International Journal of Engineering Research and Development
e-ISSN: 2278-067X, **p**-ISSN: 2278-800X, www.ijerd.com
Volume 6, Issue 9 (April 2013), PP. 59-66

Simulation Model for Biodiesel Production using Plug Flow reactor: Non Isothermal Operation

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Abstract:- A mathematical model capable to predict the dynamic behavior of Non Isothermal tubular (plug) flow reactor on transesterification of sun flower oil has been developed. The model was simulated using the k values obtained from literature and temperatures of 25^o C , 40^o C and 60^oC with varying feed ratios of 3:1, 6:1 and 9:1 respectively. The simulated results showed the effect of temperature and feed ratio on conversion and yield of biodiesel. The highest conversion was observed at 60^oC while that of yield was at 40^oC. A conversion of 90 % was achieved with a reactor volume of 487.7 liters.

Keywords:- Modeling, Plug flow Reactor, Trans esterification, Vegetable oil, Biodiesel production

I. INTRODUCTION

Due to the drastic increase in environmental problems, associated with petroleum based fuel, a search for alternative fuel has gained significant attention in recent times (Rie *et al* ., 2006; Ayhan *et al*., 2005 and Fukuda *et al*., 2001). Among the different possible resources, biodiesel fuel derived from triglycerides of renewable lipid feedstock (Kusdiana *et al*., 2004) such as vegetable oil or animal fats has shown a great potential as a reliable substitute for petroleum based diesel fuel (Lalita *et al*., 2004). Biodiesel produced from vegetable oil is a good alternative to petroleum based diesel because of the similar properties exhibited to those of petroleum based diesel such as viscosity, volumetric heating value, high cetane and flash points (Lalita *et al*., 2004). Also, its environmental benefits such as renewability, carbon neutral property, biodegradability and low emission profile could be a solution to global warming (Rie *et al* ., 2006). The direct use of vegetable oils in fuel engines is problematic due to the high viscosity (about 11-17 time higher than that of petroleum based diesel) and lower volatility. There is incomplete combustion problem, which may lead to carbon deposit in the fuel injector [Gumpon *et al*., 2000; Kisidiana *et al*., 2001, Guan *et al*., 2007; Robiromerd and Martinez ., 2006).. Therefore, several methods have been employed to improve the properties of vegetable oil for fuel purposes. the trans esterification with ethanol or methanol to produce methyl ester that burned directly in unmodified diesel engines, with very low deposit formation (Noureddini *et al*.,1997). Generally, biodiesel is produced using batch reactor but the use of batch reactor is of high cost and achieving a very small quantity of the product.

Several research groups (He *et al*., 2007; Rie *et al*., 2006; Saka *et al*., 2001 and Kusdiana *et al* ., 2001) have studied biodiesel production experimentally using CSTR (reactive distillation technique) with efficient and high yield, non catalytic with supercritical methanol, high reaction rate (batch reactor) but required high temperature (350-450^o C and pressure (45-65 MPa) and fixed bed reactor. In order to reduce the temperature and pressure to 270^o C and 7 Mpa, Saka *et al*., (2001) has proposed a two step mechanism – by hydrolysis and trans-esterification. Guoging *et al*., (2007) used micro-tube reactor to achieve high conversion because of increased high flow rate, high contact surface and small reaction volume.

Kinetic model for trans-esterification of vegetable oil has been developed on various reactor types, with various catalyst system including plug flow reactors (Bambase *et al*., 2007; Demibas , 2002; Marjanovic *et al*., 2010 and Darnoko *et al*., 2000). However, very little has been reported of the development of dynamic behavior which actually predicts the performance of plug flow reactors. In this study, mathematical model to predict the dynamic effect of the trans esterification of vegetable oil in a non-isothermal plug flow reactor have been developed. The developed models were simulated using Polymath soft ware package to predict the optimal temperature and reactor volume to achieve high conversion and yield.

