Optimization of Biodiesel Synthesis in a Batch reactor using Maximum Principle

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Abstract: Biodiesel is produced through transesterification of vegetable oils or animal fats. It significantly depends on many reaction parameters among which temperature and stirring are most important. In transesterification, reaction rates follow Arrhenius equation (i.e. function of temperature). Also, effect of stirring effect on reaction rate can be described as Boltzmann sigmoid form (as a function of stirring). On that outlook, in this research article, a mathematical model is proposed to study the simultaneous effect of stirring and temperature on transesterification process. A two control parameters optimal control problem is formulated to optimize biodiesel yield. Maximum principle is used to solved the optimal control problem. The optimal control problem is solved using numerically method in Matlab. Simulation results of the optimality system are plotted in figures and found to be satisfactory.

Key-Words: Biodiesel, Transesterification, Stirring, Temperature, Maximum Principle.

1 Introduction

Fatty acid methyl esters (FAME) collectively known as biodiesel. It is derived from edible or non-edible vegetable oils such as Jatropha oil [1, 2], rapeseed oil [3], soybean oil [4] or animal fat [5, 6] and can be used as an alternative fuel for diesel [7, 8].

Transesterification or alcoholysis is commonly employed to convert vegetable oil to biodiesel in the presence of catalyst [2, 9]. Transesterification of oil is influenced by different parameters such as molar ratio between alcohol and triglycerides, reaction time, catalyst concentration, and reaction temperature [10]. Among these parameters, stirring and temperature are the most important factors of transesterification process [5, 11].

Effects of temperature in transesterification is investigated in case of biodiesel production from different vegetable oils [12]. With increase in reaction temperature, conversion to biodiesel is also increased. But after a certain level of temperature (above $50^{\circ}C$), biodiesel yield decreases [13, 11].

Vegetable oil (triglycerides) is immiscible with alcohol due to their different polarity. Thus, biodiesel production process is very slow initially. This problem is avoided introducing stirrer rotation that increase the diffusion rates [1, 14]. Peterson et al. [15] has studied the effect of stirrer speed on the transesterification of vegetable oil with alcohol. The effect of the stirring in the transesterification of cotton seed oil has been analyzed by Rashid et al. [16]. He has shown that beyond an optimum stirrer speed production is decreased. Kafuku and Mbarawa [17] have also observed that the conversion efficiency increased with the increase in agitation rate until an optimum levels after that cavitation phenomena appeared which leads to a decrease in the overall efficiency. Thus, temperature and stirring are the most important control variables in biodiesel production. So, optimal temperature and stirring profiles are required for maximum production of biodiesel.

In this article, a mathematical model is proposed to study the effect of temperature and stirring on biodiesel yield. Using maximum principle, optimal stirring and temperature profile, as a control pair, is determined so that maximum amount of biodiesel can be obtained from transesterification of Jatropha oil. Simulation results are shown in figures to fulfill the analytical outcomes.

2 Mathematical Model for Biodiesel Production

Formulation of the mathematical model with basic assumptions are given below.

Biodiesel is produced reacting triglycerides with methanol in a batch reactor. The reactions happen in three reversible steps. During the course of reaction of triglycerides and methanol, some intermediates (diglyceride and monoglyceride) are considered. Therefore, we consider here that three consecutive reversible reactions occurred during the production of biodiesel. The schematic explanation of the reaction is given below [2, 18].

$$TG + AL \rightleftharpoons_{k_2}^{k_1} DG + BD$$

$$DG + AL \rightleftharpoons_{k_4}^{k_3} MG + BD \qquad (1)$$

$$MG + AL \rightleftharpoons_{k_6}^{k_5} GL + BD.$$

Here k_1 , k_3 , k_5 are forward reaction rates and k_2 , k_4 , k_6 are backward reaction rates. Mixing in the reaction system has significant effect on overall reaction rates. Here, we use k_s as the effect of stirring on reaction rate and the term can be defined as Boltzmann sigmoid form [1, 19]:

$$k_s = \frac{a}{1 + e^{-b(F-c)}},$$
 (2)

where F is the speed of stirrer and a, b and c are constants. Also, the dependency of reaction rate constants on temperature can be expressed by the Arrhenius equation [11, 18]:

$$k_i' = \alpha_i e^{\frac{-\beta_i}{T}}, \qquad (3)$$

where, T is the reaction temperature, α_i is the frequency factor, and values of α_i and β_i are given in Table 1. Thus, the combined effect of temperature and stirring on reaction rates can be expressed as [20]:

$$k_i = k'_i k_s = \left(\frac{a}{1 + e^{-b(F-c)}}\right) \alpha_i e^{\frac{-\beta_i}{T}}.$$
 (4)

