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Research Article

Modelling and Simulation of Biodiesel Transesterification Reactor

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A B S T R A C T

The present work deals with studying the dynamic behaviour. The oil reacts with methanol to produce biodiesel. A mathematical model for the biodiesel transesterification reactor is derived based on the mass balance and energy balance of triglyceride (TG), diglyceride (DG), monoglyceride (MG), glyceride (G), alkyl ester (E), and methanol inside the reactor. The simulation of the biodiesel transesterification reactor is solved by using ''MATLAB/ Simulink''. The dynamic model of the open-loop system is implemented by measuring the exit reactor temperature, exit concentration of triglyceride, alkyl ester (biodiesel) and exit concentration of glycerol responses of Biodiesel Transesterification Reactor to step-change disturbances in the feed triglyceride concentration from 1.11 kmol/m^3 to 1.22 kmol/m^3, 1.33 kmol/m^3, 1.44 kmol/m^3, 1.55 kmol/m^3, 1.67 kmol/m^3, 1 kmol/m^3, 0.89 kmol/m^3, 0.79 kmol/m^3,0.67 kmol/m^3 and 0.56 kmol/m^3, feed methanol concentration from 6.66 kmol/m^3 to 7.33 kmol/m^3, 7.99 kmol/m^3, 8.66 kmol/m^3, 9.3 kmol/m^3, 10 kmol/m^3, 5.99 kmol/m^3, 5.33 kmol/m^3, 4.66 kmol/m^3, 3.97 kmol/m^3 and 3.33 kmol/m^3, volumetric flow rate from 27.08 m^3/hr to 30 m^3/hr, 35 m^3/hr, 40 m^3/hr, 45 m^3/hr, 22 m^3/hr, 20 m^3/hr, 18 m^3/hr and 15 m^3/hr and feed temperature from 333 K to 335 K, 338 K, 328 K, 323 K, 318 K and 313 K. The simulation results of the open-loop system showed that the sensitivity index value when the methanol is stepped from 6.66 kmol/m^3 to 7.33 kmol/m^3, 7.99 kmol/m^3, 8.66 kmol/m^3, 9.3 kmol/m^3, 10 kmol/m^3, 5.99 kmol/m^3, 5.33 kmol/m^3, 4.66 kmol/m^3, 3.97 kmol/m^3, and 3.33 kmol/m^3 is higher than from the other input variable. The results of model simulation were compared with the simulation data achieved by Mjalli (2009) to validate the model. The results showed excellent agreement between the simulation model and the simulation data of Mjalli.

Keywords: Biodiesel Transesterification Reactor, Reactor Modeling, Dynamic Behavior, Mathematical Model, Mass Balance, Energy Balance, Triglyceride, Diglyceride, Monoglyceride, Glycerol, Alkyl Ester

Introduction

Invented by Rudolph Diesel in the 1890s, the diesel engine quickly became known worldwide for its durability, efficiency, and high fuel economy. Early tests with vegetable oil as fuel were conducted by Dr. Diesel and the French government, as they explored alternatives to petroleum, especially for agricultural machinery in remote areas. This early interest laid the groundwork for biodiesel. While initial research into converting vegetable oils into fatty acid methyl esters began in Belgium during the 1930s, the commercial biodiesel industry only started to take shape in Europe in the late 1980s.

Biodiesel has become increasingly popular due to its various advantages over traditional petroleum-based diesel fuel. Key benefits of biodiesel include lower emissions, enhanced performance, renewability, and a higher safety profile. Given these benefits, it might be surprising that petrodiesel is still widely used. However, petrodiesel remains the preferred choice for many due to certain advantages, such as its established supply and performance characteristics. On the other hand, biodiesel requires careful storage conditions and may lead to engine deposits, factors that should be considered when choosing between the two fuels.

