found from analysis and design using laboratory scale with implemented RTO. Next, from that laboratory scale we are increasing the volume to pilot plant. For further development in final stages would be used to scale up for industrial production.

The process and parameter for scale up was used from current achievement in our research in Chapter 3 page 16 that has result from real time simulation with SO that supported with sensors, has succesfully increase the volume of production up to 17% and process duration reduction by 36 minutes in pilot plant scale with stirring speed 300rpm to 450rpm and reduced average temperature that used in the reactor.

This conceptual stages also supported with research by Donati and Paludetto (1997) that stated the models contained all physical, chemical and disturbance effected on the performance of the process and the basic knowledge generated from laboratory scale experiments that have been implemented excellently and repeated ideas to implement in industrial unit or pilot unit.

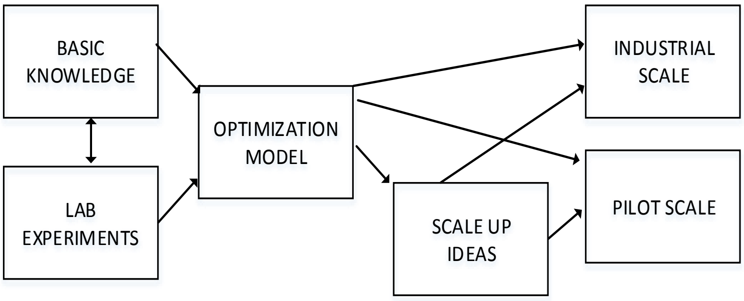


Figure 38 Knowledge flow for scaling up
(Modified from Donati and Paludetto 1997)

As shown in Figure 38 the flow was started with basic knowledge block that related to our research in Chapter 3 that real-time simulation for esterification process succeeded to improve the volume of production by implemented mid-IR sensor to identify the quality of product with Self Optimization and based on Chapter 4 that has successfully implemented the identification of esterification stages using three states or cluster with implementation of mid-IR sensors. Next, by doing laboratory experiments for optimization of esterification of oleic acid with glycerol in SBRC, we combined with basic knowledge block that we obtained in Chapter 4 in application of mid-IR sensors, we have developed the optimization model block in minimization process time that discussed in Chapter 5 with optimization model using real-time adaptive control that has an improvement for clustering the data from sensor and optimization model in maximization of yield in Chapter 6 that implemented a gradient adaptive selection to improve the precision of identification from infrared sensors. As the result of our experiments using application of RTO with adaptive selection sensors has successfully increased the yield up to 14%. Finally, from these model optimization block we want to implement the scale-up ideas to develop in pilot and industrial scale.

Physical Design

For physical design in scaling up of esterification process, we proposed the model measurement aspect as a complete steady state model of the reactor using the kinetics and mass transfer behavior built in the laboratory which predicts all of the design parameters and the input from sensors. Those aspects has several differences such as volume, temperature in reactor, stirrer speed, process time, control parameter and response of control. Especially in control activator with the same control parameter that we has implemented in laboratory scale we has design the model for scale up. The esterification process was designed at different stirring speeds of 300, 400, 500 and 600 rpm in a reactor with volume 25L, under the same reaction parameters of 65°C and 100°C, and reaction time range between 200 and 240 minutes.

In this chapter we started from analysis the esterification process as we have achivement in research from Chapter 3 using BPMN 2.0 diagrams that has different swimlane between SO and NCO operation from our current achivement in RTO with adaptive clustering in Chapter 5 and next development with gradient adaptive selection sensor in Chapter 6.

Table 24 Regression for scale up process

Volume (L)	Temperature (°C)	Process Time(minutes)	
1	210	100	$V = -71+0.2T+0.3P$ $R^2=1$
5	200	120	
10	210	130	
25	172	205	←Extrapolated

The design of RTO with scaling up was used assumption the process and parameter was have same characteristic between laboratory bench scale and scaling up system as empirical scale up method (Harmsen 2013). In Table 23 for volume 25L was extrapolated for process time predictions for 205 minutes. This model of requirements hopefully run smoothly and meet the time needed for esterification in high capacity production that operated in alternating current power phase for the heater as shown in Appendix 4.

ASPECT	LABORATORY-SCALE	SCALE-UP
VOLUME (L)	1	25
TEMPERATURE (°C)	200-240	150-190
RATE OF AGITATION (RPM)	200-400	150-300
CATALYST	MESA	MESA
PROCESS TIME (minutes)	105-120	205-210
CONTROL PARAMETER	TEMPERATURE & AGITATION RATE	TEMPERATURE & AGITATION RATE
CONTROL ACTIVATOR	SERVO ON POTENTIOMETER	DIGITAL SELECTOR POWER SWITCH
RESPONSE OF CONTROL	SLOW in TEMPERATURE	VERY SLOW in TEMPERATURE
Microcontroller	Arduino Uno	Arduino Mega

Figure 39 Scale-up measurement planning model

In Figure 39 we described the same control parameter and catalyst, for different aspects as process time, control activator and response of control from laboratory scale to scale up for pilot plant. To control the design parameters of scale-up model, we propose certain several aspects as available in pilot plant at SBRC as listed physical design has conversion from laboratory scale that apparatus of esterification process consists of heater and magnetic strirrer, condensation tube, sensor and microcontroller Arduino to execute the control of RTO.