II. MODEL FORMULATION

2.1 Reaction mechanism for alkali trans esterification of triglycerides and methanol

The alkali trans esterification of triglycerides to produce biodiesel proceeds through three steps reversible reaction (Ayhan *et al*., 2005).

$$TG + CH_3OH \underset{K_2}{\overset{K_1}{\rightleftarrows}} DG + R_1COOCH_3 \qquad 1$$

$$DG + CH_3OH \underset{K_4}{\overset{K_3}{\rightleftarrows}} MG + R_2COOCH_3 \qquad 2$$

$$MG + CH_3OH \underset{K_6}{\overset{K_5}{\rightleftarrows}} GL + R_3COOCH_3 \qquad 3$$

Equation (1) through (3) could be explicitly written as;

$$TG + A \xrightarrow{K1} DG + ME \qquad 4$$

$$DG + ME \xrightarrow{K2} TG + A \qquad 5$$

$$DG + A \xrightarrow{K3} MG + ME \qquad 6$$

$$MG + ME \xrightarrow{K4} DG + A \qquad (7)$$

$$MG + A \xrightarrow{K5} GL + ME \qquad (8)$$

$$GL + ME \xrightarrow{K6} MG + A \qquad (9)$$

Where TG is triglycerides, A is alcohol (methanol), DG is diglycerides. MG Is monoglycerides, ME is methyl ester, GL is glyceride and K_1 through K_6 are the rate constants.

The rate expressions for the alkaline trans esterification of triglycerides and alcohol to methyl 10

$$r_{TG} = -K_1 C_{TG} C_A + K_2 C_{DG} C_{ME} \qquad (10)$$

$$r_{DG} = K_1 C_{TG} C_A - K_2 C_{DG} C_{ME} - K_3 C_{DG} C_A + K_4 C_{MG} C_{ME} \qquad (11)$$

$$r_{MG} = K_3 C_{DG} C_A - K_4 C_{MG} C_{ME} - K_5 C_{MG} C_A + K_6 C_{GL} C_{ME} \qquad (12)$$

$$r_A = -K_1 C_{TG} C_A + K_2 C_{DG} C_{ME} - K_3 C_{DG} C_A + K_4 C_{MG} C_{ME} - K_5 C_{MG} C_A + K_6 C_{GL} C_{ME} \qquad (13)$$

$$r_{GL} = K_5 C_{ME} C_A - K_6 C_{GL} C_{ME} \qquad (14)$$

$$r_{ME} = K_1 C_{TG} C_A - K_2 C_{DG} C_{ME} + K_3 C_{DG} C_A - K_4 C_{MG} C_{ME} + K_5 C_{MG} C_A - K_6 C_{GL} C_{ME} \qquad (15)$$

Where $-r_{TG,}\, r_{DG},\, r_{MG},\, -r_{A,}\, r_{GL},\, r_{ME}$ are the reaction rates of triglycerides, diglycerides, monoglycerides, alcohol, glycerides and methyl ester

2.2 Model Equations

The following assumptions were made for the modeling of plug flow reactor. (1) The reactor is operated at steady state (2) The reaction is a liquid phase reaction, density is constant so the volumetric flow rate at the inlet is equal to that of the outlet (3) The reaction is carried out in a non-isothermal condition (4) The work term is negligible (5) The specific heat capacity is constant (6) The summation of the energy is negligible, compared to the enthalpy.

A mole balance on species j at instant in time, t, yields the following equation (Fogler, 2008)

$$\begin{pmatrix} Rate\ of \\ flow\ of \\ j\ into \\ the\ system \\ \left(\frac{moles}{time}\right) \end{pmatrix} - \begin{pmatrix} Rate\ of \\ flow\ of \\ j\ from \\ the\ system \\ \left(\frac{moles}{time}\right) \end{pmatrix} + \begin{pmatrix} Rate\ of \\ generation \\ of \\ j\ within\ the \\ system \\ \left(\frac{moles}{time}\right) \end{pmatrix} = \begin{pmatrix} Rate\ of \\ accumulation \\ of \\ j\ within\ the \\ system \\ \left(\frac{moles}{time}\right) \end{pmatrix} \qquad (16)$$

$$F_{jo} - F_j + G_j = \frac{dN}{dt} \qquad (17)$$

Where F_{jo} and F_j are the inlet and outlet molar flow rate of species $_j$, G_j is the generation term, $\frac{dN}{dt}$ is the accumulation term. Applying the model assumptions on equation 17, the mathematical model for the trans esterification of triglycerides and alcohol in non isothermal plug flow reactor is derived as follow;