We denote the concentration of triglycerides, diglycerides, monoglycerides, methanol (alcohol) and glycerol by C_T, C_D, C_M, C_A, C_G respectively. The mathematical model for the production of biodiesel in a batch reactor is governed by the following Ordinary Differential Equations, derived from the mass balance of the batch reactor:

$$\frac{dC_B}{dt} = k_1 C_T C_A - k_2 C_D C_B + k_3 C_D C_A - k_4 C_M C_B + k_5 C_M C_A - k_6 C_G C_B$$

$$\frac{dC_T}{dt} = -k_1 C_T C_A + k_2 C_D C_B,$$

$$\frac{dC_D}{dt} = k_1 C_T C_A - k_2 C_D C_B
- k_3 C_D C_A + k_4 C_M C_B,$$

$$\frac{dC_M}{dt} = k_3 C_D C_A + k_4 C_M C_B
- k_5 C_M C_A + k_6 C_G C_B,$$

$$\frac{dC_A}{dt} = -k_1 C_T C_A + k_2 C_D C_B
- k_3 C_D C_A + k_4 C_M C_B
- k_5 C_M C_A + k_6 C_G C_B,$$

$$\frac{dC_G}{dt} = k_5 C_M C_A - k_6 C_G C_B,$$
(5)

where k_i satisfies the relation (4). The initial concentrations are,

 $C_T(0) = C_{T_0}, C_B(0) = 0, C_D(0) = 0, C_M(0) = 0, C_A(0) = C_{A_0}, C_G(0) = 0.$

3 Optimal control approach

In [11], Diwekar and Benavides formulated an optimal control problem taking temperature only as control parameter. Here, we formulate a two control parameter optimal control problem taking stirring and temperature as control parameters. The aim is to maximize biodiesel production by controlling stirring and temperature simultaneously. Using maximum principle, we try to find control profile for temperature (T) and stirring (F) for maximum biodiesel production. The objective function is taken in the following manner:

maximize $J = C_B(t_f)$,

subject to the state system:

$$\frac{dC_B}{dt} = k_1 C_T C_A - k_2 C_D C_B
+ k_3 C_D C_A - k_4 C_M C_B
+ k_5 C_M C_A - k_6 C_G C_B$$

$$\frac{dC_T}{dt} = -k_1 C_T C_A
+ k_2 C_D C_B,$$

$$\frac{dC_D}{dt} = k_1 C_T C_A - k_2 C_D C_B - k_3 C_D C_A
+ k_4 C_M C_B,$$

$$\frac{dC_M}{dt} = k_3 C_D C_A + k_4 C_M C_B
- k_5 C_M C_A + k_6 C_G C_B,$$

$$\frac{dC_A}{dt} = -k_1 C_T C_A + k_2 C_D C_B
- k_3 C_D C_A + k_4 C_M C_B
- k_5 C_M C_A + k_6 C_G C_B,$$

$$\frac{dC_G}{dt} = k_5 C_M C_A - k_6 C_G C_B.$$
(6)

The initial concentrations for the state system are: $C_T(0) = C_{T_0}, C_A(0) = C_{A_0} \text{ and } C_B(0) = 0, C_D(0) = 0, C_M(0) = 0, C_G(0) = 0 \text{ and } t_f \text{ is the final time. The system (6) can be written in compact form as:}$

$$\frac{dC_i}{dt} = f_i(t, x, T, F), \tag{7}$$

where f_i (i=1,...,6) are the right sides of system (6) and C_i (i=1,...,6) are the state variables representing the concentration of each components and temperature T, Stirring F are the two control parameters.

