ABUAD Journal of Engineering Research and Development (AJERD)
Volume 1, Issue 1, 49-60

# Modelling, Simulation and Control of a Reactive Distillation Process for Biodiesel Production

Abdulwahab GIWA[1,*], Michael Ayomide OGUNWARE[1]

[1]*Chemical and Petroleum Engineering Department, College of Engineering, Afe Babalola University, KM. 8.5, Afe Babalola Way, Ado-Ekiti, Ekiti State, Nigeria*

agiwa@abuad.edu.ng/michaelogunware7130@gmail.com

[*]*Corresponding Author:* agiwa@abuad.edu.ng

*Date of First Submission:* 29/09/2017
*Date Accepted:* 31/10/2017

***Abstract:*** *In this work, the production of biodiesel via reactive distillation process has been modelled and simulated with the aid of ChemCAD for both steady state and dynamics. Also, the control of the process has been carried out using MATLAB/Simulink. In order to achieve the aim of the work, dynamics data showing the response of biodiesel mole fraction in the column bottoms (controlled variable) to a change in reboiler duty (manipulated variable) and reflux ratio (selected disturbance variable) were extracted from the ChemCAD dynamic simulation of the developed process model and used to obtain the first-order-plus-dead-time transfer function relation between biodiesel mole fraction in the column bottoms, reboiler duty and reflux ratio with the aid of MATLAB. The open loop simulation was done by applying steps to the input variables (reboiler duty and reflux ratio). Furthermore, the set-point tracking and disturbance rejection control of the system were carried out using a PID controller tuned with Zeigler- Nichols, Cohen-Coon and trial-and-error techniques. It was observed that the controller parameters obtained by Zeigler-Nichols and Cohen-Coon tuning were not able to achieve the set-point tracking control of the system appropriately, and this necessitated the use of trial-and-error technique to obtain the controller parameters used to handle the system in the desired manner for set-point tracking of maintaining the mole fraction of biodiesel at 0.7. Nonetheless, Zeigler-Nichols and Cohen-Coon techniques were sufficient to successfully tune the controller to carry out the disturbance rejection of the process. However, it was observed that the performance of Cohen-Coon tuning technique was better than that of Zeigler-Nichols tuning technique in the disturbance rejection control simulation because it had lower Integral Square Error and lower Integral Absolute Error values. It has, thus, been discovered that biodiesel could be produced in high purity via reactive distillation process, and the system could be efficiently handled to behave as desired using PID control system.*

**Keywords**: Fatty acid methyl ester, reactive distillation, PID control, set-point tracking, disturbance rejection, MATLAB, ChemCAD.

## 1. INTRODUCTION

Due to an increased demand of energy by the world population and the non-renewability of crude oil, the development of renewable energy generation techniques for future has gained great importance over the century [1]. One of these renewable energy types has been identified to be biodiesel.

Apart from being renewable, biodiesel is a non-toxic, biodegradable substitute for diesel produced from crude oil. Generally, it is produced through transesterification of vegetable oils and animal fat by short chained aliphatic alcohols. Commercially, the production of biodiesel from vegetable oils and fats still have various drawbacks. Both batch and continuous processes utilize almost 100% excess alcohol than the stoichiometric molar requirement (3:1) in order to drive the transesterification reaction to completion and produce maximum amount of biodiesel per unit consumption of oil [2]. At the end of the process, unreacted alcohol must be recovered by a separate distillation column for alcohol recovery. This increases capital as well as operating cost. Therefore, there is the need to develop alternative means for commercial production of biodiesel which minimizes cost without reducing the yield and quality of biodiesel produced. Reactive distillation is one of such means.

Reactive distillation combines separation and reaction into a single vessel to minimize operation and equipment costs [2-23]. In this process, the products formed are removed as soon as they are formed. This characteristic makes it possible to overcome the equilibrium thermodynamics of a reaction, reaching high conversion and selectivity. Thus, it is particularly effective for reversible reactions such as the transesterification of vegetable oil and fats to biodiesel [24]. However, the combination of reaction and separation into a single unit that resulted in many complexities of the process has made the dynamics and control of this process a challenge to Process Engineers.

Dynamics in Chemical Engineering is the study of how process variables vary with time in response to some input types. As all real-life process variables vary with time, and biodiesel production parameter being one of them, it is important to study the dynamics of the biodiesel production process. Furthermore, Process Control is defined as external intervention needed to guarantee the satisfaction of

operational requirements such as safety, production specifications, environmental regulations, operational constraints, and economics [25] of a process. Since the structure of biodiesel reactive distillation process is complex, because of the need to maximize mass and energy raw materials, there is the need to develop a suitable control system for the process.

Some research studies have already been carried out on the subject matter (modelling, simulation and control) of this work. Among them, Santander et al. [26] employed response surface methodology and Aspen Plus to simulate the production of biodiesel using castor oil in a reactive distillation column in order to find the best conditions for producing large amount of fatty acid esters. Also, Simasatitkul et al. [27] used Aspen HYSYS to investigate the production of biodiesel from the transesterification reaction between soybean oil and methanol in a reactive distillation column and analysed the effects of some operating and design parameters on biodiesel production. Furthermore, Samakpong et al. [28] used Aspen PLUS to simulate a design of esterification of rubber seed oil and methanol catalyzed by sulphuric acid using reactive distillation column, and they were able to obtain the optimum values of the process operating parameters such as molar distillate rate and molar reflux ratio. Orifici et al. [29] used Aspen-HYSYS to simulate and optimize the process of biodiesel production from the transesterification reaction of crude palm oil with methanol. Chavdar and Egveni (2014) also carried out a ChemCAD simulation of biodiesel production by transesterification of vegetable oil, the two final target products-biodiesel and glycerol, were obtained with purity values of 98% and 99%, respectively. Giwa et al. [30] used proportional-integral-derivative approach to the control of a reactive distillation process used for biodiesel production as a servo system. Giwa et al. [31] carried out the proportional-integral-derivate control of a reactive distillation process used for producing biodiesel as a regulatory system. The application of model predictive control to the development of a renewable energy type via reactive distillation process was carried out by Giwa et al. [32]. Furthermore, Giwa et al. [33] applied the toolbox of MATLAB to work on the control of a reactive distillation process using model predictive control approach, and the production of biodiesel which was taken as the case study of the work.

It can be noticed from the literature review carried out that most of the researches reported were based on production of biodiesel using esterification process. Studies on the control of biodiesel production via transesterification processes are limited. Therefore, this research is aimed at providing an outlook at the dynamics of biodiesel production by transesterification reactive distillation process as well as developing a control system for the process. The aim of the work was achieved via the following objectives: developing and simulating the ChemCAD steady-state model of the process, converting the steady-state model into a dynamic type to generate dynamic data, using the generated dynamic data to develop the process transfer functions with the aid of MATLAB, using the transfer function model of the process to obtain the tuning parameters of a PID controller, and applying the PID controller to make the mole fraction of the biodiesel be at the desired set-point value.