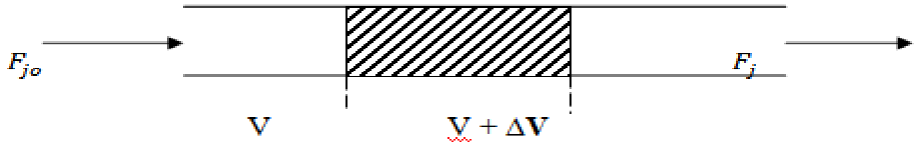

Figure 1 Schematic diagram of a plug flow reactor.

From equation 17, the model equation is

$$F_{jo}\Big|V - F_j\ \Big|V + \Delta V + r_j\Delta V = 0 \qquad (18)$$

Dividing through by ΔV and taking a limit as ΔV -------- 0,

$$\frac{dF_j}{dV} = r_j \qquad (19)$$

But $F_j = vC_j$ Therefore, equation 19 becomes

$$\frac{vdC_j}{dV} = r_j \qquad (20)$$

$$\frac{dC_j}{d\left(\frac{V}{v}\right)} = \frac{dC_j}{d\tau} = r_j \qquad (21)$$

Substituting equation (10) through (15) into equation (21) will give the mathematical model of PFR used for the trans esterifcation of triglycerides and methanol as

$$\frac{dC_{TG}}{d\tau} = -K_1C_{TG}C_A + K_2C_{DG}C_{ME} \qquad (22)$$

$$\frac{dC_{DG}}{d\tau} = K_1C_{TG}C_A - K_2C_{DG}C_{ME} - K_3C_{DG}C_A + K_4C_{MG}C_{ME} \qquad (23)$$

$$\frac{dC_{MG}}{d\tau} = K_3C_{DG}C_A - K_4C_{MG}C_{ME} - K_5C_{MG}C_A + K_6C_{GL}C_{ME} \qquad (24)$$

$$\frac{dC_A}{d\tau} = -K_1C_{TG}C_A + K_2C_{DG}C_{ME} - K_3C_{DG}C_A + K_4C_{MG}C_{ME}$$

$$-K_5C_{MG}C_A + K_6C_{GL}C_{ME} \qquad (25)$$

$$\frac{dC_{GL}}{d\tau} = K_5C_{ME}C_A - K_6C_{GL}C_{ME} \qquad (26)$$

$$\frac{dC_{ME}}{d\tau} = K_1C_{TG}C_A - K_2C_{DG}C_{ME} + K_3C_{DG}C_A - K_4C_{MG}C_{ME}$$
$$+K_5C_{MG}C_A - K_6C_{GL}C_{ME} \qquad (27)$$

The conversion of the species j within the reactor is given as

$$X_{TG} = 1 - \frac{C_{TG}}{C_{To}} \qquad (28)$$

The yield of methyl ester within the reactor is

$$Y = \frac{C_{ME}}{C_{TGO}} \qquad (29)$$

The volume of the reactor is given as

$$V = v_o\left(\frac{1}{-r_{TG}}\right)\int_{x_{TG}=0}^{x_{TG}} dX_{TG} \qquad (30)$$

For an open system in which energy is added by the surrounding and mass flow across the boundaries, the energy balance for the case of only one specie entering and leaving the system is given as (Fogler 2008).