Biodiesel has attracted much attention from researchers as it is considered to be an alternative source of energy. Also, the negative impact of fossil fuel emissions that create global warming and climate change issues could accelerate the biodiesel commercialisation process. Biodiesel, also called fatty acid methyl ester (FAME) or fatty acid ethyl ester (FAEE), can be produced by different approaches, such as transesterification or esterification reactions (Pasha et al., 2021; Silva et al., 2018). The biodiesel transesterification process can be performed by the reaction of long-chain glyceride-derived oil with alcohol in the presence or absence of a catalyst to produce fatty acid alkyl esters (FAAE) and glycerol (López Zapata et al. 2018). Biodiesel can be produced by batch and continuous reactors. Sakai et al. (Sakai, Kawashima, and Koshikawa 2009) investigated an economic comparison between batch and continuous processes, and they reported that batch processes are more expensive than continuous processes. Fonseca et al. (Pasha et al. 2021) examined that batch processes have higher production than single continuous stirred tank reactors (CSTR), but a series of CSTRs showed higher production than batch reactors. A simplified block diagram for a typical CSTR biodiesel reactor is shown in Figure 1.

Biodiesel can be produced through the transesterification process by reacting one mole of triglyceride (TG) with three moles of alcohol (ROH), as shown in Equation.

$$TG + 3\ ROH \leftrightarrow G + 3\ E \quad (1)$$

The overall reaction is the result of three consecutive reactions, as shown in Equation 2.

$$TG + ROH \leftrightarrow DG + E$$

$$DG + ROH \leftrightarrow MG + E$$

$$MG + ROH \leftrightarrow G + E \quad (2)$$

Where TG, DG, MG, G, and E are triglyceride, diglyceride, monoglyceride, glycerol, and alkyl ester, respectively. The forward reaction is considered to be second order. Soap formation and carboxylic acid formation accompanied biodiesel production as side reactions. To shift the reaction for higher biodiesel production and soap formation, the alcohol-to-triglyceride molar ratio has to be in excess. Therefore, the alcohol concentration becomes constant, and the rate of reaction depends only on the triglyceride concentration. In the meantime, these side reactions have a marginal impact on the overall reaction process and thus will be neglected in the calculations (Mjalli et al. 2009). The dynamic of transesterification can be modelled based on the kinetic rate of the reaction with the aid of mass and energy balance equations done on the system, taking into account the heat of the reaction, change in specific heat, and density (López Zapata et al. 2018).

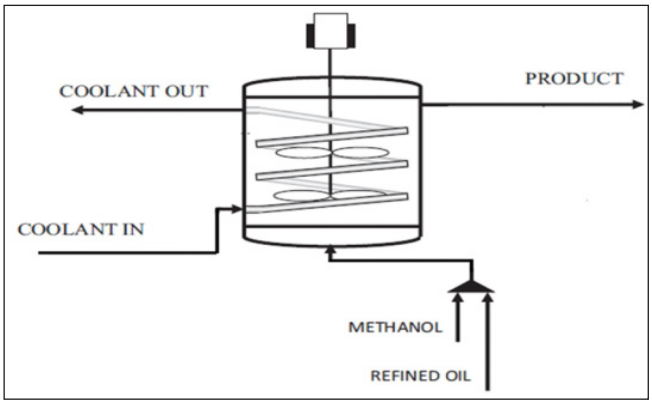

Figure 1.Simplified Schematic of the CSTR Reactor of Biodiesel

Research Background

The purpose of a mathematical model simulation is to predict the process behaviour in a general form to cover both inside and outside the range of experimental data. Also, high flexibilities for both the inlet and outlet streams in the reactor simulation are expected. Furthermore, mathematical models and simulations can assist in optimising the chemical processes and reducing the cost and time of the experiment. To build a mathematical model for any process, at least three main things have to be done: describe the process system comprehensively and accurately, convert the physical and chemical process into mathematical terms, and solve the set of equations and conditions employing the numerical method (Gál and Lakatos, 2004).

To model a transesterification reactor, it's imperative to

understand the reaction kinetic mechanism related to this reaction. Biodiesel transesterification reactors have sets of complex chemical reactions and heat transfer characteristics. Also, the operating conditions of the transesterification reactor are widely ranged and highly intricate. Freedman and his colleagues initiated the search for the chemical kinetics of biodiesel production in the early 1980s. Different types of vegetable oils have been researched for transesterification reactions, such as soybean, sunflower, rapeseed, and safflower seed (Freedman, Mounts). They proposed a kinetic model with only one overall reaction in which one molecule of triglyceride (TG1) reacts with three molecules of alcohol (ROH).