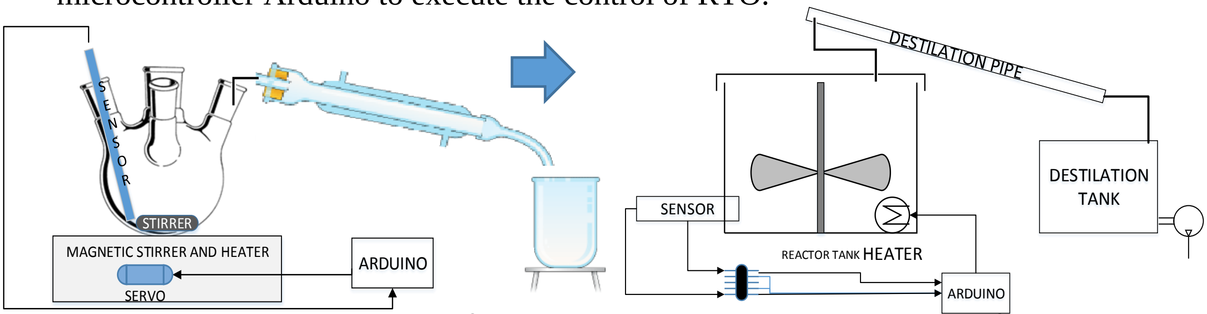


Figure 40 Proposed diagram of physical scale-up conversion apparatus for pilot plant

In Figure 40 we have conversion diagram of apparatus in identically for scale-up system. In this conversion we defined a reactor with heater and stirrer in pilot plant was represented the magnetic stirrer and heater in laboratory scale. For another apparatus such as condensation tube and beaker glass was converted to condensation pipe and distillation tank as detailed. The sensors in proposed system has three types of wavelength that connected to Arduino. This proposed system has discussed completely in Chapter 4 and Chapter 5 especially for optimization in esterification of oleic acid with glycerol.

The physical design for scaling up with controlled the temperature and stirrer speed as optimization has discussed in Chapter 6 to meet up the requirement of scale up which has difference from laboratory scale such as volume, stirrer speed, power and control of heater

StateFlow simulation

To simulate the control parameter that was related to process parameter in scale up, we used simulator software Stateflow® (Mathworks 2014). Stateflow® is an environment for modeling and simulating combinatorial and sequential decision logic based on state machines and flow charts. Stateflow lets us to combine graphical and tabular representations, including state transition diagrams, flow charts, state transition tables, and truth tables, to model how your system reacts to events, time-based conditions, and external input signals. The Stateflow includes state machine animation and static and run-time checks for testing design consistency and completeness before implementation.

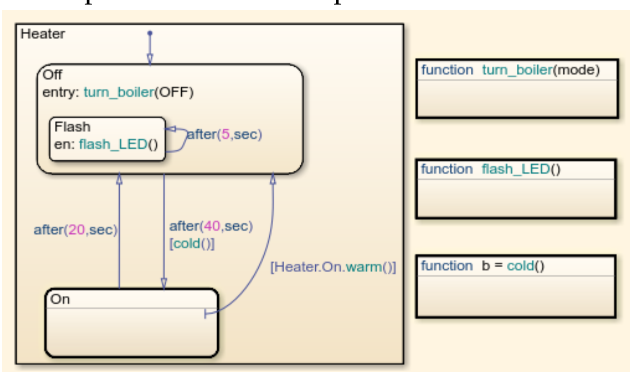


Figure 41 Stateflow model for controlling the reactor heater in pilot scale

In Figure 41, with Stateflow, we have designed logic for adaptive control in esterification process in heater block diagram as an on/off function that supported with three state of control.

Physical Scale-up Control

In this chapter, a new system for scaling up control with real-time optimization using microcontroller and voltage converter (Ramirez 2001) were proposed. As functional requirement to select the three control states of temperature based on our previous research, we design a control system using three units of transformers that were set at different power, that connected to voltage converter block to meet the design requirement for supply the specific energy rate from alternating current phase to the heater in the reactor as physical requirement. In esterification reactor block in this design the three types of mid-IR sensor and temperature sensor named thermocouple were used. The sensor for measuring stirrer speed that not stated in esterification reactor block because we were still not found a sensor that operate able in high temperature and high pressure as condition in the reactor.

As contributions of current achievement in real-time monitoring, real-time adaptive control and real-time optimization using gradient adaptive selection of infrared sensors for glycerol esterification that succeeded to implement as conceptual and physical scale up. In this chapter, we have designed the scale-up models based on functional requirement that using real-time data to ensure feasibility of the inputs based on temperature and mid-IR measurement as the implementation of RTO method by using microcontroller in scale-up.

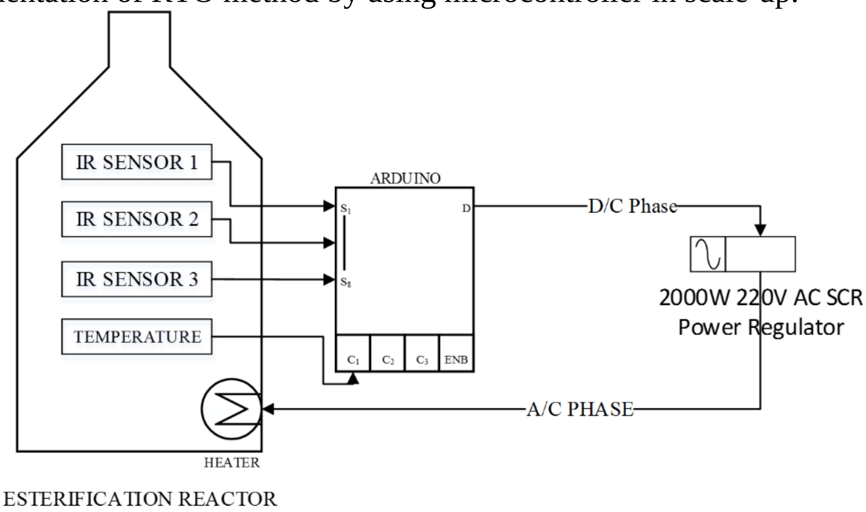


Figure 42 Diagram of proposed physical scale-up control

As shown in Figure 42 A physical design model that the control activator of the heater was constructed with Stateflow, which was supported by data classification Support Vector Machine (SVM) was included in the model by using 2000W 220 V AC SCR Power Regulator which was supported with a microcontroller that connected to a digital input of SCR (Shandilya 2016).