$$\begin{pmatrix} \text{Rate of} \\ \text{accumulation} \\ \text{of energy} \\ \text{within the} \\ \text{system} \end{pmatrix} = \begin{pmatrix} \text{Rate of} \\ \text{flow of} \\ \text{heat from the} \\ \text{surrounding} \\ \text{to the} \\ \text{system} \end{pmatrix} - \begin{pmatrix} \text{Rate of} \\ \text{work done} \\ \text{by the} \\ \text{system on} \\ \text{the} \\ \text{surrounding} \end{pmatrix} + \begin{pmatrix} \text{Rate of energy} \\ \text{added to the} \\ \text{system} \\ \text{by mass} \\ \text{flow to} \\ \text{the system} \end{pmatrix}$$

$$- \begin{pmatrix} Rate\ of\ energy \\ removed\ from \\ the\ \ system \\ by\ mass \\ flow\ from \\ the\ system \end{pmatrix} \qquad (31)$$

$$\frac{dE}{dt}\ syst\ = \mathrm{Q}\ -\ \mathrm{W}\ +\ \textstyle\sum_{i=1}^{n} E_{io} F_{io}\ -\ \sum_{i=1}^{n} \mathrm{E_i F_i} \qquad (32)$$

For flow reactors at steady state, the accumulation term is equal to zero. Therefore equation (32) becomes

Q-W+$\sum_{i=1}^{n} \mathrm{E_{io} F_{io}}$-$\sum_{i=1}^{n} \mathrm{E_i F_i} = 0$ (33) Where Q is the heat flow from the surrounding to the system, W is the work done by the system, E_{io} and E_i are the energies added or removed from the system by mass flow into or out of the system respectively.

Based on the model assumptions, the energy balance for a PFR in which heat is added or removed through the cylindrical wall and by mass flow across the boundaries is as follows

$$\Delta Q = Ua\Delta A(Ta - T) = Ua\Delta V(Ta - T) \qquad (34)$$

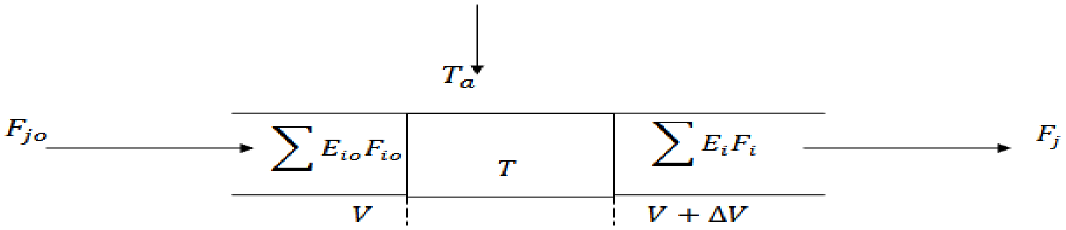

Figure 2, Schematic Diagram of PFR with heat lost or gained.

$$\Delta Q + \textstyle\sum F_{io} H_{io} \Delta V - \Delta V + \Delta V = 0 \qquad (35)$$

Where $\mathrm{H_i}$ is the enthalpy of the specie j, Q , the heat transfer coefficient of the surrounding fluid, ΔA is the total surface area through which heat is transferred to the system, Ta and T are the temperature of the surrounding fluid “ambient temperature” and temperature of the reactor respectively.

$$\mathrm{U_a \Delta V}(T_a - T) + \textstyle\sum F_{io} H_{io} \Big|V - \sum F_i H_i \Big| V + \Delta V = 0 \qquad (.36)$$

Dividing through by ΔV and taking a limit as ΔV 0,

$$\mathrm{U_a}(T_a - T) - \sum \frac{dF_{iH_i}}{dV} = 0 \qquad (.37\ )$$

Differentiating equation (.37),

$$\mathrm{U_a}(T_a - T) - \sum \frac{H_i dF_i}{dV} - \sum \frac{F_{idH_i}}{dV} = 0 \qquad (\ .38)$$

From mole balance on specie,

$$\frac{dF_i}{dV} = r_i = v_i(-r_i) \qquad (\ 39)$$

Where v_i is the volumetric flow rate of specie i and $-r_j$ is the overall reaction rate of the whole species within the reactor.

Also, $\frac{dH_i}{dV} = \frac{C_{PdT}}{dV}$ (40)

Where C_p is the specific heat capacity of the specie i.