$$TG + 3\ ROH \leftrightarrow G + 3\ E$$

This reaction consists of three steps. The triglyceride (TG) decomposes to diglyceride (DG) and monoglyceride (MG) with the production of glycerol (G) and alkyl ester (E). This is represented as:

$$TG + ROH \leftrightarrow DG + E$$

$$DG + ROH \leftrightarrow MG + E$$

$$MG + ROH \leftrightarrow G + E$$

Mittelbach, Trathnigg, and Wiss (1990) investigated the transesterification kinetics in a methanol-sunflower oil reaction using potassium hydroxide as a catalyst. Their study examined how temperature, catalyst concentration, and the type of vegetable oil affect the reaction process. They observed that the reaction initially forms a two-phase system within the first two minutes, which transitions into a one-phase system for approximately 5 to 10 minutes, before reverting to a two-phase system as glycerin accumulates. This complex behavior impacts the reaction kinetics, which must be carefully considered. Contrary to Freedman's findings, they concluded that triglyceride conversion does not follow second-order kinetics.

In a related study, Noureddini and Zhu (1997) analyzed the transesterification kinetics of soybean oil, adapting Freedman's kinetic model to include the effects of mixing intensity, measured via Reynolds number. They also evaluated temperature's impact on the reaction by determining the Arrhenius parameters for both the standard and a modified Arrhenius equation.

$$k = A\ T^{n}\ \exp(-Ea/RT)$$

where n is an experimentally derived parameter.

Boocock et al. 1998 reported that their data didn't follow the second-order kinetics model. They proposed that the reaction rate declines over time because of inadequate mixing and the reduced effectiveness of the catalyst due to the reduced polarity.

Darnoko and Cheryann (2000) reported the kinetics of palm oil transesterification to produce methyl ester in the presence of KOH as a catalyst in a batch reactor. In the initial stages of the reaction, a pseudosecond-order model was suggested to best fit their data followed by first-order kinetics for the rest of the reaction. Thus, the rate equation for the h reaction is expressed as:

Their reaction rate constants data at different temperatures fitted within 96% accuracy. They also showed that the rate was enhanced as the temperature increased up to 60°C. Then, at temperatures higher than 60°C, there was no reduction in the time to obtain the maximum conversion.

The selected kinetic model can be formulated in terms of a general reaction equation. For any reaction (j) and component (i), the general kinetic model can be written as:

$$rj = kj\ [Ci]^{2}$$

Assuming the j-th reaction rate constant, kj, is related to temperature:

$$kj = Aj\ \exp(-Ea/RT)$$

By plotting ln(kj) against 1/T, the gradient of the straight line is calculated as the j-th reaction activation energy (Eaj).

Romain Richard (Richard, Thiebaud-Roux, and Prat 2013) studied the kinetic modelling for biodiesel transesterification of sunflower oil with ethanol employing continuous microreactors. Their kinetic data were obtained in the first seconds of the reaction. They also identified the chemical reaction kinetics and mass transfer coefficients at 65°C.

Zapata (López Zapata et al. 2018) proposed the design and validation of a mathematical model of the transesterification reaction. They tried to simulate the real case of a batch reactor in designing a dynamic model employing the reaction kinetics, mass, and heat balance.

Adokiye (Adokiye, Gunorubon, and Kenkugile 2020) proposed dynamic models for the inlet and outlet concentration using CSTR for the transesterification of used cook oil with the aid of mass conservation basics. A feedback control was developed with a proportional-integral (PI) controller and a closed-loop model.

Mathematical Modelling of the Biodiesel Transesterification Reactor

The biodiesel transesterification reactor consists of a complex set of chemical reactions and heat transfer characteristics (Mjalli et al. 2009). Thus, it is necessary to understand the complex reaction kinetic mechanism, mass balance, and energy balance equations to obtain the modelling of transesterification reactors.

Investigation regarding biodiesel transesterification kinetics was started at the beginning of the 1980s by Freedman and his co-workers. They proposed only one overall reaction,

which made it limited to use for this reaction. In their proposition, one molecule of triglyceride (TG) reacts with three molecules of alcohol (ROH) to produce glycerol (G) and alkyl ester (E), as shown in Equation 3:

TG + 3 ROH ↔ G + 3 E (3)

In more detail, the reaction in Equation 1 is a result of three consecutive steps as presented in Equation 4:

TG + ROH ↔ DG + E

DG + ROH ↔ MG + E

MG + ROH ↔ G + E (4)

where diglyceride (DG) and monoglyceride (MG) are reaction intermediates.