## 2. METHODOLOGY

### 2.1 Model Development and Steady-State Simulation

The sodium hydroxide catalysed transesterification reaction mechanism used in the reactive distillation process modelling involving triolein (the reacting triglyceride) and methanol (alcohol) to produce methyl oleate (biodiesel) and glycerol as the by-product is shown in Equations 1-3.

$$C_{57}H_{104}O_6 + CH_3OH \leftrightarrow C_{39}H_{72}O_5 + C_{19}H_{36}O_2 \quad (1)$$

$$C_{39}H_{72}O_5 + CH_3OH \leftrightarrow C_{21}H_{40}O_4 + C_{19}H_{36}O_2 \quad (2)$$

$$C_{21}H_{40}O_4 + CH_3OH \leftrightarrow C_3H_8O_3 + C_{19}H_{36}O_2 \quad (3)$$

The process was modelled and simulated using ChemCAD process simulator [34] by following the steps outlined below:

1. **Component Selection:** The chemical components involved in the process were chosen from the ChemCAD database, and they were:
    - Triolein
    - Diolein
    - Monoolein
    - Methanol
    - Methyl oleate
    - Glycerol
    - Sodium hydroxide
2. **Thermodynamic Package Selection:** UNIFAC, the thermodynamic package suggested by the ChemCAD simulator based on the components chosen, was adopted and used for the simulation.
3. **Flowsheet Development:** Two process feed streams and two product streams were added to the flowsheet from the piping and flow palette. A Simultaneous Correction Distillation System (SCDS) column type was also added from the separators palette. The feed and product streams were connected to the column appropriately by using the stream connector from the piping and flow palette. The developed ChemCAD model of the process is shown in Figure 1.

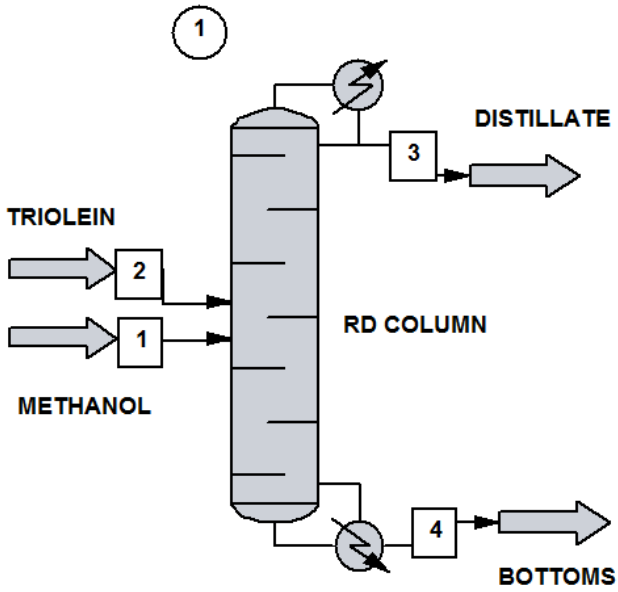


Figure 1: ChemCAD model of the reactive distillation process used for methyl oleate production

4. **Feed Stream Specification:** The conditions of the feed streams were specified, and the specified parameters were as given in Table 1 and 2 for streams 1 and 2 respectively.

Table 1: Operating parameters for stream 1

| Stream Name | METHANOL |
|---|---|
| Temperature (°C) | 25 |
| Pressure (atm) | 1 |
| Total Flow (kmol/hr) | 60 |
| Methanol (mol%) | 100 |

Table 2: Operating parameters for stream 2

| Stream Name | TRIOLEIN |
|---|---|
| Temperature (°C) | 25 |
| Pressure (atm) | 1 |
| Total Flow (kmol/hr) | 15 |
| Triolein (wt%) | 99.5 |
| Sodium Hydroxide (wt%) | 0.5 |

5. **Equipment Specification:** The conditions of the SCDS column were specified using the parameters given in Table 3.

Table 3: Parameter data for the SCDS column

| Column Name | RD COLUMN |
|---|---|
| Condenser type | Total |
| Number of stages | 30 |
| Feed stage for stream 1 | 19 |
| Feed stage for stream 2 | 8 |
| Reflux ratio | 2 |
| Reboiler duty (kJ/sec) | 25 |

6. **Reaction Specification:** The general data used for modelling the reaction of the process are given in Table 4.

Table 4: General reaction data

| | |
|---|---|
| Number of liquid reactions | 6 |
| Number of vapour reactions | 0 |
| Molar flow unit | kmol |
| Activation Energy | Cal |
| Volume unit | Litres |
| Time unit | Minutes |
| Temperature unit | Kelvin |

The transesterification reaction occurring within the column was modelled using 6 kinetic rate expressions, as shown in Equations 4-9, for the forward and backwards reactions given in Equations 1-3.

$$r_1 = k_1 C_{TO} C_{MOH} \quad (4)$$

$$r_2 = k_2 C_{DO} C_{MOE} \quad (5)$$

$$r_3 = k_3 C_{DO} C_{MOH} \quad (6)$$

$$r_4 = k_4 C_{MO} C_{MOE} \quad (7)$$

$$r_5 = k_5 C_{MO} C_{MOH} \quad (8)$$

$$r_6 = k_6 C_G C_{MOE} \quad (9)$$

where TO, DO, MO, MOH, MOE, and G denote triolein, diolein, monoolein, methanol, methyl oleate and glycerol respectively. The rate constant is given as in Equation (10), and the kinetic data used were obtained from the work of Agarwal et *al.* [1] as given in Table 5.

$$k = k^0 \times e^{-\frac{E}{RT}} \quad (10)$$

Table 5: Kinetic data for the transesterification of tri-olein using methanol

| | |
|---|---|
| $k_1^0$ (mol$^{-1}$ litre min$^{-1}$) | $1.469 \times 10^8$ |
| $E_1$ (cal/kmol) | 14040 |
| $k_2^0$ (mol$^{-1}$ litre min$^{-1}$) | 105100 |
| $E_2$ (cal/kmol) | 10739 |
| $k_3^0$ (mol$^{-1}$ litre min$^{-1}$) | $1.19 \times 10^{10}$ |
| $E_3$ (cal/kmol) | 16049 |
| $k_4^0$ (mol$^{-1}$ litre min$^{-1}$) | $1.725 \times 10^8$ |
| $E_4$ (cal/kmol) | 13907 |
| $k_5^0$ (mol$^{-1}$ litre min$^{-1}$) | 24940 |
| $E_5$ (cal/kmol) | 7173 |
| $k_6^0$ (mol$^{-1}$ litre min$^{-1}$) | 627700 |
| $E_6$ (cal/kmol) | 10997 |

Source: Agarwal *et al.* [1]

After developing the model of the process using ChemCAD, it was run using the 'Run All' icon on the ChemCAD flowsheet ribbon until convergence.

### 2.2 Process Dynamics Simulation

The following steps were followed to simulate the dynamics of reactive distillation process used for methyl oleate production:

1. **Conversion of steady-state model to dynamics model:** Using the developed converged steady state reactive distillation model, from the 'Convergence Parameters' sub-menu, the dynamics option was selected from the 'Steady state/Dynamics' drop down menu.
2. **Dynamics simulation:** The dynamics simulation was carried out using two run time steps. The 'Select streams from flowsheet' option was checked from the 'Record streams' sub-menu.
    - First run time step: A time of 1.5 min with a 0.1 min interval was used for the first run time step. The dynamic simulation was run from initial steady state in this case.
    - Second run time step: A time of 15 min with a 0.1 min interval was used for the second run time step. The reboiler duty and reflux ratio of the column were changed to 5 kJ/s and 1 respectively in this case. The dynamic simulation was then run from the current state.
3. **Dynamics data extraction:** The dynamics data obtained were the mole fraction of methyl oleate in the column BOTTOMS stream (stream 4). The run time plot for the mole fraction of methyl oleate in stream 4 was obtained using the 'Plot Dyn Streams' icon on the ChemCAD flowsheet ribbon. From the plot, the dynamics data were extracted to an excel worksheet by clicking the 'Data to Excel CSV file' option from the 'Chart' drop down menu.

### 2.3 Process Transfer Function Generation

The process model used in this work was formulated by developing the transfer function relations between methyl oleate mole fraction (output variable) and reboiler duty and reflux ratio (input variables) using the data generated from the developed ChemCAD model. A first-order-plus-dead-time (FOPDT) transfer function model (Equation 11) was chosen as the model type.

$$x_{biod}(s) = \frac{K_{pp}e^{(-T_{dp}s)}}{\tau_{pp}s+1}Q(s) + \frac{K_{pd}e^{(-T_{dd}s)}}{\tau_{pd}s+1}R(s) \qquad (11)$$

In Equation (11), $x_{biod,}$ is the methyl oleate mole fraction, $Q$ is the column reboiler duty, and $R$ is the column reflux ratio. The process model formulation was done with the aid of a MATLAB [35] script file.