Substituting equation (.39) and (40) into equation (38),

$$\mathrm{U_a}(T_a - T) - \textstyle\sum v_i H_i(-r_j) - \sum \frac{\mathrm{F_i C_p dT}}{dV} = 0 \qquad (\ 41)$$

Where $v_i H_i$ is the heat of reaction and is represented as ΔH_{RX}.

Rearranging equation (41) gives;

$$\frac{dT}{dV} = \frac{r_i \Delta H_{RX} + \mathrm{U_a}(T_a - T)}{\sum F_i C_{pi}} \qquad (42)$$

Equation (.42) is the energy balance equation showing the rate of change of temperature with respect to the change in volume of a non-isothermal PFR.

III. RESULTS AND DISCUSSION

Kinetic models were developed and simulation was carried out on the transesterification of vegetable oil (sunflower oil) in a non isothermal plug flow reactor and the results are presented in Figures 2 to 8

Figure 2 shows that at time (t=0), there was no change in composition but just after 14 minutes, the composition of the reactants (C_{TG} and C_A) started decreasing while that of the products (C_{ME} and C_{GL}) increased with time. Intermediates (C_{DG}, and C_{MG}) were also formed but disappeared as the reaction proceeds with time. It also shows that the rate of formation of the desired product (biodiesel) is higher than that of the undesired product (glycerol) and the rate of disappearance of the major reactant (triglycerides) is higher than that of the alcohol indicating that the limiting reactant is triglycerides and the overall reaction rate is time dependent. The observed result is comparable to the report in literature (Noureddini *et al* ., 1997).

The effect of temperature on the dynamic behavior of triglycerides conversion as reaction proceeds with time is shown in Figure 4. Initial conversions of 0.0695, 0.199 and 0.524 were obtained at 25°C, 40°C and 60°C respectively. The result shows that, the reaction rate is faster at higher temperature but as the reaction proceeds with time, the conversion at 60°C remain constant while that of 25°C and 40°C increases linearly with time. This is an indication that higher temperatures will favor backward reaction, which agreed with reports in literature (Noureddini *et al*., 1997). Figure 5 shows that the yield increases as time increases. The yield obtained with time of 14 min was 0.127, 0.282 and 1.15 at 25^0C, 40^0C, and 60^0C respectively. Also confirming the effect of temperature on yield but at time (t=598 minutes) the yield at 60°C remained constant while that at 25°C and 40 ^{o} C linearly increased. Also, close to the end of the reaction, the yield at 40°C increased to 2.501 which is the highest compared to the other temperatures

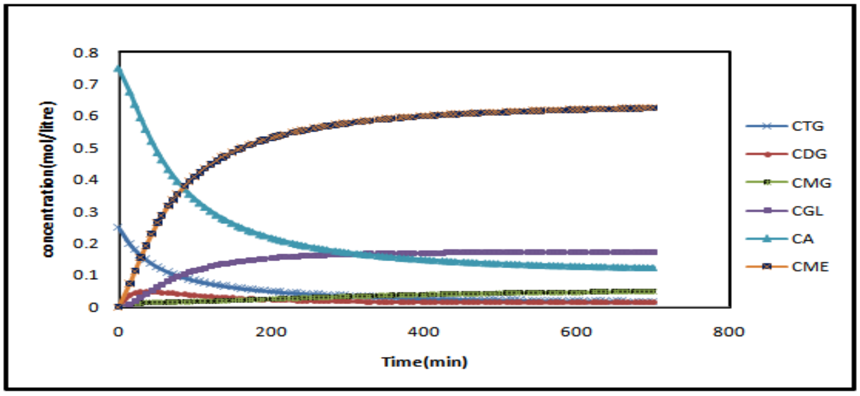

Figure 3 Composition profile versus time at constant temperature = 40°C, feed ratio = 3:1