3.1 Optimality of the system

The Hamiltonian is taken as:

$$H(\xi_i(t), C_i(t), T, F) = \sum_{i=1}^{6} \xi_i f_i(x, T, F), \ i = 1, \ 2, \ \dots (8)$$

where ξ_i (i = 1, ..., 6) are the adjoint variables. The adjoint equations satisfy the following relations:

$$\frac{d\xi_i}{dt} = -\sum_{j=1}^6 \xi_j(\frac{\partial f_i}{\partial C_i}), \ i = 1, ..., 6.$$
(9)

Using equation (8) and (9) we obtain the following

adjoint system:

$$\frac{d\xi_1}{dt} = -\xi_1(-k_2C_D - k_4C_M + k_6C_G) + \\
\xi_2k_2C_D + \xi_3(-k_2C_D - k_3C_A + k_4C_M) \\
-\xi_4(-k_4C_M + k_6C_G) + \xi_5k_2C_D + \xi_6k_6C_G$$

$$\frac{d\xi_2}{dt} = -\xi_1k_1C_A + \xi_2k_1C_A - \xi_3k_1C_A + \xi_5k_1C_A$$

$$\frac{d\xi_3}{dt} = \xi_1(k_2C_B - k_3C_A) - \xi_2k_2C_B + \xi_3(k_2C_B + k_3C_A) - \xi_4k_3C_A - \xi_5(k_2C_B - k_3C_A)$$

$$\frac{d\xi_4}{dt} = -\xi_1(-k_4C_B + k_5C_A) - \xi_3k_4C_B - \xi_4(-k_4C_B - k_5C_A) - \xi_5k_4C_B - \xi_6k_5C_A$$

$$\frac{d\xi_5}{dt} = -\xi_1(k_1C_T + k_3C_D + k_5C_M) + \xi_2k_1C_T$$

$$-\xi_3(k_1C_T - k_3C_D)$$

$$-\xi_4(k_3C_D - k_5C_M) - \xi_5(-k_1C_T - k_3C_D - k_5C_M) + \xi_6k_6C_B$$

$$\frac{d\xi_6}{dt} = \xi_1k_6C_B - \xi_4k_6C_B + \xi_6k_6C_B.$$
(10)

The boundary conditions for the adjoint variable are $\xi_1(t_f) = 1$, and $\xi_i(t_f) = 0$ for i=2, 3, 4, 5, 6.

Using the optimality condition, we have

$$\frac{dH}{dT^*} = 0 \text{ and } \frac{dH}{dF^*} = 0.$$
(11)

Since, temperature (T) and stirring (F) are given in

Table 1: Values of parameters used in numerical calculation [11].

Parameters	Value	Parameters	Value
α_1	3.92e7	β_1	6614.83
α_2	5.77e5	β_2	4997.98
α_3	5.88e12	β_3	9993.96
α_4	0.098e10	β_4	7366.64
$lpha_5$	5.35e3	β_5	3231.18
$lpha_6$	2.15e4	β_6	4824.87

implicit forms. Thus, the derivative of Hamiltonian, with respect to T and F, can not be calculated directly. So, the optimal control pair $(T^*(t), F^*(t))$ can be obtained by maximizing the Hamiltonian, $H(t, T^*, F^*)$, using the following manner:

The derivative of Hamiltonian, with respect to T and



Figure 1: Concentration trajectories of all components at constant temperature (313K) taking stirrer speed F=600 rpm, $C_T(0) = 1 \ mol/L$, $C_A(0) = 6 \ mol/L$ and other parameters as given in Table 1.

F, are given respectively by

$$\frac{dH}{dT} = \sum_{i=1}^{6} \frac{\partial H}{\partial C_i} \frac{dC_i}{dT} + \sum_{i=1}^{6} \frac{\partial H}{\partial \xi_i} \frac{d\xi_i}{dT}$$
$$= \sum_{i=1}^{6} \frac{\partial H}{\partial C_i} \theta_i + \sum_{i=1}^{6} \frac{\partial H}{\partial \xi_i} \phi_i$$
(12)

and

$$\frac{dH}{dF} = \sum_{i=1}^{6} \frac{\partial H}{\partial C_i} \frac{dC_i}{dF} + \sum_{i=1}^{6} \frac{\partial H}{\partial \xi_i} \frac{d\xi_i}{dF}$$
$$= \sum_{i=1}^{6} \frac{\partial H}{\partial C_i} \alpha_i + \sum_{i=1}^{6} \frac{\partial H}{\partial \xi_i} \beta_i.$$
(13)