### 2.4 Simulink Modelling and Open-Loop Simulation of the Process

After obtaining the transfer function of the process, the model of which was as given in Equation 11, the reactive distillation system was modelled in Simulink environment contained in MATLAB by combining the different appropriate blocks required. The developed Simulink model for the open loop case of the system was as shown in Figure 2. Thereafter, the open loop dynamics of the process was studied by applying unit step changes to the process manipulated and disturbance variables, and running the Simulink open loop model using codes written in a MATLAB script file.

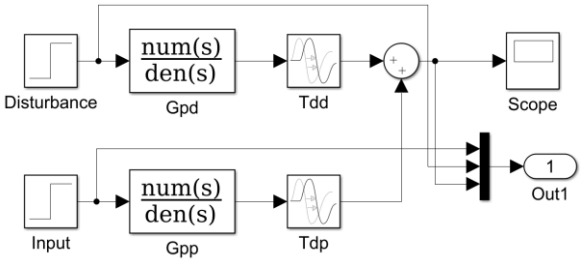


Figure 2: Open loop Simulink model of the process

### 2.5 Proportional-Integral-Derivative Controller Tuning

A Proportional-Integral-Derivative (PID) control approach was adopted for the control of the process studied in this work in order to obtain a reasonably high mole fraction of the output variable (methyl oleate mole fraction). With the transfer function of the controller given as in Equation (12), Zeigler-Nichols and Cohen-Coon tuning techniques were applied using the expression in Table 6 with the aid of a MATLAB script file.

$$G_c(s) = K_c\left(1+\frac{1}{\tau_I s}+\tau_D s\right) \qquad (12)$$

Table 6: Tuning parameter expressions for proportional-integral-derivative (PID) control system

| Controller Parameter | Zeigler-Nichols technique | Cohen-Coon technique |
|---|---|---|
| $K_c$ | $\frac{K_u}{1.7}$ | $\frac{1}{K}\frac{\tau_{pp}}{\tau_{pd}}\left(\frac{4}{3}+\frac{T_{dp}}{4\tau_{pp}}\right)$ |
| $\tau_I$ | $\frac{P_u}{2}$ | $T_{dp}\left(\frac{32+6T_{dp}/\tau_{pp}}{13+8T_{dp}/\tau_{pp}}\right)$ |
| $\tau_D$ | $\frac{P_u}{8}$ | $T_{dp}\left(\frac{4}{11+2T_{dp}/\tau_{pp}}\right)$ |

Source: Stephanopoulos [25]

### 2.6 Procedures for Simulink modelling and Closed loop simulation of the Process

After developing the Simulink model of the process and studying its open loop behaviours, its closed loop simulation was also carried out using the developed closed loop Simulink model shown in Figure 3a and using the value of the tuning parameters obtained by applying the expressions given in Table 6 and trial-and-error method with the model given in Figure 3b. The closed loop dynamics of the process was studied for both set-point tracking and disturbance rejection using codes written in a MATLAB script file.

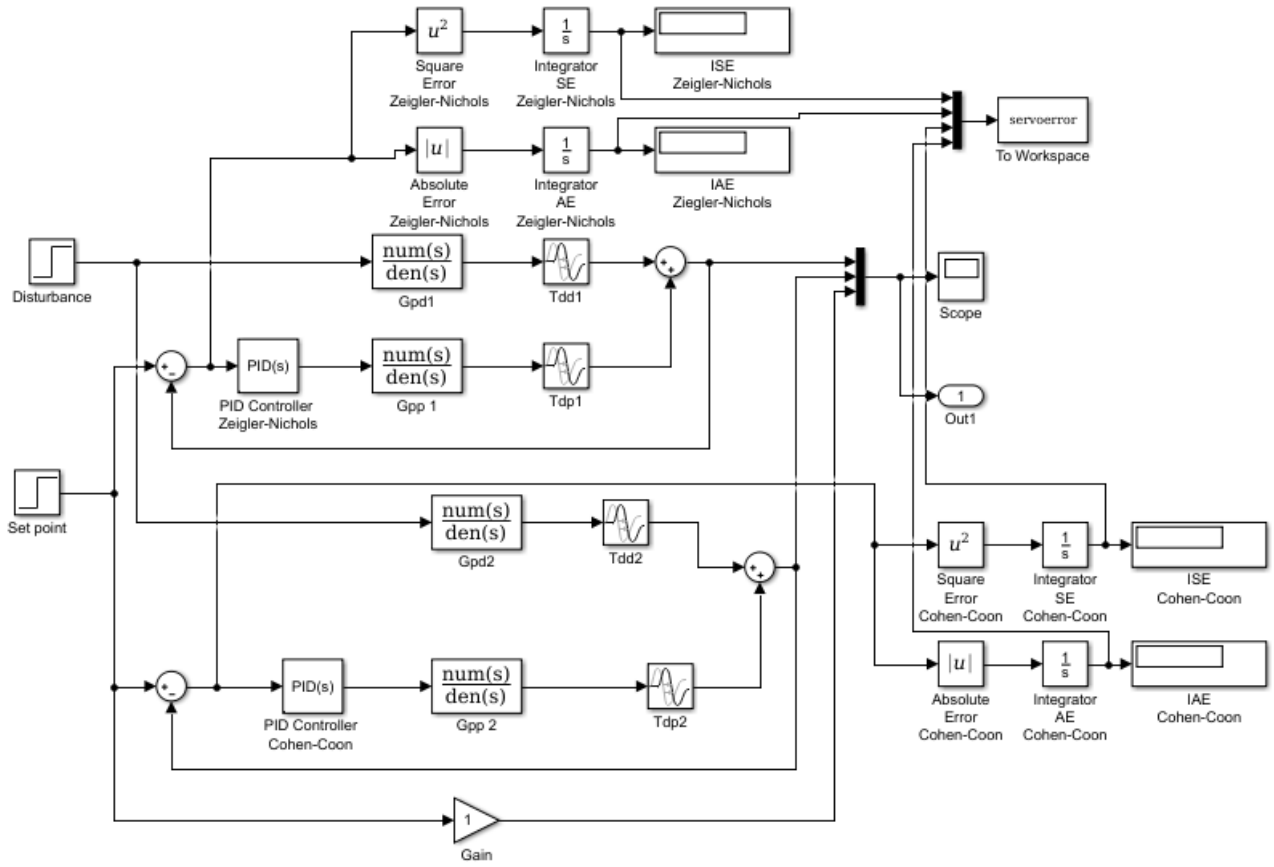


Figure 3a: Closed loop Simulink model of the process with PID controllers tuned with Ziegler-Nichols and Cohen-Coon method