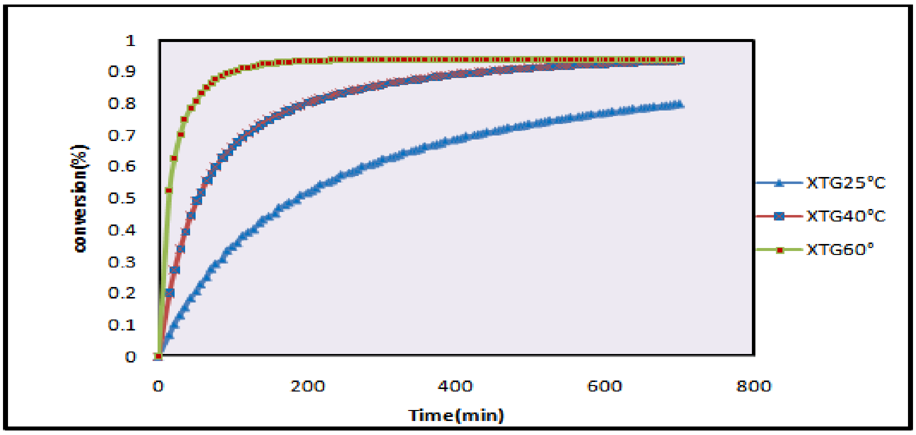

Figure 4 Effect of temperature on triglycerides conversion at constant feed ratio=3:1

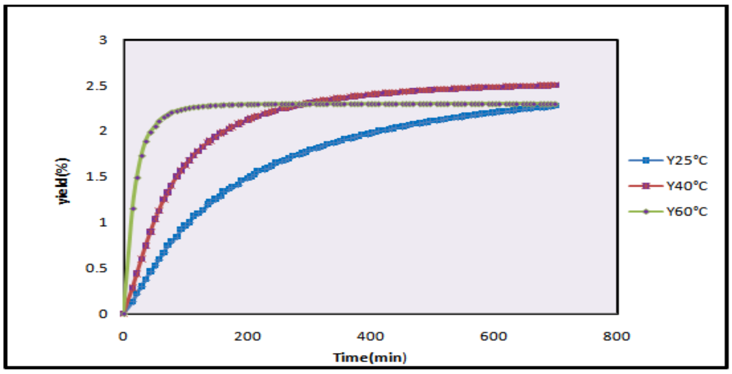

Figure 5 Effect of temperature on methyl ester yield at constant feed ratio =3:1.

3.1 Effect of Feed Ratio

The conversions for 3:1, 6:1, and 9:1 feed ratios are 0.199, 0.223 and 0.245 respectively (See Figure 6). As the reaction time increased to 700 minutes, the conversion also increased to 0.934, 0.998, and 0.999 for 3:1, 6:1 and 9:1 feed ratio respectively. This shows that increasing the feed ratio results to higher conversion. However, increasing the ratio above 9:1 may not result to any significant increase in conversion since there was convergence for 6:1 and 9:1 feed ratios. The yield for 3:1, 6:1, and 9:1 feed ratio 0.1999, 0.328, and 0.373 respectively (See Figure 7). Also the yield increased to 0.934, 2.941 and 2.971 with an increase in the reaction time to (700 minutes). Figure 8 shows the dynamic behavior of the conversion with respect to reactor volume. The conversion of triglycerides increases linearly with the volume of the reactor with constant volumetric flow rate of 0.01 liters per minutes. As the reactants moves from one point to another in the reactor, the volume of the reactor continues to increase hence results to increase in the triglycerides conversion and the total volume required to obtain a maximum of 0.934 conversion of sunflower at 40°C and 3:1 is Figure 8 shows the dynamic behavior of the conversion with respect to reactor volume. The conversion of triglycerides increases linearly with the volume of the reactor with constant volumetric flow rate of 0.01 liters per minutes. As the reactants moves from one point to another in the reactor, the volume of the reactor continues to increase hence results to increase in the triglycerides conversion and the total volume required to obtain a maximum of 0.934 conversion of sunflower oil at 40°C and 3:1 is 487.7 liters.