This model is derived from the mass balance equations for the three-step reaction by assuming the overall order of the forward reaction is second order.

This kinetic model can be expressed in terms of a general reaction equation as represented in Equation 5:

$$r_1=k_1[C_{TG}]^2$$
$$r_2=k_2[C_{DG}]^2$$
$$r_3=k_3[C_{MG}]^2 \quad (5)$$

while (j) and (i) represent any reactions and components, respectively. By assuming the rate constant, k_j, depends on the reaction temperature as shown in Equation 6:

$$k_j = A_j \exp\left(-\frac{E_a}{RT}\right) \quad (6)$$

The activation energy (E_a) can be calculated from the slope of the straight line by plotting ln (k_j) versus 1/T. Thus, the reaction rate as a function of temperature can be evaluated.

However, at very high alcohol molar ratios to triglycerides, alcohol concentration is presumed to be constant. In this case, the rate of reaction is dependent on the triglyceride concentration and was called "pseudo first-order" by Freedman. Another proposition, "shunt reaction," was included in the case of the data that does not fit the consecutive model. This reaction is considered to have a fourth-order kinetic rate (1st order with triglyceride and 3rd order with alcohol) (Mjalli et al. 2009).

Material balance for biodiesel production in CSTR can be obtained for each compound as described below:

$$V\frac{dC_i}{dt} = Fi_oCi_o - FiCi - \sum_{j=1}^{n} rjV \quad (7a)$$

$$\frac{dC_{TG}}{dt} = \frac{F_{TGo}C_{TGo}}{V} - \frac{F_{TG}C_{TG}}{V} - r_1 \quad (7b)$$

$$V\frac{dC_{DG}}{dt} = F_{DGo}C_{DGo} - F_{DG}C_{DG} - (-r_1 + r_2) \quad (7c)$$

$$V\frac{dC_{MG}}{dt} = F_{MGo}C_{MGo} - F_{MG}C_{MG} - (-r_2 + r_3) \quad (7d)$$

$$V\frac{dC_G}{dt} = F_{Go}C_{Go} - F_GC_G - (-r_3) \quad (7e)$$

$$V\frac{dC_E}{dt} = F_{Eo}C_{Eo} - F_EC_E - (-r_1 - r_2 - r_3) \quad (7f)$$

$$V\frac{dC_A}{dt} = F_{Ao}C_{AO} - F_AC_A - (r_1 + r_2 + r_3) \quad (7g)$$

Since heat is transferred between the biodiesel transesterification reactor and the cooling coil, an energy balance has to be obtained for both of them.

Energy Balance on the Reactor

The energy balance on the CSTR-type reactor can be described in the equation below (Mjalli et al. 2009):

$$V(Cp_{TG}C_{TG} + Cp_{DG}C_{DG} + Cp_{MG}C_{MG} + Cp_GC_G + Cp_AC_A + Cp_EC_E)\frac{dT}{dt}$$
$$=(F_{TGO}C_{TGO}Cp_{TG} + F_{AO}C_{AO}Cp_A)(T_O - T)-(V(r_1\Delta H_1 + r_2\Delta H_2 + r_3\Delta H_3)$$
$$-(UA_H\Delta T) \quad (3.6) \quad (8)$$

Energy Balance on the Cooling Coil

The energy balance on the cooling coil can be expressed as the following (Mjalli et al. 2009):

$$\frac{dT_C}{dt} = \frac{F_{Co}}{V_C}(T_C - T) + \frac{UA_H\Delta T}{\rho_C V_c\ Cp_c} \quad (9)$$

$$\frac{1}{UA_H} = \frac{1}{h_oA_o} + \frac{X_w}{K_wA_w} + \frac{1}{h_i\ A_i} \quad (10)$$

$$Nu = \left[\left(3.66 + \frac{4.343}{a}\right)^3 + 1.158\left(\frac{Re(d_i/C)^{1/2}}{b}\right)^{3/2}\right]^{1/3}\left(\frac{\mu}{\mu_s}\right)^{0.14} \quad (11)$$

$$a = 1 + \frac{927((C/D)}{Re^2\,Pr} \quad (12)$$

$$b = 1 + \frac{0.477}{Pr} \quad (13)$$

$$h_i = \frac{k_i}{d_i}Nu(3.12) \quad (14)$$

$$h_o = \frac{k_{max}}{D_a}\left(1.10\left(\frac{ND_a^2\rho}{\mu}\right)^{0.62}\right)\left(\frac{C_p\mu^{0.333}}{k_{max}}\right) \quad (15)$$