As seen in above description and discussion, our previous achievements hassled to novel effort in constructing higher level of production approach into pilot plant scale. These achievement led to some advantages in case of increasing yield of production, reducing time in esterification process and lower average temperature in reactor, it means increased the efficiency of process.

This scale up level will also be beneficial for industrial purposes by enabling critical point previously unknown or partially understood well in terms of real-time operations and related issue.

Yet there are some disadvantages which had limited contributions to enhance a real completed operations such as these system needed a sensor with high specification and operate able in high temperature and high pressure to put them in the reactor especially for mid-IR sensor and rotation sensor. All these disadvantages are among potential future research to solve.

Conclusion

A formulation the optimization in scale up process using pseudo-homogeneous model was described for the kinetics of oleic acid esterification and a conceptual and physical design for scaling up esterification process was found. By support of design requirement that was simulated the control state with Stateflow and implemented the opportunity and potential of current research achievements such as real time monitoring, real time adaptive control and real time adaptive control with gradient selection. In scale up system in real situation to meet the physical requirement, it is essential to set up the control that interfaced with alternating power phase. A voltage converter to operate the control between DC and AC phase was used. Finally, the design set of control using three types of activator that represented by transformer in the scale-up plan was proposed. From our research that remaining requirements to handle in the future work is to find the sensor for measure the rotation speed of the stirrer. The reactor also should be insulated perfectly from surrounding environment as to increase the efficiency of energy used.

8 CONCLUSION AND RECOMMENDING REMARKS

Conclusion

The modeling of a real time simulation model of the production system of glycerol esterification with self-optimization was developed with discrete simulation and contributes to support the production system of GMO. The analysis of the model was described for lower level process detailed in BPMN 2.0 diagram. The experiments of real-time simulation showed that the evaluation of the proposed parameter such as temperature between at range of 240^0C to 260^0C, and volume at range 25L to 30L for each batch.

The system of real-time monitoring glycerol esterification process was built with mid-IR sensors classifying with SVM contributes to support the identification esterification status in every minute and to get information for the time needed for the esterification process. This esterification status achieved a good performance when classifying into three statuses: initialize, on process and finish as tested with ROC

As a new approach for optimization process esterification of oleic acid with glycerol was developed using method Relief which is feature selection, worked well to select the sensors at specified time intervals and adaptive Pillar k-means data clustering algorithm to determine the set control parameters like temperature and rotational speed of stirrer in real time adaptive control with three sets of cluster control.

The best performance of this research was found in real-time optimization with an adaptive control and sensors selection with gradient measurement supported with classification, was improved as the responsiveness of control increased product yield up to 14% and reduced the duration of process up to 20 minutes, with an effective range of stirrer rotation set between 300rpm to 450rpm and process temperature in reactor between 200°C until 210°C.

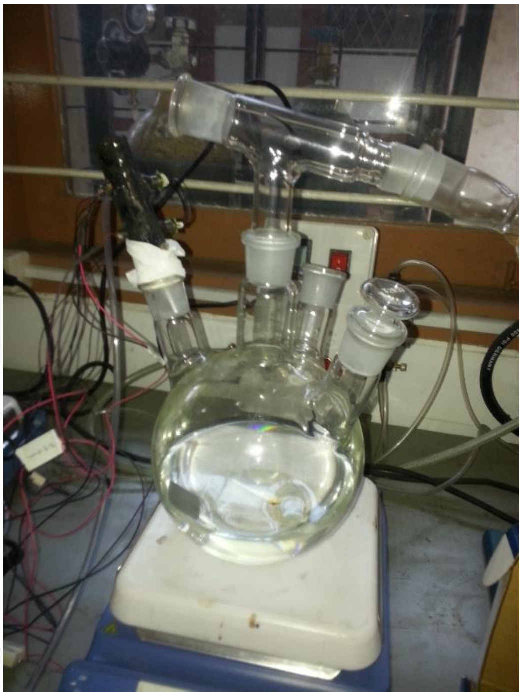
We also proposed the scale-up models based on implementation system using real-time data to ensure feasibility of the inputs based on temperature and mid-IR measurement using a control system with microcontroller.