Here $\theta_i = \frac{dC_i}{dT}$ and $\phi_i = \frac{d\xi_i}{dT}$, $\alpha_i = \frac{dC_i}{dF}$ and $\beta_i = \frac{d\xi_i}{dF}$. The values of θ_i , ϕ_i , α_i and β_i are calculated by the following properties:

$$\frac{d}{dT}\left(\frac{dC_i}{dt}\right) = \frac{d}{dt}\left(\frac{dC_i}{dT}\right) = \frac{d\theta_i}{dt},$$
(14)

$$\frac{d}{dT}\left(\frac{d\xi_i}{dt}\right) = \frac{d}{dt}\left(\frac{d\xi_i}{dT}\right) = \frac{d\phi_i}{dt},$$
(15)

$$\frac{d}{dF}\left(\frac{dC_i}{dt}\right) = \frac{d}{dt}\left(\frac{dC_i}{dF}\right) = \frac{d\alpha_i}{dt} \text{ and } (16)$$

$$\frac{d}{dF}\left(\frac{d\xi_i}{dt}\right) = \frac{d}{dt}\left(\frac{d\xi_i}{dF}\right) = \frac{d\beta_i}{dt}.$$
 (17)

The differential equations for θ_i , ϕ_i , α_i and β_i are given below in compact form as

$$\frac{d\theta_i}{dt} = F(C_i, \theta_i, T, F),$$

$$\frac{d\phi_i}{dt} = G(C_i, \phi_i, \theta_i, T, F),$$

$$\frac{d\alpha_i}{dt} = F(C_i, \theta_i, T, F),$$

$$\frac{d\beta_i}{dt} = G(C_i, \phi_i, \theta_i, T, F).$$
(19)

As for example, the differential equation for θ_1 is given respectively by the following equation:

$$\frac{d\theta_1}{dt} = \frac{dk_1}{dT}C_TC_A + k_1\theta_2C_A + k_1C_T\theta_5 - \left(\frac{dk_2}{dT}C_DC_B + k_2\theta_3C_B + k_2C_D\theta_1\right) + \frac{dk_3}{dT}C_DC_A + k_3\theta_3C_A + k_3C_D\theta_5 - \left(\frac{dk_4}{dT}C_MC_B + k_4\theta_4C_B + k_4C_M\theta_1\right) + \frac{dk_5}{dT}C_MC_A + k_5\theta_4C_A + k_5C_M\theta_5 - \left(\frac{dk_6}{dT}C_GC_B + k_6\theta_6C_B + k_6C_G\theta_1\right)$$

and the differential equation for ϕ_1 is:

$$\begin{aligned} \frac{d\phi_1}{dt} &= \phi_1 k_2 C_D + \xi_1 \frac{dk_2}{dT} C_D + \\ &\quad \xi_1 k_2 \theta_3 + \phi_1 k_4 C_M + \xi_1 \frac{dk_4}{dT} C_M + \xi_1 k_4 \theta_4 \\ &\quad + \phi_1 k_6 C_G + \xi_1 \frac{dk_6}{dT} C_G + \xi_1 k_6 \theta_6 \\ &\quad + \phi_2 k_2 C_D + \xi_2 \frac{dk_2}{dT} C_D + \\ &\quad \xi_2 k_2 \theta_3 + \phi_3 k_2 C_D + \\ &\quad \xi_3 \frac{dk_2}{dT} C_D + \xi_3 k_2 \theta_3 + \phi_3 k_2 C_A + \\ &\quad \xi_3 \frac{dk_2}{dT} C_A + \xi_3 k_2 \theta_5 - (\phi_3 k_4 C_M \\ &\quad + \xi_3 \frac{dk_4}{dT} C_M + \xi_3 k_4 \theta_4) + \phi_4 k_4 C_M + \\ &\quad \xi_4 \frac{dk_4}{dT} C_M + \xi_4 k_4 \theta_4 - (\phi_4 k_6 C_G \\ &\quad + \xi_4 \frac{dk_6}{dT} C_G + \xi_4 k_6 \theta_6) \\ &\quad + \phi_5 k_2 C_D + \xi_5 \frac{dk_2}{dT} C_D + \xi_5 k_2 \theta_3 + \phi_6 k_6 C_G \\ &\quad + \xi_6 \frac{dk_6}{dT} C_G + \xi_6 k_6 \theta_6. \end{aligned}$$