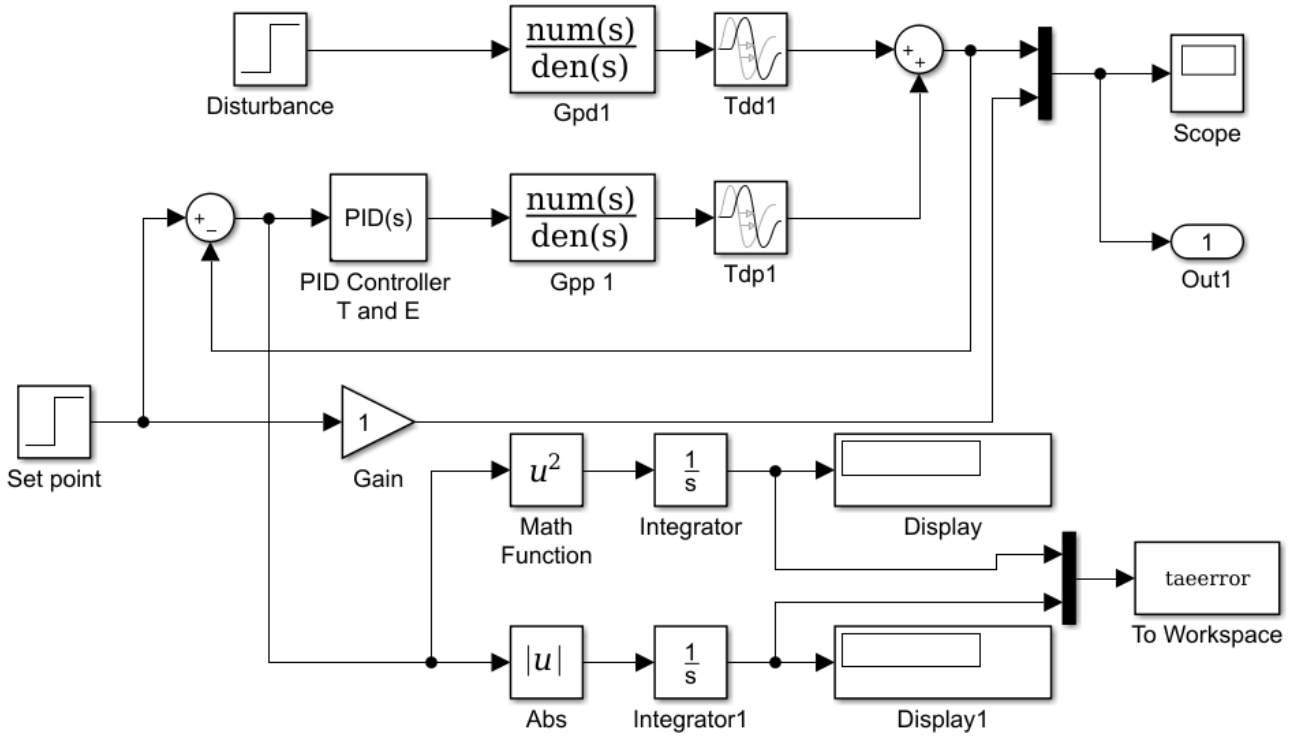


Figure 3b: Closed loop Simulink model of the process with PID controller tuned with trial-and-error method

## 3. RESULTS AND DISCUSSION

### 3.1 Steady State Results

The results obtained from the steady-state simulation of the developed ChemCAD process model of the reactive distillation process for methyl oleate production were as given in Tables 7-9. Shown in Table 7 are the calculated parameters of the column. It can be seen from the results that the developed ChemCAD model of the process was working properly because, for instance, the condenser duty was obtained to be a negative value, which was an indication that heat was being given out by the condenser, and this was found to follow one of the general principles of a condenser as obtainable in the literature.

Table 7: Column calculated parameters

| Parameter | Value |
|---|---|
| Condenser duty (kJ/sec) | -722.138 |
| Reflux rate (mol/sec) | 13.649 |

Table 8 shows the properties of column distillate stream. Among all the components involved in the process, only unreacted methanol was found to be in the distillate stream, in pure form, because of its volatility, which is also generally related to its boiling point. This further showed that there was effective separation of methanol to the column distillate stream, which was fed into the column in excess of triolein in the ratio of 6:1 as opposed to the stoichiometric ratio of 3:1 based on the proposed reaction mechanism.

Table 8: Column distillate stream properties

| Parameter | Value |
|---|---|
| Temperature (°C) | 64.48 |
| Pressure (atm) | 1 |
| Vapour fraction | 0.6 |
| Enthalpy (kJ/sec) | -1463.24 |
| Distillate rate (mol/sec) | 6.82 |
| Methanol mole fraction | 1 |

The properties of the bottom stream from which the product of the process was obtained is also given in Table 9. Observing the table, as evidenced from the molar composition of the stream, it was discovered that there was production of methyl oleate (biodiesel), glycerol, monoolein, and diolein. In support of this, a reduction in triolein and methanol initial molar flow was an indication of a reaction taking place within the column. This has, thus, confirmed that biodiesel (methyl oleate) could be produced using the reactive distillation approach based on the proposed reaction mechanism and column operating parameters.

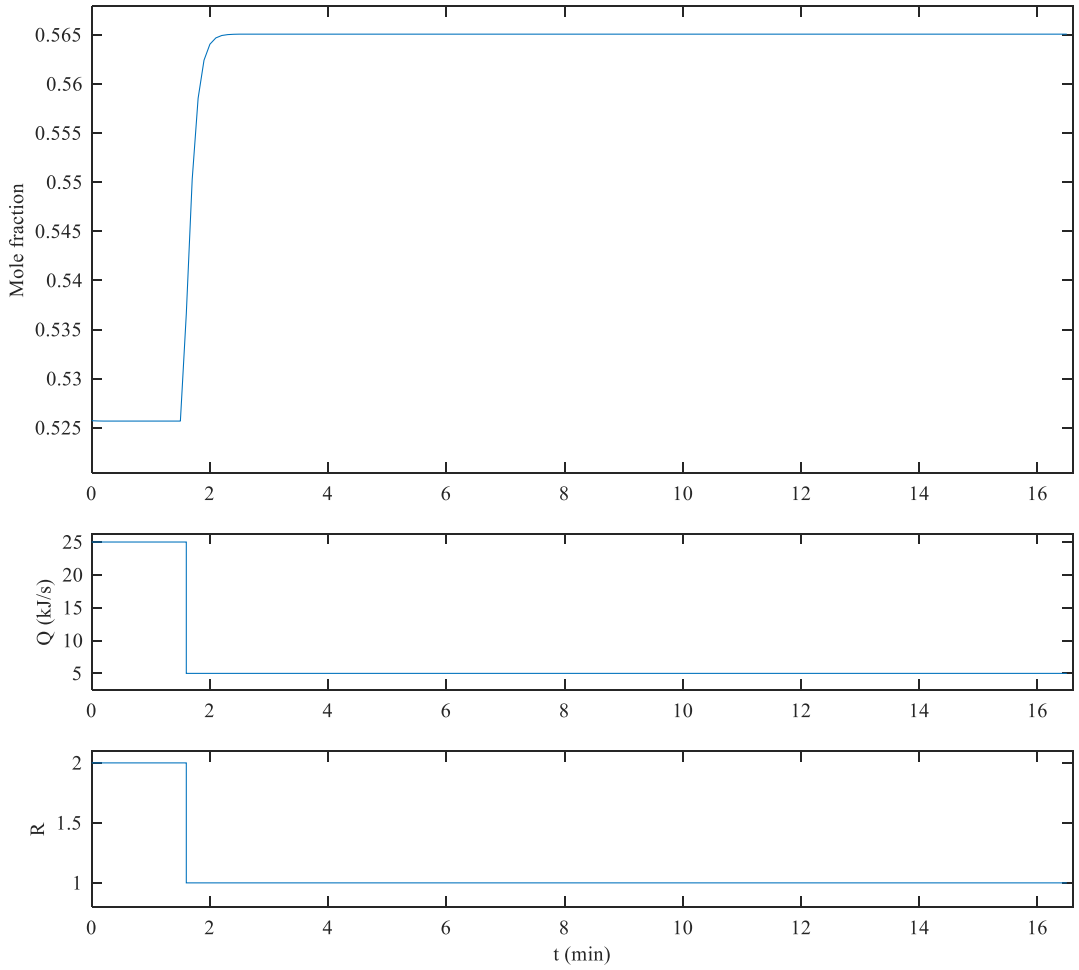


Figure 4: Dynamics response of methyl oleate production system

Table 9: Column bottom stream properties

| Parameter | Value |
|---|---|
| Temperature (°C) | 116.112 |
| Pressure (atm) | 1 |
| Vapour fraction | 0 |
| Enthalpy (kJ/sec) | 10441.4 |
| Bottoms rate (mol/sec) | 14 |
| Triolein mole fraction | 0 |
| Methanol mole fraction | 0.1768 |
| Methyl oleate mole fraction | 0.5257 |
| Glycerol mole fraction | 0.0015 |
| Diolein mole fraction | 0.011 |
| Monoolein mole fraction | 0.2551 |
| Sodium hydroxide mole fraction | 0.0298 |