Recommending remarks

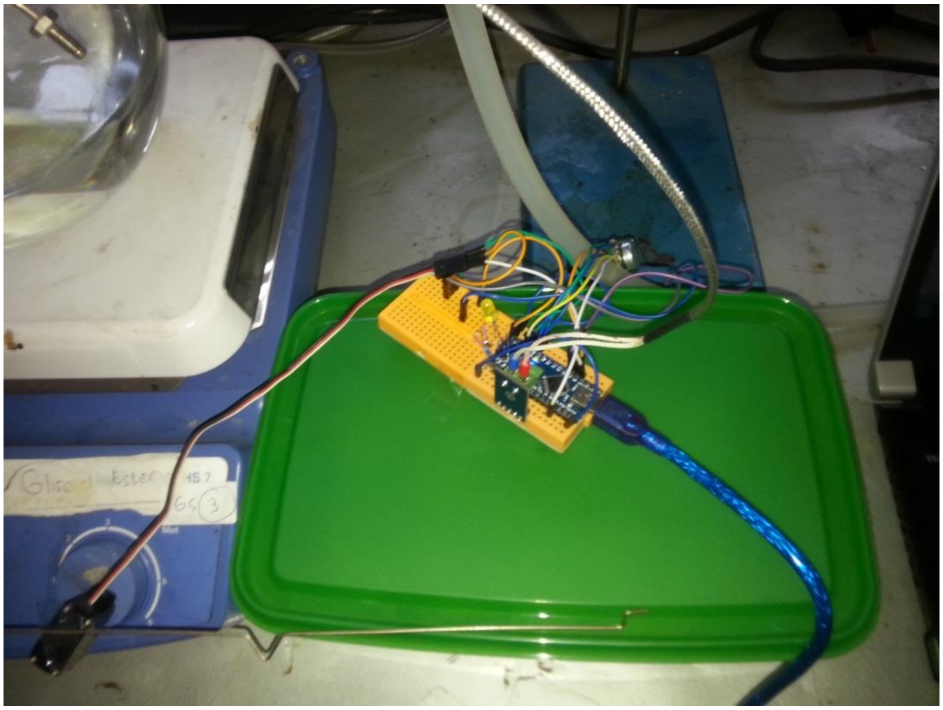
According to the existing disadvantages, in optimization subject, there are still some gaps to fill for the real-time aspect, especially in real time measurement and control for the parameter that is related to optimization, to reduce time span needed for data processing supported with computational intelligence, as well as sensor development that can be operated in several wavelengths by adjusting the voltage and high temperature environment. Future work will be best devoted to the extension of proposed approaches to more complex reaction schemes, e.g. reactions characterized by second-order kinetics. Also, the integration of the proposed model-based within sensors and control device using a universal approximation for the estimation of the model uncertainties supported with another computational data mining is currently the subject of investigation.

APPENDIX

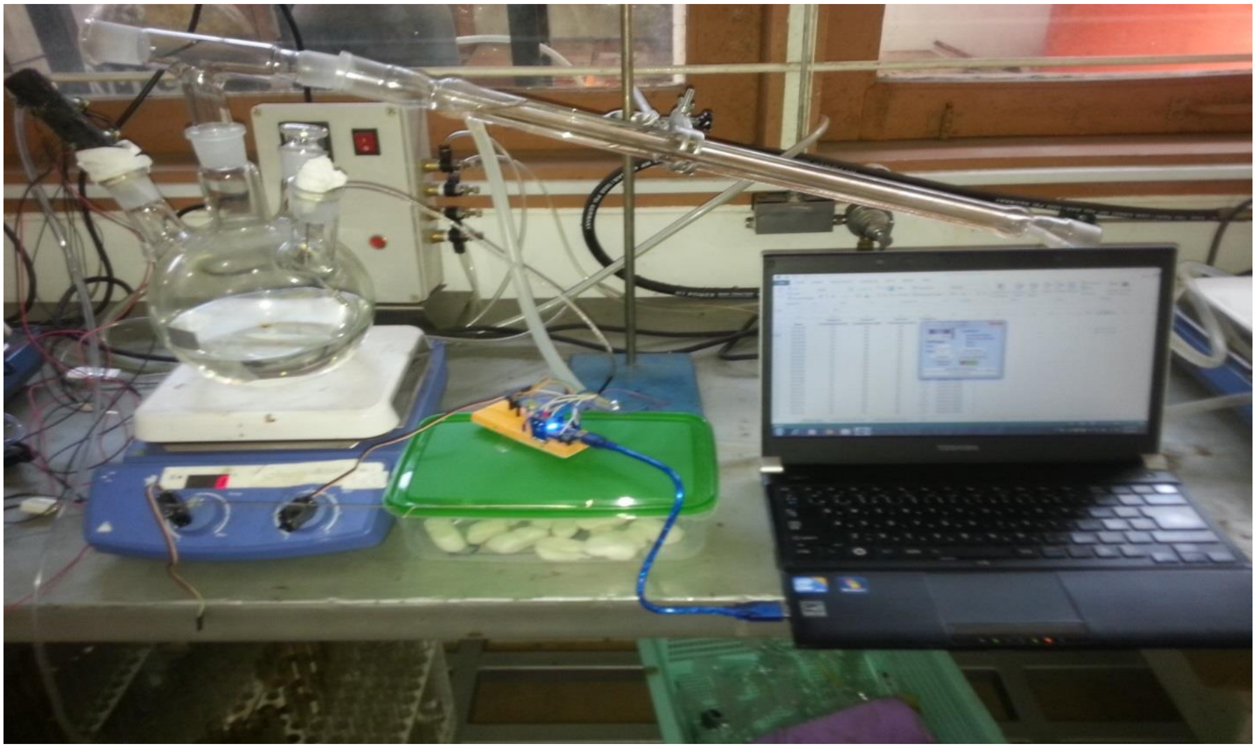
Appendix 1 Photo of reactor with sensor attached