$$= \beta_{1}k_{2}C_{D} + \xi_{1}\frac{dk_{2}}{dF}C_{D} + \xi_{1}k_{2}\alpha_{3} \\ +\beta_{1}k_{4}C_{M} + \xi_{1}\frac{dk_{4}}{dF}C_{M} + \xi_{1}k_{4}\alpha_{4} + \\ \beta_{1}k_{6}C_{G} + \xi_{1}\frac{dk_{6}}{dF}C_{G} + \xi_{1}k_{6}\alpha_{6} \\ +\beta_{2}k_{2}C_{D} + \xi_{2}\frac{dk_{2}}{dF}C_{D} + \xi_{2}k_{2}\alpha_{3} \\ +\beta_{3}k_{2}C_{D} + \xi_{3}\frac{dk_{2}}{dF}C_{D} + \xi_{3}k_{2}\alpha_{3} + \\ \beta_{3}k_{2}C_{A} + \\ \xi_{3}\frac{dk_{2}}{dF}C_{A} + \xi_{3}k_{2}\alpha_{5} - (\beta_{3}k_{4}C_{M} \\ +\xi_{3}\frac{dk_{4}}{dF}C_{M} + \xi_{3}k_{4}\alpha_{4}) + \\ \beta_{4}k_{4}C_{M} + \\ \xi_{4}\frac{dk_{4}}{dF}C_{M} + \xi_{4}k_{4}\alpha_{4} - (\beta_{4}k_{6}C_{G} \\ +\xi_{4}\frac{dk_{6}}{dF}C_{G} + \xi_{4}k_{6}\alpha_{6}) + \\ \beta_{5}k_{2}C_{D} + \\ \xi_{5}\frac{dk_{2}}{dF}C_{D} + \xi_{5}k_{2}\alpha_{3} + \beta_{6}k_{6}C_{G} \\ +\xi_{6}\frac{dk_{6}}{dF}C_{G} + \xi_{6}k_{6}\alpha_{6}. \end{cases}$$

 $\frac{d\beta_1}{dt}$

3.2 Solution technique for the OCP

The differential equation for α_1 is given by the following equation:

$$\frac{d\alpha_1}{dt} = \frac{dk_1}{dF} C_F C_A + k_1 \alpha_2 C_A + k_1 C_F \alpha_5 - (\frac{dk_2}{dF} C_D C_B + k_2 \alpha_3 C_B + k_2 C_D \alpha_1) + \frac{dk_3}{dF} C_D C_A + k_3 \alpha_3 C_A + k_3 C_D \alpha_5 - (\frac{dk_4}{dF} C_M C_B + k_4 \alpha_4 C_B + k_4 C_M \alpha_1) + \frac{dk_5}{dF} C_M C_A + k_5 \alpha_4 C_A + k_5 C_M \alpha_5 - (\frac{dk_6}{dF} C_G C_B + k_6 \alpha_6 C_B + k_6 C_G \alpha_1),$$

and the differential equation for β_1 is:

Finally, we solve the system of equations (18) numerically for θ_i with initial conditions, $\theta_i(0) = 0$, $\alpha_i(0) =$ 0, i=1, ...,6. On the other hand, to compute ϕ_i , we solve the system of equation (19) by backward integration along with RKF method with the is boundary conditions are, $\phi_i(t_f) = 0, \ \beta_i(t_f) = 0, \ i=1,...,6$. We solve The system of equation (10) by backward integration along with RKF method using RKF45 solver. In order to obtain a solution, some iterative techniques including the shooting method and steepest descent of the Hamiltonian method are used. To reduce the computational intensity, the optimal stirring and temperature profile for the system is achieved by using the approach proposed by Diwekar and Benavides [11], where the maximum principle is used. This algorithm starts with the initial estimate stirring and temperature F(t) and T(t). Subsequently, Equations (6) and (10) are solved. Next, the values of $\frac{dH}{dF}$ and $\frac{dH}{dT}$ at each time are computed and then the convergence criterion:

$$\frac{dH}{dF}$$
 < tolerance $\frac{dH}{dT}$ < tolerance,

are satisfied. If the convergence criterion is not



Figure 2: Concentration of biodiesel is plotted as a function of temperature taking stirrer speed as F=600 rpm.