Also noticed from the results given in Tables 8 and 9 was that there was no triolein in both the column distillate and bottom streams. This was seen to be an indication that the triolein involved in the reaction was completely converted within the column due to the excess of methanol that was fed into the column and the continuous removal of products within the column that was in favour of the forward reactions of the mechanism. It was also noticed from the results given in Table 9 that the column bottom stream contained various products in different fractions including the desired product (methyl oleate). Thus, it was deemed beneficial to improve the biodiesel mole fraction in the column bottom stream. However, before going into the control of the process, there was the need to have some knowledge about the time-dependent behaviour, that is, the dynamics, of the process.

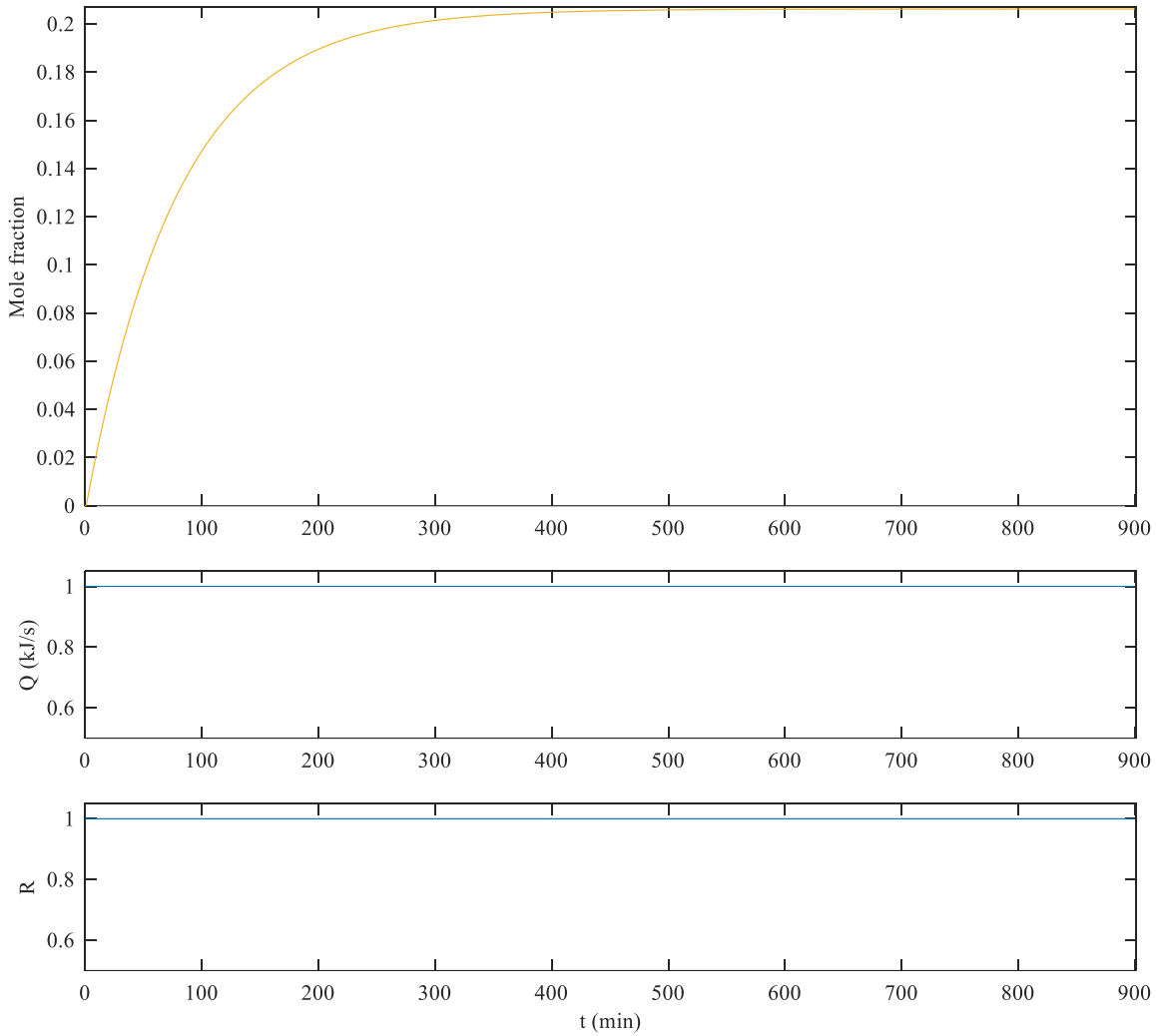


Figure 5: Open loop dynamic response of the process to a unit step change in each of reboiler duty and reflux ratio

### 3.2 Process Dynamics Results

Figure 4 shows the response obtained from the dynamic simulation of the developed ChemCAD model of the reactive distillation process of methyl oleate production in form of a plot of methyl oleate mole fraction, which was the output variable, and the input variables (the reboiler duty and the reflux ratio) against time as obtained for two run time steps: 0-1.5 min for verification of the steady state of the process and 1.5-16.5 min for a unit decrease in column reflux ratio and a 20-unit decrease in the reboiler duty applied at 1.5 min of the process. It can be seen from Figure 4 that biodiesel mole fraction was significantly affected by the change in the input variables of the process.

For the changes applied to the reboiler duty and reflux ratio, methyl oleate mole fraction was found to increase from its initial steady state value of 0.525 to another steady state value of 0.565. It was therefore seen that the reboiler duty and reflux ratio were appropriate input variables of the process for producing methyl oleate using reactive distillation process. The change that occurred in the output variable was observed to be due to the combination of the effects caused by changes in reflux ratio and reboiler duty, and they include:

- increase in separation of components across the column due to increase in reflux ratio, posing an indirect resistance to mass transfer between methanol and triolein and, thus, affecting the occurring reactions and causing a reduction in methyl oleate produced, and
- increase in monoolein and diolein recycle from the column bottoms stream due to an increase in reboiler duty and, thus, leading to more methyl oleate being produced and a higher methyl oleate mole fraction in the column bottom stream.

### 3.3 Open-loop Simulation Results

Using developed MATLAB script codes, the process overall transfer function relating methyl oleate mole fraction in the column bottom (controlled/output variable) to the column reboiler duty (manipulated variable) and the column reflux ratio (selected disturbance variable) was obtained to be as given in Equation (12),

$$x_{biod}(s) = \frac{0.112e^{(-0.8245s)}}{81.1609s+1}Q(s) + \frac{0.0942e^{(-0.8696s)}}{76.9263s+1}R(s) \quad (12)$$

where Q(s) is reboiler duty and R(s) is reflux ratio in Laplace transform.

The transfer function model showed that the process had small static gains but large time constants because of the complexities involved in it as a result of the combination of reaction and separation in a single unit.

The open loop response of the process controlled output variable was obtained by applying a unit step change to both the process manipulated and disturbance variables, at time $t = 0$, using the developed transfer function, with the aid of Simulink, and the response given by the process is shown in Figure 5 that was observed to be like that of a first order system. As can be seen from the figure, the system could attain a steady state value of biodiesel mole fraction of approximately 0.2 at about 400 min, and this was found to be in conformity with the obtained steady state gains of the process. It was also noticed from Figure 5 that the change in reboiler duty and reflux ratio could make the system have stable dynamics. However, the need to have a product with high purity earlier than the settling time given by this open loop response necessitated control of the system.