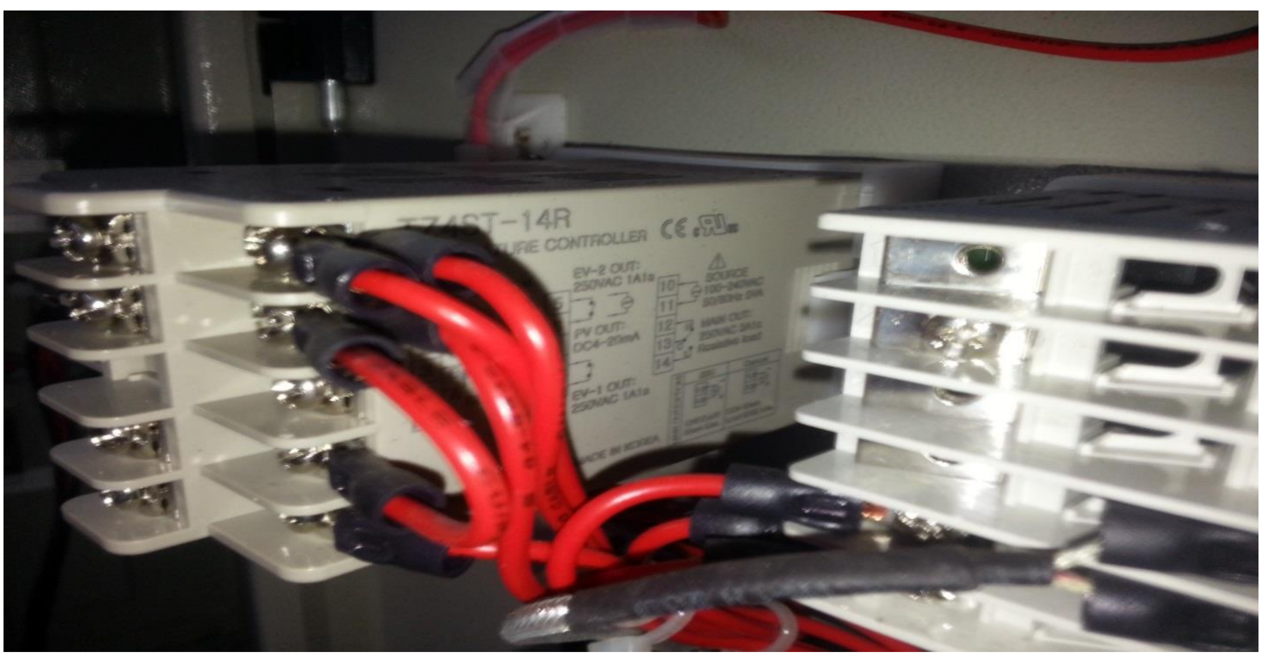
Appendix 2 Photo of Arduino microcontroller



Appendix 3 Photo of real-time optimization system



Appendix 4 Photo of existing reactor scale-up controller



Appendix 5 Datasets of attribute parameter for quality control

Number	Water (%) x_1	Viscosity (poise) x_2	Free Glycerin (%) x_3	pH x_4	Purity (%) x_5	Rel. Density x_6	Quality Pass
1	0.5	0.0548	3	4	72	1.01	NO
2	0.1	0.0543	1	4	63	0.98	NO
3	0.4	0.0549	3	3	88	1	YES
4	0.3	0.0523	1	3	84	0.96	NO
5	0.5	0.0535	3	5	86	0.96	NO
6	0.4	0.0546	3	4	69	0.97	NO
7	0.2	0.0536	3	4	69	0.98	NO
8	0.2	0.052	1	5	69	1.01	NO
9	0.2	0.0549	2	4	90	0.98	YES
10	0.1	0.0529	1	4	70	1.01	NO
11	0.1	0.051	2	4	81	1.02	YES
12	0.4	0.0543	2	3	71	1.01	NO
13	0.2	0.0526	3	4	83	0.96	YES
14	0.2	0.0519	2	4	62	1.02	NO
15	0.1	0.0513	3	5	78	0.96	YES
16	0.4	0.0523	1	5	72	0.95	NO
17	0.2	0.0518	1	5	71	1.02	NO
18	0.4	0.0516	3	5	74	1.01	NO
19	0.3	0.0529	3	5	72	1	NO
20	0.1	0.0547	2	3	82	1.01	YES
21	0.4	0.0531	2	5	61	1	NO
22	0.4	0.054	1	4	82	0.96	YES
23	0.3	0.0523	3	3	74	1.01	NO
24	0.5	0.0547	1	5	83	0.97	YES
25	0.4	0.0538	3	4	84	0.95	YES
26	0.2	0.0523	2	5	61	1	NO
27	0.1	0.0517	3	5	75	0.98	NO
28	0.2	0.0535	2	3	69	0.98	NO
29	0.5	0.0522	2	3	70	0.97	NO
30	0.2	0.0545	3	4	90	1.01	YES

Appendix 6 Sample of data using 3 sensor in real time

Time (Second)	Sensor 3.4µm (Value)	Sensor 5.5µm (Value)	Sensor 7µm (Value)
193	5	40	10
194	8	38	9
195	6	38	5
196	5	35	7
197	7	30	9
198	8	39	8
199	7	34	10
200	5	38	6
201	6	40	7
202	7	40	6
203	5	34	7
204	7	32	8
205	8	33	7
206	8	38	6
207	5	39	6
208	8	34	9
209	9	32	10
210	8	36	9
211	11	37	5
212	8	37	8
213	11	30	7
214	8	34	9
215	8	34	10
216	10	33	6
217	8	32	7
218	8	30	10
219	12	36	6
220	8	35	8
221	12	34	7
222	11	37	7
223	12	31	9
224	9	36	9
225	12	31	5
226	11	33	6