Table 1.Operating Conditions and CSTR Type Reactor Specifications (Mjalli et al. 2009)

Unit	Value	Symbol	Parameter
°C	60	T	Reactor temperature
atm	1	P	Reactor pressure
m^3/hr	27.08	F_O	Reactant flow rate
m^3/s	0.00268	F_C	Coolant flow rate
rps	6	N	Stirrer rotational speed

kmol/m^3	1.11	C_{TGo}	Initial concentration of TG
kmol/m^3	6.66	Cao	Initial concentration of MeOH
–	69176.45	Re	Reynolds number (shell side)
m^3	27.08	V	Reactor volume
m	0.913	Da	Agitator diameter
m^2	50.48	A_H	Overall heat transfer area
m	0.0779	d_i	Inner diameter of cooling coil
M	0.0889	d_o	Outer diameter of cooling coil
m^3	0.86145	V_C	Volume of cooling coil
M	180.75	L	Length of cooling coil
M	0.0055	Xw	Wall thickness of cooling coil

Simulation Work

A simulation programme is constructed for a biodiesel transesterification reactor by using the programme MATLAB/ Simulink. It is a powerful software for the simulation of dynamic model analysis. It consists of a Simulink part to build the models and study the characteristics of dynamic situations. Figure 2 shows the MATLAB/ Simulink of the dynamic model.

Result and Discussion

Effect of Triglyceride Concentration

Figure 3 shows the exit temperature responses due to a step change in the feed concentration of triglyceride. Initially, when the feed concentration of triglyceride was 1.11 kmol/m^3, the exit temperature from the reactor was 316.7 K, while the feed concentration of triglyceride stepped to 1.22 kmol/m^3, 1.33 kmol/m^3, 1.44 kmol/m^3, 1.55 kmol/m^3, and 1.67 kmol/m^3, the reactor temperature responses decreased to 316.1 K, 315.9 K, 315.6 K, 314.9 K, and 314.9 K, respectively. Because of the increase in the feed concentration, the number of triglyceride moles increases at a constant volume of the reactor; therefore, the energy to break down more triglyceride molecules becomes insufficient.

Effect of Methanol Concentration

Figure 4 shows the exit temperature responses due to a step-change in the feed concentration of methanol. Initially, when the feed concentration of methanol was 6.66 kmol/m^3, the exit temperature from the reactor was 316.7 K, while the feed concentration of triglyceride stepped to 7.33 kmol/m^3, 7.99 kmol/m^3, 8.66 kmol/m^3, 9.3 kmol/m^3, and 10 kmol/m^3, the reactor temperature responses increased to 317 K, 317.3 K, 317.7 K, 317.9 K, and 318.2 K, respectively. With the negative step-change in methanol concentration from 6.66 kmol/m^3 to 5.99 kmol/m^3, 5.33 kmol/m^3, 4.66 kmol/m^3, 3.97 kmol/m^3, and 3.33 kmol/m^3, the reactor temperature response was decreased from 316.7 K to 316.5 K, 316.1 K, 315.7 K, 315.3 K, and 315 K, respectively.