### 3.4 Controller Tuning Results

With the aid of MATLAB script codes, using the PID control approach and the process transfer function relating methyl oleate mole fraction to reboiler duty, the obtained controller parameters for both Zeigler-Nichols and Cohen-Coon tuning methods were estimated to be as shown in Table 10, and those values were further used for the control of the system for both set-point tracking and disturbance rejection control problems of the process.

Table 10: Controller parameters using Zeigler-Nichols and Cohen-Coon Tuning

| Controller Parameter | Zeigler-Nichols Tuning | Cohen-Coon Tuning |
|---|---|---|
| $K_c$ | 812.1163 | 1174.1 |
| $\tau_i$ | 1.6490 | 2.0208 |
| $\tau_d$ | 0.4123 | 0.2993 |

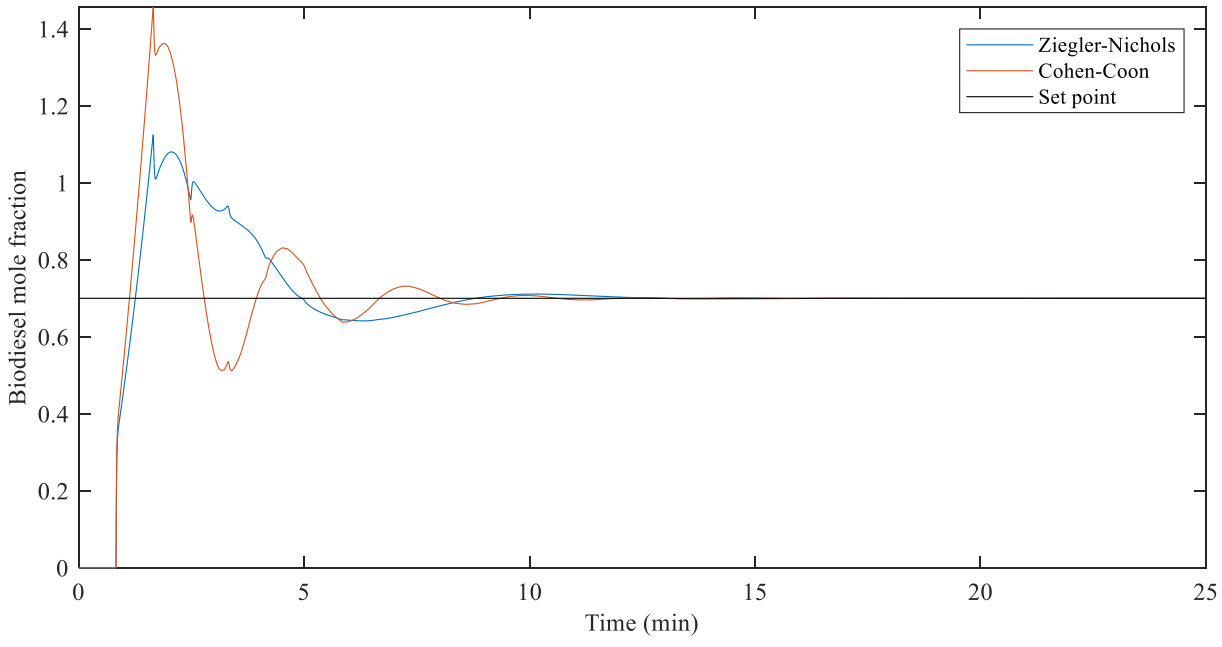

Figure 6: Closed loop response of the process to a 0.7 step change in the set point using Zeigler-Nichols and Cohen-Coon methods

### 3.5 Closed Loop Response for Set-Point Tracking

The closed loop response of the process to a step change of 0.7 in biodiesel mole fraction and no change in reflux ratio (disturbance variable) using the obtained controller parameters, of both Zeigler-Nichols and Cohen-Coon tuning methods, with the aid of Simulink that was run via a MATLAB script, was obtained to be as shown in Figure 6. It can be seen from Figure 6 that the PID control using both tuning techniques could reach the desired set point of 0.7, but the closed-loop response of the process controlled variable was found to be unrealistic because the responses were found to exceed the maximum obtainable mole fraction of 1. This showed the need for using another method to tune the PID controller of the process. By trial and error, different PID controller parameters were obtained as shown in Table 11.

Table 11: Controller parameters obtained using trial-and-error method

| **Parameter** | **Value** |
|---|---|
| $K_c$ | 105 |
| $\tau_I$ | 15 |
| $\tau_D$ | 0.5 |

Using the controller parameters obtained with the trial-and-error method to run the closed-loop model of the system using a MATLAB script, it was discovered that a set point of 0.7 biodiesel mole fraction was achieved within 90 min without any overshoot above the maximum obtainable mole fraction, as can be seen in Figure 7. It was, thus, found out that a biodiesel purity of 0.7 mole fraction could be achieved in the reactive distillation column as a bottom product using the PID controller tuning parameters obtained with trial-and-error method.

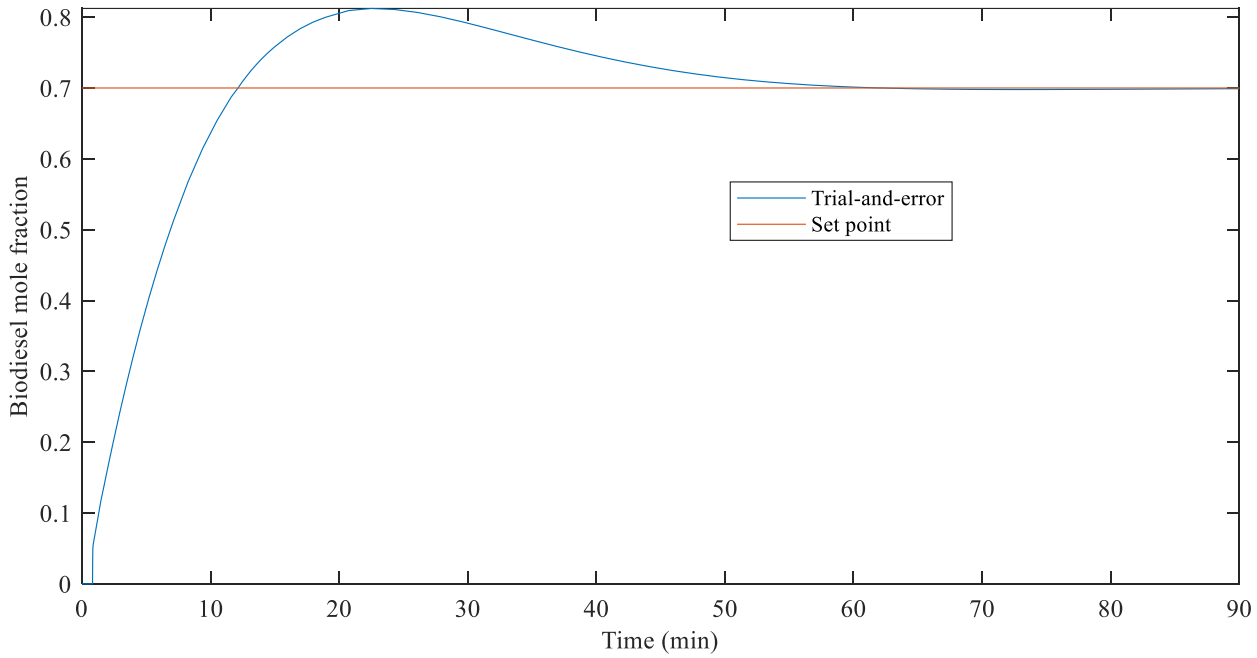


Figure 7: Closed loop response of the process to a set point of 0.7 using trial-and-error tuning technique

### 3.6 Closed loop Response for Disturbance Rejection

The closed loop response of the process model to a 3-unit step change in reflux ratio (disturbance variable) only using the obtained controller parameters calculated via a similar approach as that of set point tracking, was as shown in Figure 8.