Appendix 7 Sample of data using 2 sensor in real time

Time (Minutes)	Sensor 3.4µm (Transmittance)	Sensor 5.5µm (Transmittance)	Esterification Status
30	8	32	Initialization
31	8	30	Initialization
32	12	36	Initialization
33	8	35	Initialization
34	12	34	Initialization
35	11	37	Initialization
36	12	31	Initialization
37	9	36	Initialization
38	12	31	Initialization
39	11	33	Initialization
60	9	32	Work On Process
61	10	30	Work On Process
62	8	35	Work On Process
63	9	34	Work On Process
64	10	35	Work On Process
65	12	35	Work On Process
66	11	37	Work On Process
67	11	38	Work On Process
68	10	31	Work On Process
69	8	37	Work On Process
70	8	31	Work On Process
71	8	33	Work On Process
72	9	33	Work On Process
73	12	34	Work On Process
74	12	38	Work On Process
75	8	35	Work On Process
76	12	32	Work On Process
77	9	37	Work On Process
78	12	37	Work On Process
79	9	38	Work On Process
80	8	32	Work On Process
81	10	40	Work On Process
82	11	35	Work On Process
83	12	38	Work On Process
84	11	36	Work On Process
85	10	37	Work On Process

Time (Second)	Sensor 3.4µm (Transmittance)	Sensor 5.5µm (Transmittance)	Esterification Status
86	9	37	Work On Process
87	8	36	Work On Process
88	10	38	Work On Process
89	12	30	Work On Process
90	9	39	Work On Process
91	8	40	Work On Process
92	12	31	Work On Process
93	8	39	Work On Process
94	12	37	Work On Process
95	8	40	Work On Process
96	12	36	Work On Process
97	9	37	Work On Process
98	12	36	Work On Process
99	11	30	Work On Process
100	8	30	Work On Process
101	9	34	Work On Process
102	12	36	Work On Process
103	10	37	Work On Process
104	12	33	Work On Process
105	11	39	Work On Process
106	9	31	Work On Process
107	11	38	Work On Process
108	11	31	Finishing
109	8	37	Finishing
110	9	40	Finishing
111	9	39	Finishing
112	8	32	Finishing
113	10	34	Finishing
114	12	38	Finishing
115	10	31	Finishing
116	11	31	Finishing
117	10	35	Finishing
118	9	39	Finishing
119	8	40	Finishing
120	11	39	Finishing
121	9	34	Finishing
122	8	40	Finishing
123	8	36	Finishing

Appendix 8 Arduino code

```
// the setup routine runs once when you press reset:
#include <Servo.h>
Servo myservo;

const int ledPin1 = 13;       // pin that the LED is attached to
const int ledPin2 = 12;
const int threshold1 = 50;   // an arbitrary threshold level that's in the range of the analog
input
const int threshold2 = 60;
// ThermoCouple
int thermo_gnd_pin = 6;
int thermo_vcc_pin = 5;
int thermo_so_pin  = 4;
int thermo_cs_pin  = 3;
int thermo_sck_pin = 2;

void setup() {
  {
  myservo.attach(9);  // attaches the servo on pin 9 to the servo object
}
  pinMode(12, OUTPUT);
  // initialize serial communication at 9600 bits per second:
  Serial.begin(9600);
  Serial.println("CLEARDATA");
  Serial.println("LABEL,Time,Sensor1 ,Sensor2 ,Sensor3,Temperature, State
Temperature,State Agitation");
  pinMode(thermo_vcc_pin, OUTPUT);
  pinMode(thermo_gnd_pin, OUTPUT);
  digitalWrite(thermo_vcc_pin, HIGH);
  digitalWrite(thermo_gnd_pin, LOW);}

// the loop routine runs over and over again forever:
void loop() {
  // read the input on analog pin 0:
  Serial.print(("DATA,TIME, "));
  int sensorValue1 = analogRead(A0);
  int sensorValue2 = analogRead(A1);
  int sensorValue3 = analogRead(A2);
  int sensorValue4 = analogRead(A3);
  int kalibrasi1=sensorValue1*(100.0/1023.0);
  int kalibrasi2=sensorValue2*(100.0/1023.0);
  int kalibrasi3=sensorValue3*(100.0/1023.0);
  int kalibrasi4=sensorValue4*(100.0/1023.0);
```