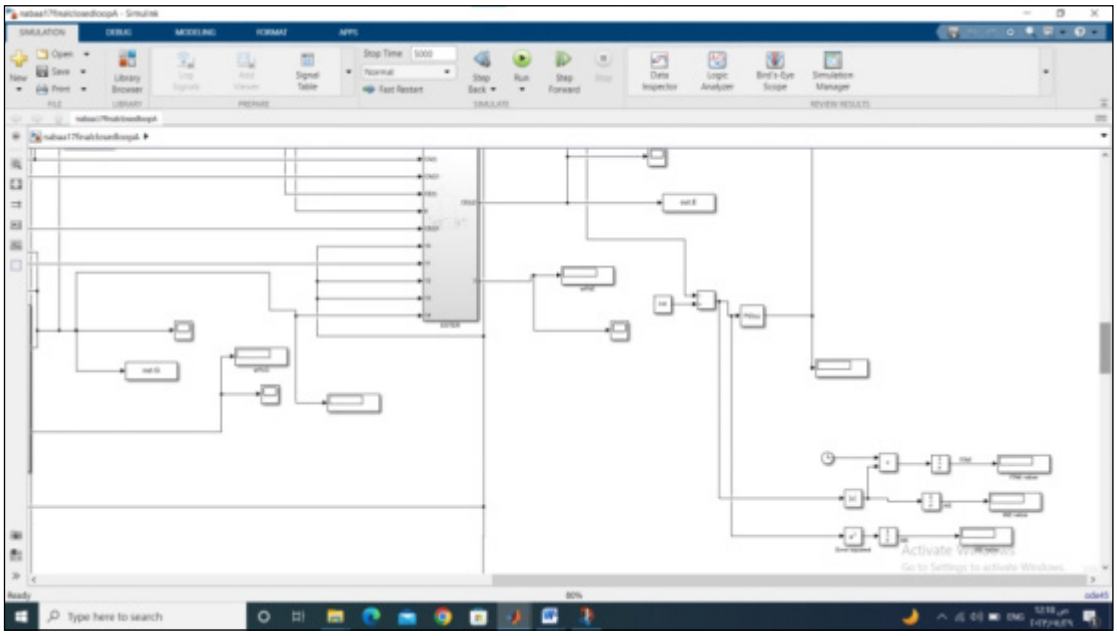

Figure 2. Simulation Work of the PID Controller for the Biodiesel Transesterification Reactor

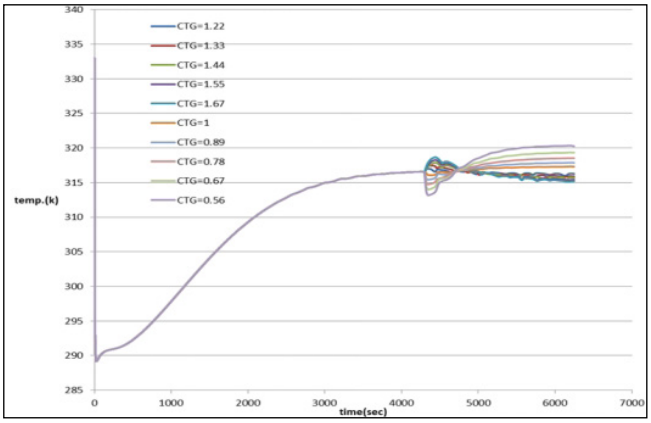

Figure 3.Exit Temperature Responses due to Step Change in Triglyceride Concentration (kmol/m3)

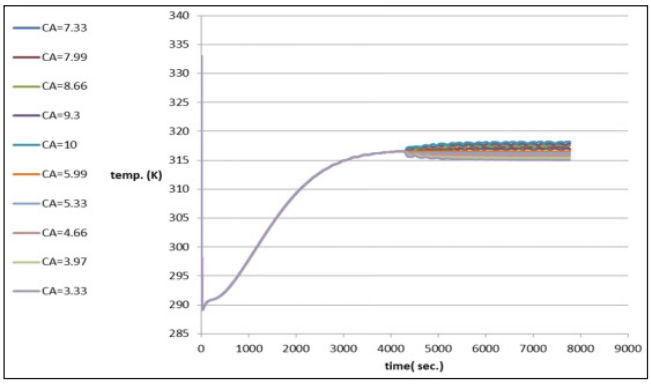

Figure 4.Exit Temperature Responses due to Step Change in Methanol Concentration (kmol/m3)

Effect of Feed Temperature

Figure 5 shows the exit reactor temperature responses when the feed temperature is stepped from 333 K to 335 K and 338 K. Initially, the exit reactor temperature was 316.7 K. When the feed temperature of TG was 333 K, while the feed temperature stepped from 333 K to 335 K and 338 K, the exit reactor temperature was increased from 316.7 K to 318 K and 320.1 K.

When there was a negative step change in feed temperature from 333 K to 328 K, 323 K, 318 K, and 313 K, the exit reactor temperature decreased from 316.7 K to 313.3 K, 310 K, 306.7 K, and 303.1 K.