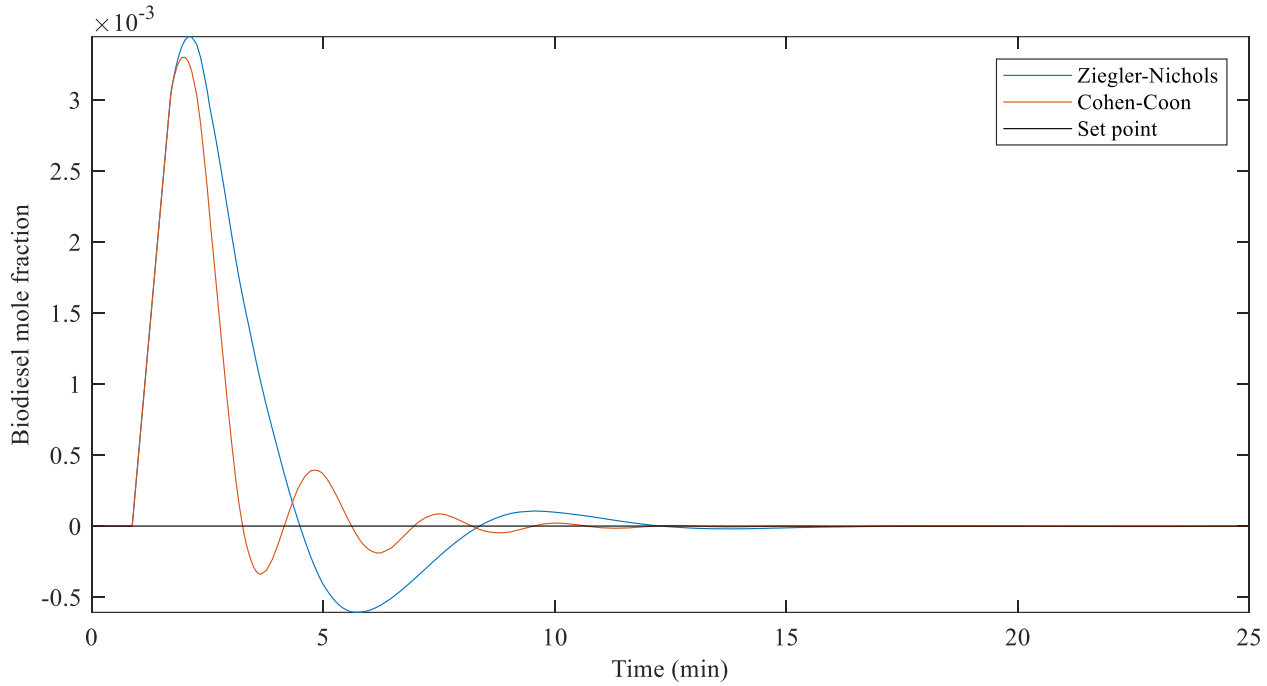


Figure 8: Closed response of the process model to a 3-unit step change in reflux ratio

It can be seen from Figure 8 that the PID control approaches using both Zeigler-Nichols and Cohen-Coon controller parameters were able to make the system return to its initial value of zero mole fraction, which was required for a regulatory control by suppressing any effect that the step change in the reflux ratio (disturbance variable) could have on the process output. For both Zeigler-Nichols- and Cohen-Coon-tuned PID control systems, the process output was observed to return to the set point within approximately 15 min. The process response to both Zeigler-Nichols and Cohen-Coon controller parameters were seen to have similar overshoots, that of Cohen-Coon method was found to decay faster, though with more oscillations.

In order to clearly determine the controller parameters that were able to control the process more efficiently and effectively, the performance criteria values, Integral Square Error (ISE) and Integral Absolute Error (IAE), for both the Zeigler-Nichols and Cohen-Coon-tuned PID control systems were estimated, and the values were as given in Table 12.

Table 12: Performance criteria values

| **Performance Criterion** | **Zeigler-Nichols Tuning** | **Cohen-Coon Tuning** |
|---|---|---|
| ISE | 1.7109e-05 | 1.151e-05 |
| IAE | 0.0082658 | 0.0053962 |

According to the results given in Table 12, Cohen-Coon technique proved to give a better regulatory control, in this case of reactive distillation for biodiesel production, with a lower ISE and IAE than other tuning method.

## 4. CONCLUSIONS

After achieving the objectives of this research work, which was on modelling, simulation and control of a reactive distillation process for biodiesel production, and analysing the results obtained, the following findings have been discovered:

1. biodiesel could be produced via reactive distillation process through sodium hydroxide catalysed transesterification reaction of triolein and methanol,
2. changes in column reboiler duty and reflux ratio resulted in stable dynamic response of biodiesel mole fraction,
3. the PID control of biodiesel mole fraction with reboiler duty as the manipulated variable and reflux ratio as the disturbance variable has been achieved,
4. Cohen-Coon and Zeigler-Nichols tuning methods were found to give controller tuning parameters for set-point tracking control of the process that resulted in unrealistic biodiesel mole fraction,
5. the set-point tracking control of a set-point step change of 0.7 in biodiesel mole fraction was achieved using $K_c$ = 105, $\tau_I$ = 15, $\tau_D$ = 0.5 as the controller parameters obtained by trial-and-error method, and
6. Cohen-Coon tuning technique provided a better controller performance than Zeigler-Nichols tuning method for regulatory control of biodiesel mole fraction during production.

## ACKNOWLEDGEMENT

Special thanks go to Aare Afe Babalola, LL.B, FFPA, FNIALS, FCIArb, LL.D, SAN, OFR, CON – The Founder and President, and the Management of Afe Babalola University, Ado-Ekiti, Ekiti State, Nigeria for providing the necessary materials that enabled the accomplishment of this research work.

## NOMENCLATURES

| | |
|---|---|
| C | Concentration |
| DO | diolein |
| E | Activation energy |
| G | Glycerol |
| Gc(s) | Controller transfer function |
| IAE | Integral Absolute Error |
| ISE | Integral Squared Error |
| $k_1$, $k_2$, $k_3$, $k_4$, $k_5$, $k_6$ | Reaction rates constants |
| $K_c$ | Proportional gain of the controller |
| $K_p$, $K_{pd}$ | Static gains of the process |
| $K_u$ | Ultimate gain |
| MO | monoolein |
| MOE | methyl oleate |
| MOH | methanol |
| PID | Proportional-Integral-Derivative |
| $P_u$ | Ultimate period (min/cycle) |
| Q | Reboiler duty |
| R | Universal gas constant |
| $r_1$, $r_2$, $r_3$, $r_4$, $r_5$, $r_6$ | Reaction rates |
| T | Temperature |
| $T_{dp}$, $T_{dd}$ | Dead times of the process |
| TO | triolein |
| $x_{biod}$ | Bottom biodiesel mole fraction |
| $\tau_D$ | Derivative time of the controller |
| $\tau_I$ | Integral time of the controller |
| $\tau_{pp}$, $\tau_{pd}$ | Time constants of the process |

## REFERENCES

[1] Agarwal, M., Singh, K., Chaurasia, S.P. (2012). Simulation and Sensitivity Analysis for Biodiesel Production in a Reactive Distillation Column, *Polish Journal of Chemical Technology*, 14(3), 59-65.

[2] Kiss, A.A., Dimian, A.C., Rothenberg, G. (2008). Biodiesel by Catalytic Reactive Distillation Powered by Metal Oxides, *Energy Fuels*, 22(1), 598-604.