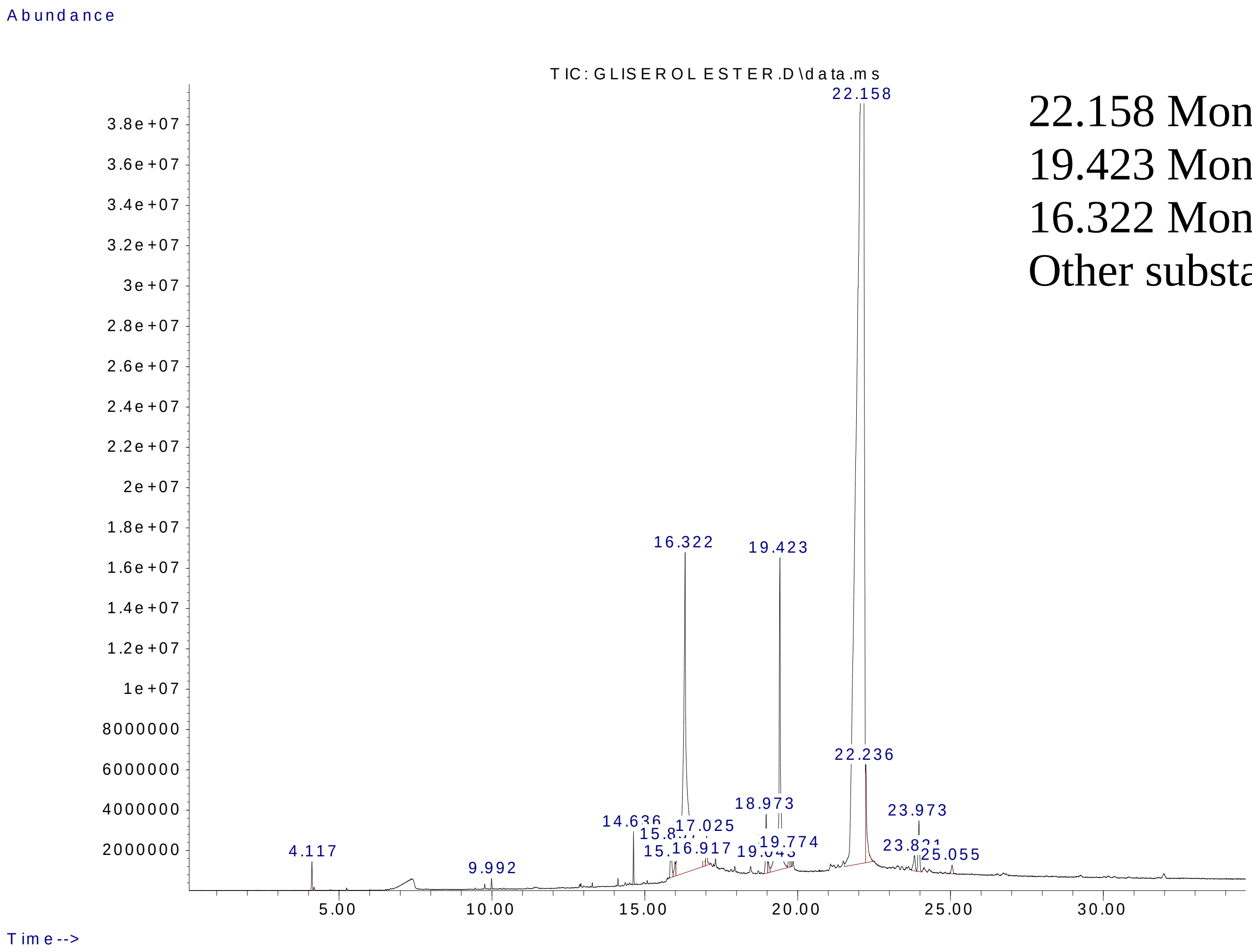
```
  char* state_temperature;
  char* state_agitation;

   if (kalibrasi1 < 30 and kalibrasi3 < 30 ) {
    digitalWrite(ledPin1, HIGH);state_temperature="1";state_agitation="1";
  }
  else {
    digitalWrite(ledPin1,LOW);state_temperature="2";state_agitation="3";
  }
  if (kalibrasi1 > 50 and kalibrasi2>60 ) {
    digitalWrite(ledPin1, LOW);state_temperature="3";state_agitation="3"; }

  // print out the value you read:
  Serial.print(Calibration#1);
  Serial.print(",");
  Serial.print(Calibration#2);
  Serial.print(",");
  Serial.print(Calibration#3);
  Serial.print(",");
  Serial.print(Calibration#4);
  Serial.print(",");
  Serial.print(state_temperature);
  Serial.print(",");
  Serial.print(state_agitation);
  Serial.print(",");
  Serial.println();

      delay(1000);        // delay in between reads for stability
}
```

Appendix 9 GCMS test for esterified glycerol with temperature 210^{0}C-90 minutes process time