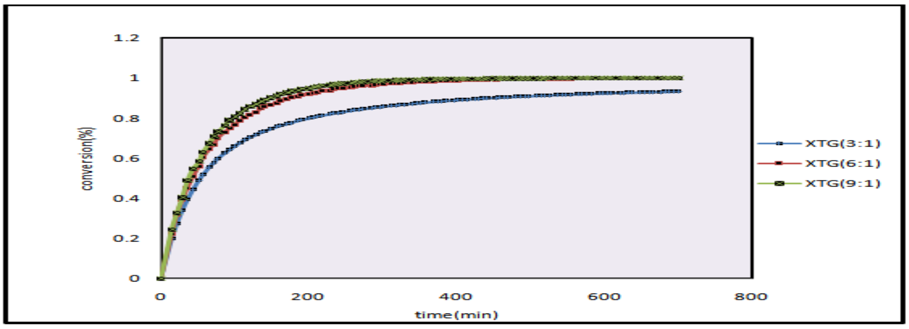

Figure 6 Effect of feed ratio on triglycerides conversion at constant temperature= $40°C$

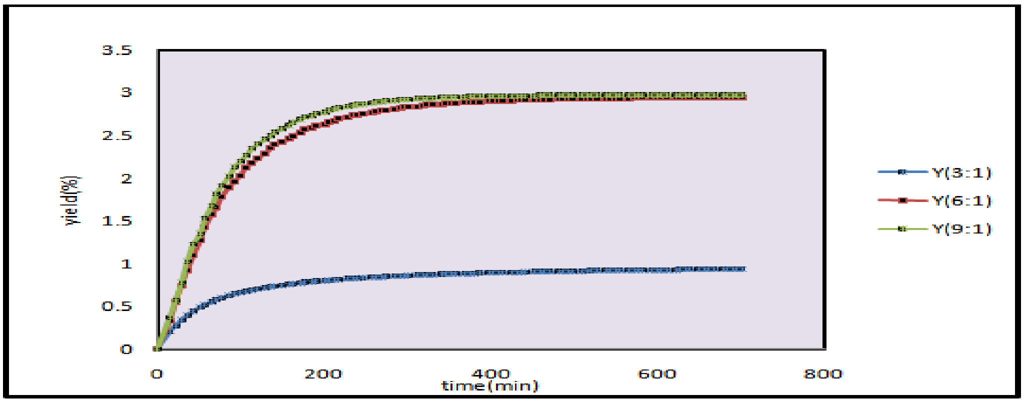

Figure 7. Effect of feed ratio on yield at constant temperature

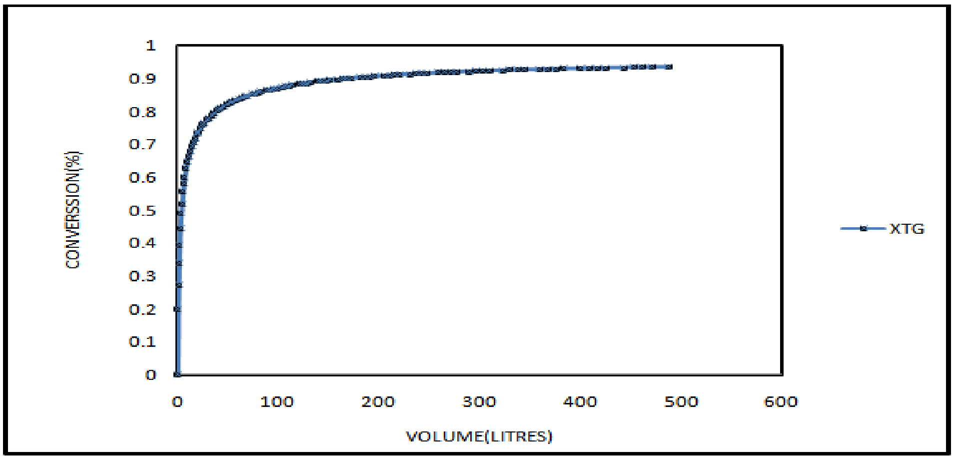

Figure 8, A Plot of conversion versus reactor volume.