Figure 3: Simultaneous effect of temperature and stirring on biodiesel yield.

satisfied, the stirring F(t) and temperature T(t) are updated in the following manner:

$$N^{new} = N^{old} + M \frac{dH}{dN} T^{new} = T^{old} + M \frac{dH}{dN}.$$

The value of M is a suitable constant that can be small enough so that no instability will result, or large enough for rapid convergence.

4 Results and Discussion

In this section, we solved the model equations and optimal control problem numerically in Matlab to understand the behavior of the system. In Figure 1, the system behaviour is shown at constant temperature 313 K and a stirring of 600 rpm. Initially rate of reaction for the formation of biodiesel is quite high and subsequently, concentration of triglycerides is decreasing along with declining nature for the concentration of DG and MG. However, after 60 min there is no significant change in concentration of biodiesel. This situation also happens with the other components.

Temperature and stirring dependency of biodiesel yield is presented in Figure 2. As reaction temperature and stirring increases, yield of biodiesel is also increased. But, high temperature should be avoided because at the high temperature, alcohol may vaporize which makes the process complicated.

Figure 4 and 5 presents the profiles of Hamiltonian



Figure 4: Solution of optimal control problem: Temperature, stirring and biodiesel concentration profile for all iterations.

derivatives $(\frac{dH}{dT}, \frac{dH}{dF})$ and Temperature per iteration respectively. From these figures, it can be seen that as the profiles of the gradients $(\frac{dH}{dT})$ decrease, the temperature profiles increase. It is observed that initially higher temperature is required for smooth biodiesel production that is also evident for experimental works. Hamiltonian derivative decreases with time that means temperature goes to its optimum value for maximum production of biodiesel that is indicated clearly by the Hamiltonian derivative. The solution technique presented in this paper proposes that the iterations proceed until the gradients reach a specific tolerance, thus, we decided a tolerance less than 0.001. Five iterations are considered to be sufficient. Moreover, it can be seen from Figure 4 that temperature profiles are not changing in the last iterations.

Figure 6 illustrates the concentration profiles of biodiesel for two cases, case 1: with optimal temperature, and case 2: without control i.e. with fixed temperature. Here we are comparing the concentration profiles at constant temperature at 60 min of reaction time. In this figure, the effect of optimal stirring and temperature as a control pair is reflected on the concentration of biodiesel. In case 1, the concentration of biodiesel at optimal control reaches its maximum value, 2.75 mol/L while in case 2, the maximum concentration is 2.50 mol/L yield is obtained. Thus, biodiesel yield is increased by 10% approximately, when the optimal control on temperature is employed. On the other hand, it is also seen that the biodiesel obtained the maximum concentration in 1 hour.



Figure 5: Solution of optimal control problem: Derivative of Hamiltonian for all iterations.



Figure 6: Optimal stirring, optimal temperature and optimal biodiesel profiles are plotted (in final iteration). Biodiesel concentrations are compared for two cases: with optimal control and with out control.

5 Conclusion

In this article, a system of biodiesel production through transesterification is considered and combined effect of temperature and stirring on biodiesel yield is studied by developing a mathematical model. A two control parameter (stirring and temperature) optimal control problem is proposes so that maximum biodiesel can be obtained. The optimal control problem is solved using maximum principle. Optimal profiles for the control parameters are obtained numerically by efficient numerical methods. Concentration of biodiesel is increased significantly when the optimal control on stirring and temperature are employed. In this way, optimal control approach provides improvement to the effectiveness of biodiesel production process.

Acknowledgements: Its a pleasure to acknowledge Dr. Pahola T. Benavides, Argonne National Laboratory Lemont, Illinois and Dr. Urmila Diwekar, Department of Industrial Engineering, University of Illinois, Chicago, USA for technical support.

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