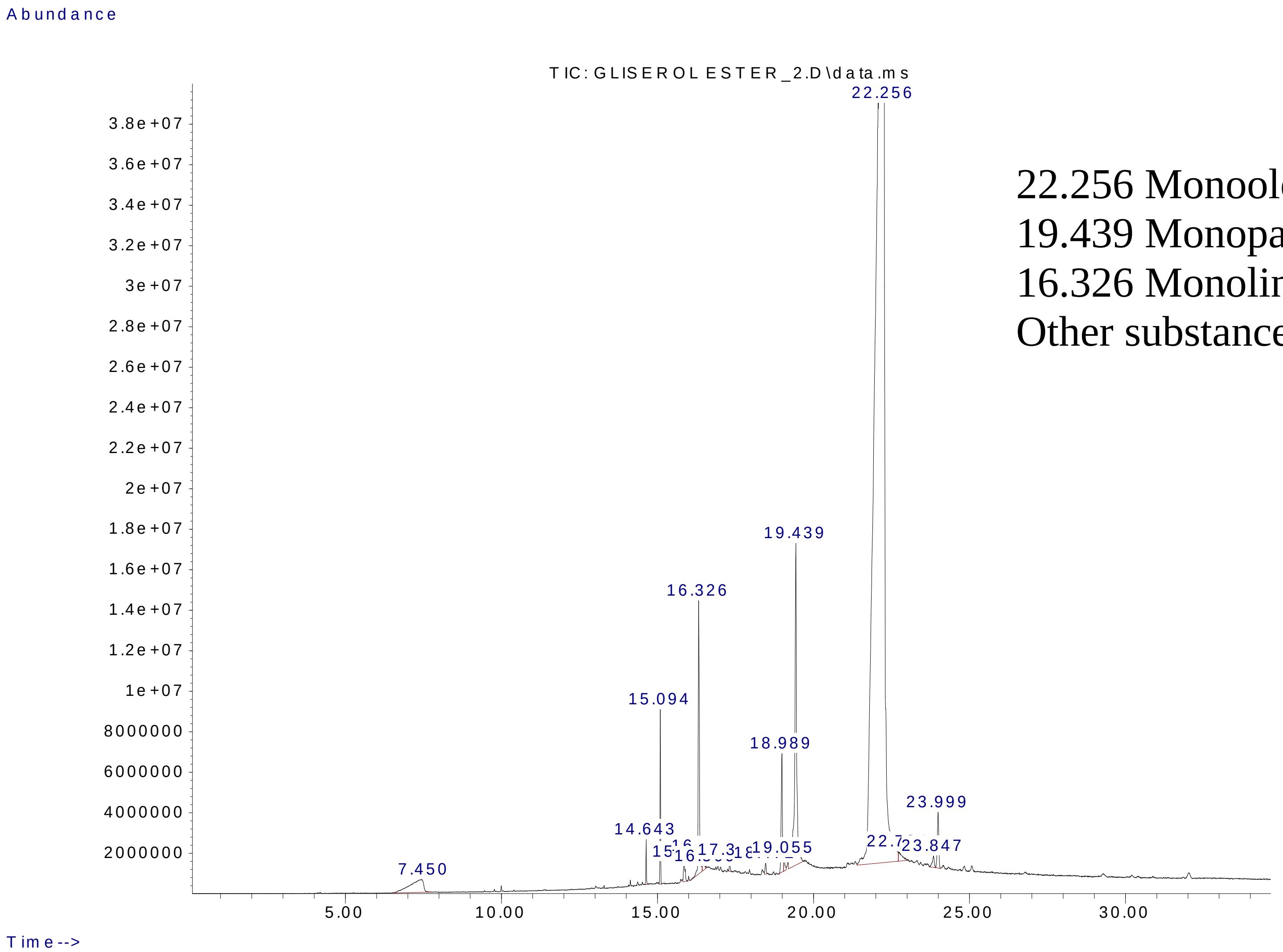
22.158 Monoolein 73.75%

19.423 Monopalmitin 11%

16.322 Monolinolein 12%

Other substance 3%

Appendix 10 GCMS test for esterified glycerol with temperature 210^{0}C-120 minutes process time



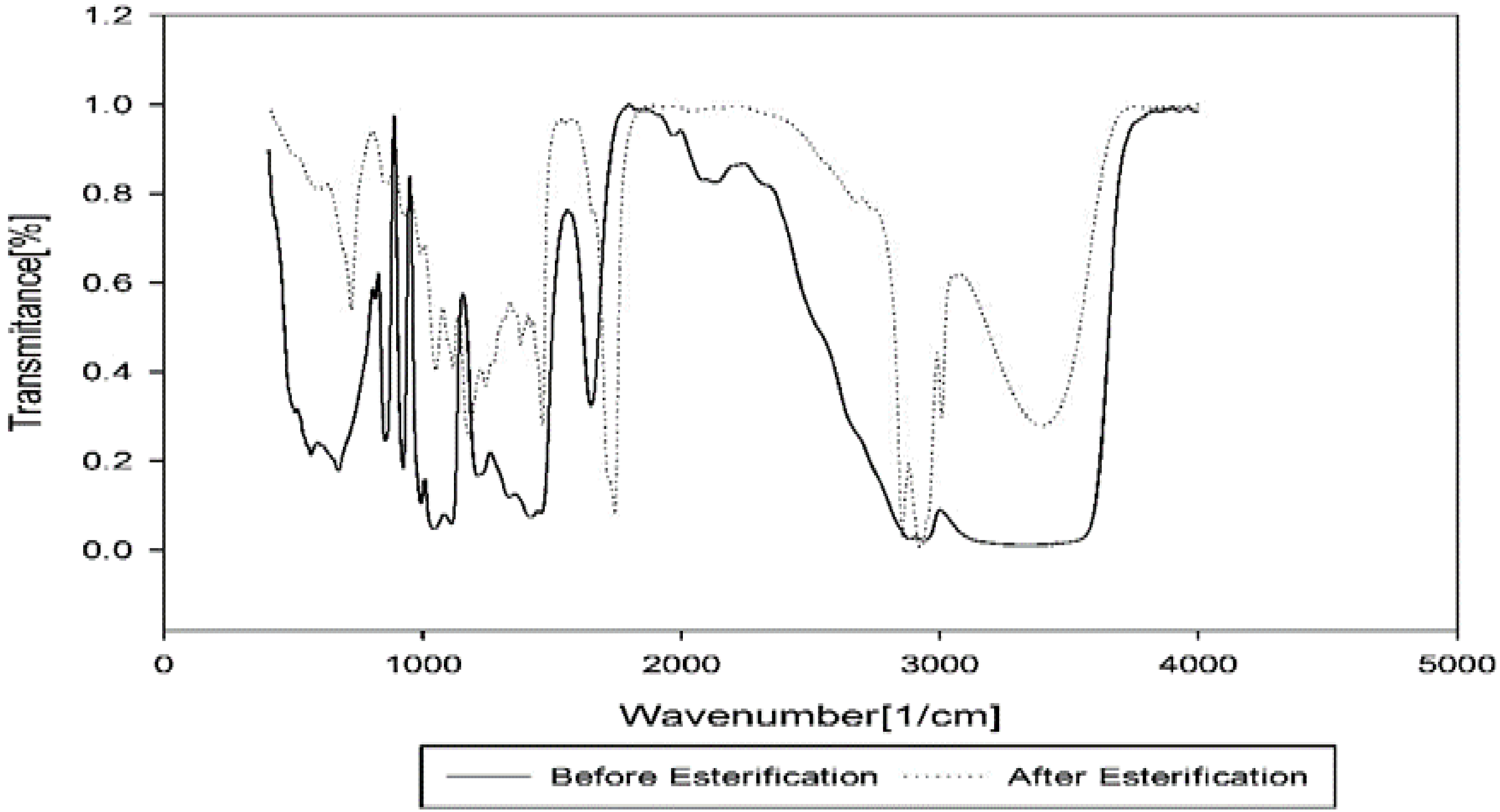
22.256 Monoolein 89.75%

19.439 Monopalmitin 5%

16.326 Monolinolein 3%

Other substance 2%

Appendix 11 FTIR test for non esterified and esterified glycerol with temperature 210^{0}C-120 minutes process time



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