IV. CONCLUSION

The mathematical model and simulation carried out shows that the transesterification of vegetable oil (sunflower) with methanol in a non isothermal tubular (plug) flow reactor is tremendously favorable by increasing the alcohol to oil ratio from 3:1 to 6:1 and 9:1. At higher ratio, the reaction mean residence time, was greatly reduced which increased the reactor performance. The developed mathematical model is found to be more reliable and more useful to predict the dynamic behavior of the trans esterification of vegetable oil and alcohol in a non isothermal plug flow reactor operating condition considered especially at 40^{o} C. The triglycerides conversion increased with temperature and remained constant as reaction time increased while the methyl ester yield decreased with increase in temperature with respect to time because the reaction is reversible. A volume of 487.7 liters can achieve a conversion of 90%

Acknowledgment

The authors would gratefully like to appreciate the Niger Delta University for the enabling environment for this study.

Nomenclature

K_1	Reaction rate constant for elementary reaction 1 ($\min^{1}$)
K_2	Reaction rate constant for elementary reaction 2 ($\min^{1}$)
K_3	Reaction rate constant for elementary reaction 3 ($\min^{1}$)
K_4	Reaction rate constant for elementary reaction 4 ($\min^{1}$)
K_5	Reaction rate constant for elementary reaction 5 ($\min^{1}$)
K_6	Reaction rate constant for elementary reaction 6 ($\min^{1}$)
$-r_1$	reaction rate for elementary reaction 1 (mol $L^{-1}\min^{-1}$)
$-r_2$	reaction rate for elementary reaction 2 (mol $L^{-1}\min^{-1}$)
$-r_3$	reaction rate for elementary reaction 3 (mol $L^{-1}\min^{-1}$)
$-r_4$	reaction rate for elementary reaction 4 (mol $L^{-1}\min^{-1}$)
$-r_5$	reaction rate for elementary reaction 5 (mol $L^{-1}\min^{-1}$)
$- r_6$	reaction rate for elementary reaction 6 (mol $L^{-1}\min^{-1}$)
r_{TG}	Reaction rate for triglyceride (mol L^{-1} $\min^{-1}$)
r_{DG}	Reaction rate for diglyceride (mol L^{-} $\min^{-1}$)
r_{MG}	reaction rate for monoglyceride (mol L^{-1} $\min^{-1}$)
r_A	Reaction rate for Alcohol (mol $L^{-}\min^{-1}$)
r_{ME}	Reaction rate for methyl ester (mol L^{-1} $\min^{-1}$)
r_G	Reaction rate for glycerol (mol L^{-1} $\min^{-1}$)
r_J	Reaction rate for species J (mol L^{-1} $\min^{-1}$)
C_{TG}	Final concentration of triglyceride (mol L^{-1})
C_{DG}	Final concentration of diglyceride (mol L^{-1})
C_{MG}	Final concentration of monoglyceride (mol L^{-1})
C_A	Final concentration of alcohol (mol L^{-1})
C_{ME}	Final concentration of methyl ester (mol L^{-1})
C_{TG0}	Initial concentration of triglyceride (mol L^{-1})
C_{DG0}	Initial concentration of diglyceride (mol L^{-1})

C_{MG0}	Initial concentration of monoglyceride (mol L^{-1})
C_{A0}	Initial concentration of alcohol (mol L^{-1})
C_{ME0}	Initial concentration of methyl ester (mol L^{-1})
C_{pi}	Specific heat capacity of outlet species (Cal K^{-1} Kg^{-1} mol
C_{p0}	Specific heat capacity of inlet species (Cal K^{-1} Kg^{-1} mol^{-1})
VR	Volume of reactor (L)
$\upsilon 0$	Inlet volumetric flow rate (L min-1)
υ	Outlet volumetric flow rate (L min-1)
τ	Space time (min)
S	Space velocity (min-1)
Q	Heat absorbed (Cal mol^{-1})
T_i	Temperature of outlet species
T_0	Temperature of inlet species

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