Effect of Volumetric Flowrate

Figure 6 shows the exit reactor temperature responses when the volumetric flow rate was stepped from 27.08 m^3/hr to 30 m^3/hr, 35 m^3/hr, 40 m^3/hr, 45 m^3/hr, 22 m^3/hr, 20 m^3/hr, 18 m^3/hr, and 0.15 m^3/hr. First, the exit reactor temperature was 316.7 K, when the volumetric flow rate was 27.08 m^3/hr. The exit reactor temperature response decreased from 316.7 K to 316.6 K, 316.3 K, 316.2 K, and 316 K when the volumetric flow rate was stepped from 27.08 m^3/hr to 30 m^3/hr, 35 m^3/hr, 40 m^3/hr, and 45 m^3/hr because of the increasing velocity of biodiesel, meaning

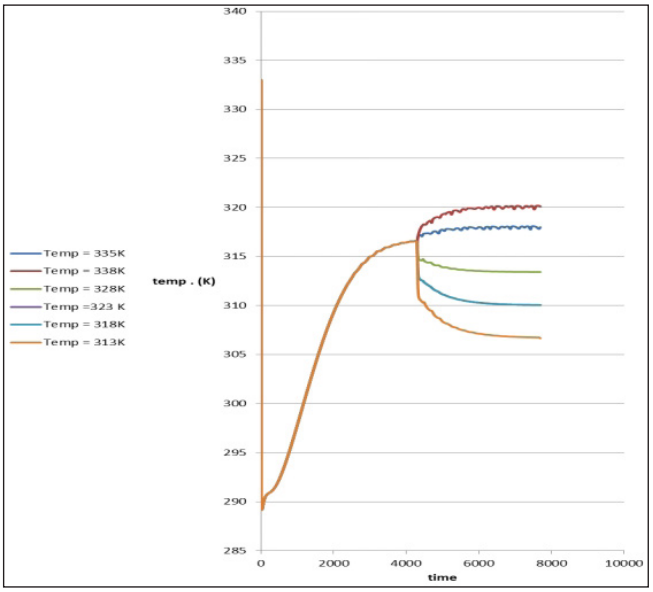

Figure 5.Exit Temperature Responses due to Step Change in Feed Temperature (K)

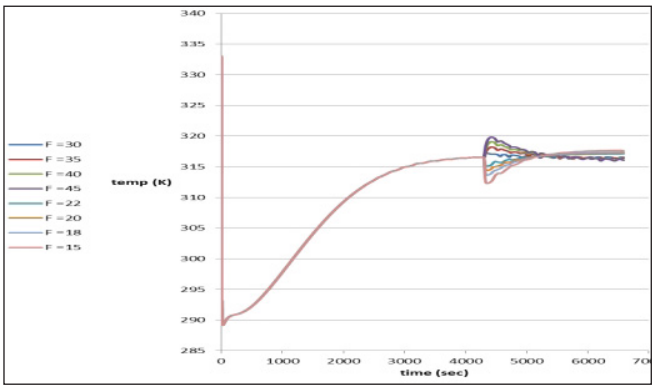

Figure 6.Exit Reactor Temperature Responses due to Step-Change in Volumetric Flow Rate (m3/hr)

that the shortness of residence time inside the reactor, therefore the biodiesel temperature will decrease. By a negative step-change of the volumetric flow rate from 27.08 m^3/hr to 22 m^3/hr, 20 m^3/hr, and 18 m^3/hr, then exit.

The reactor temperature increased to 317.2 K, 317.2 K, 317.3 K, and 317.6 K due to the residence time of biodiesel inside the reactor, which is sufficient to raise the temperature of the biodiesel.

Conclusion

Biodiesel transesterification reactors encompass a complex interplay of chemical reactions and heat transfer dynamics. The flow properties of both coolant and reactants have a nonlinear impact on the state variables of the process. In this study, the transesterification reactor model was developed and its performance was compared against data from previous research by Mjalli et al. (2009).

A simulation of the biodiesel reactor's dynamic model was designed and executed using MATLAB/SIMULINK

software. The dynamic analysis demonstrated a favorable reactor temperature response to variations in triglyceride concentration, methanol concentration, and the volumetric feed flow rate.

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