[3] Giwa, A. (2012). Steady-State Modeling of n-Butyl Acetate Transesterification Process Using Aspen Plus: Conventional versus Integrated, *ARPN Journal of Engineering and Applied Sciences*, 7(12), 1555-1564.

[4] Giwa, A. (2013). Methyl Acetate Reactive Distillation Process Modeling, Simulation and Optimization Using Aspen Plus, *ARPN Journal of Engineering and Applied Sciences*, 8(5), 386-392.

[5] Giwa, S.O., Giwa, A., Hapoglu, H. (2013). Investigating the Effects of Some Parameters on Hydrogen Sulphide Stripping Column Using Aspen HYSYS, *ARPN Journal of Engineering and Applied Sciences*, 8(5), 338-347.

[6] Giwa, A., Giwa, S.O., Hapoglu, H. (2013). Adaptive Neuro-Fuzzy Inference Systems (ANFIS) Modeling of Reactive Distillation Process, *ARPN Journal of Engineering and Applied Sciences*, 8(7), 473-479.

[7] Giwa, A., Owolabi, J.O., Giwa, S.O. (2017). System Identification and IMC-based PID Control of a Reactive Distillation Process: A Case Study of n-Butyl Acetate Production, *International Journal of Engineering Research in Africa*, 31, 104-119.

[8] Giwa, A., Giwa, S.O., Adeyi, A.A. (2015). Dynamics and Servo Control of Biodiesel Purity from a Reactive Distillation Process, *International Journal of Scientific & Engineering Research*, 6(8), 146-156.

[9] Giwa, A., Bello, A., Giwa, S.O. (2014). Performance Analyses of Fatty acids in Reactive Distillation Process for Biodiesel Production, *International Journal of Scientific & Engineering Research*, 5(12), 529-540.

[10] Giwa, A., Bello, A., Giwa, S.O. (2015). Artificial Neural Network Modeling of a Reactive Distillation Process for Biodiesel Production, *International Journal of Scientific & Engineering Research*, 6(1), 1175- 1191.

[11] Giwa, A., Karacan, S. (2012). Simulation and Optimization of Ethyl Acetate Reactive Packed Distillation Process Using Aspen HYSYS, *The Online Journal of Science and Technology*, 2(2), 57-63.

[12] Giwa, A., Karacan, S. (2012). Nonlinear Black-Box Modeling of a Reactive Distillation Process, *International Journal of Engineering Research & Technology*, 1(7), 548-557.

[13] Giwa, A., Karacan, S. (2012). Decoupling Control of a Reactive Distillation Process using Tyreus-Luyben Technique, *ARPN Journal of Engineering and Applied Sciences*, 7(10), 1263-1272.

[14] Giwa, A., Giwa, S.O. (2013). Isopropyl Myristate Production Process Optimization Using Response Surface Methodology and MATLAB, *International Journal of Engineering Research & Technology*, 2(1), 853-862.

[15] Giwa, A., Giwa, S.O., Bayram, İ., and Karacan, S. (2013). Simulations and Economic Analyses of Ethyl Acetate Productions by Conventional and Reactive Distillation Processes Using Aspen Plus, *International Journal of Engineering Research & Technology*, 2(8), 594-605.

[16] Giwa, A. (2014). Solving the Dynamic Models of Reactive Packed Distillation Process Using Difference Formula Approaches, *ARPN Journal of Engineering and Applied Sciences*, 9(2), 98-108.

[17] Giwa, A., Giwa, S.O. (2016). Modelling and Simulation of a Reactive Distillation Process for Fuel Additive Production, *Journal of Environmental Science, Computer Science and Engineering & Technology, Section C: Engineering & Technology*, 5(1), 63-74.

[18] Giwa, A., Karacan, S. (2012). Modeling and Simulation of a Reactive Packed Distillation Column

Using Delayed Neural Networks, *Chaotic Modeling and Simulation*, 2(1), 101-108.

[19] Giwa, A., Giwa, S.O. (2013). Layer-Recurrent Neural Network Modelling of Reactive Distillation Process, *Chaotic Modeling and Simulation*, 2(4), 647-656.

[20] Giwa, A., Karacan, S. 2012). Development of Dynamic Models for a Reactive Packed Distillation Column, *International Journal of Engineering*, 6(3), 118-128.

[21] Anene, R.C., Giwa, A. (2016). Modelling, Simulation and Sensitivity Analysis of a Fatty Acid Methyl Ester Reactive Distillation Process Using Aspen Plus, *International Journal of Engineering Research in Africa*, 27, 36-50.

[22] Giwa, S.O., Adeyi, A.A., Giwa, A. (2016. Application of Model Predictive Control to Renewable Energy Development via Reactive Distillation Process, *International Journal of Engineering Research in Africa*, 27, 95-110.

[23] Giwa, A., Giwa, S.O. (2012). Optimization of Transesterification Reaction Integrated Distillation Column Using Design Expert and Excel Solver, *International Journal of Advanced Scientific and Technical Research*, 2(6), 423-435.

[24] He, B.B., Singh, A.P., Thompson, J.C. (2006). A Novel Continuous-Flow Reactor Using Reactive Distillation for Biodiesel Production, *Transactions of the ASABE*. 49(1), 107–112.

[25] Stephanopoulos, G. (1984). *Chemical Process Control: An Introduction to Theory and Practice*, PTR Prentice Hall, New Jersey.

[26] Santander, C.M.G., Rueda, S.M.G., Silva, N.D.L., Costa, A.C., Filho, R.M., and Maciel, M.R.W. (2010). Simulation of the Reactive Distillation Process for Biodiesel Production, *20th European Symposium on Computer Aided Process Engineering – ESCAPE20*, 1-6.

[27] Simasatitkul, L., Siricharnsakunchai, P., Patcharavorachot, Y., Assabumrungrat, S., Arpornwichanop, A. (2011). Reactive Distillation for Biodiesel Production from Soybean Oil, *Korean J. Chem. Eng.*, 28(3), 649-655.

[28] Samakpong, M., Tangviroon, P., Svang-Ariyaskul, A. (2012). Process Simulation and Optimization of Biodiesel Production Using Reactive Distillations from Rubber Seed Oil, *Pure and Applied Chemistry International Conference 2012 (PACCON 2012)*, 653-737.

[29] Orifici, L.I., Bahl, C.D., Gely, M.C., Bandoni, A., Pagano, A.M. (2013). Modeling and Simulation of the Biodiesel Production in a Pilot Continuous Reactor, *Asociación Argentina de Mecánica Computacional*, 1451-1461.

[30] Giwa, A., Giwa, S.O., Adeyi, A.A. (2015). Dynamics and Servo Control of Biodiesel Purity from a Reactive Distillation Process, *International Journal of Scientific & Engineering Research*, 6(8), 146-156.

[31] Giwa, S.O., Adeyi, A.A., Giwa, A. (2015). Dynamics and Regulatory Control of Biodiesel Purity from a Reactive Distillation Process, *International Journal of Engineering Research and General Science*, 3(5), 371-381.

[32] Giwa, S.O., Adeyi, A.A., Giwa, A. (2016). Application of Model Predictive Control to Renewable Energy Development via Reactive Distillation Process, *International Journal of Engineering Research in Africa*, 27, 95-110.

[33] Giwa, A., Adeyi, A.A., Giwa, S.O. (2017). Control of a Reactive Distillation Process Using Model Predictive Control Toolbox of MATLAB, *International Journal of Engineering Research in Africa*, 30, 167-180.

[34] Chemstations. (2015). *ChemCAD* 6.5.7.8139, Chemstations Inc, Texas.

[35] MathWorks. (2017). *MATLAB, The Language of Technical Computing*, The MathWorks, Inc., Natick.