# Improvement of Biodiesel Production in Batch Transesterification Process

A. Chanpirak, W. Weerachaipichasgul

***Abstract*— Biodiesel production improvement in batch transesterification process can be achieved by 2 strategies: optimization and nonlinear model based control. To modify optimal temperature set point, optimization strategy is first performed with objective function to provide optimal reactor temperature: maximum product in minimum batch time (0.8481 mol/L in 51 minutes). An extended Kalman filter, (EKF) has been designed to estimate the uncertain parameter and unmeasurable states variable. Generic model control algorithm (GMC), nonlinear model based control, has been implemented to drive the reactor temperature tracking to the desired profile. In this work, improvement of batch transesterification process under uncertain overall heat transfer coefficient parameter has been proposed. Simulation results demonstrate that the EKF can provide good estimates. Moreover, the control performance of GMC is better than that of PID. As a result, the GMC with EKF is still robust and applicable in real plants.**

***Index Terms*— Biodiesel, Trtanesterification, Batch reactor, Optimization, Generic model control,**

## I. INTRODUCTION

Diesel fuel is important energy resources in the transport section that is important parameters to drive economies growing. Although demand for diesel fuel is rising, petroleum reserves are inversed. In addition, the climate change is environment problems that have been concerned using petrodiesel. Biodiesel is attended to be new alternatives energy resources that secure the future energy supplies. Biodiesel can be produced form vegetable oils, waste cooking oils, and animal fats [1-3]. Biodiesel that made from vegetable oils, consisting of methyl esters of fatty acids, is obtained by transesterification reaction between triglycerides and methanol in presence of a catalyst [4-6].

Transesterification reaction can be run in a batch reactor because it can adapt to small volume production and it can provide to scale-up processes from laboratory experiment to industrial manufacturing. Furthermore, it is also suitable for the manufacturing of difficult processes to convert to continuous operations. The yield of biodiesel affected by four main factor; the molar ratio of reactants, reaction time, catalyst, and reaction temperature [7]. The molar ratio between alcohol and triglycerides is 6:1 in alkali catalyst that is reported to guarantee the complete conversion to product esters in short time. An excess time in the reaction can give the falling conversion of triglycerides because fatty acids will form to soap. Increase concentration of catalyst helps to improve conversion of triglycerides (increasing the yield of biodiesel). The most commonly used catalyst is sodium hydroxide. An increase in reaction temperature as an increase reaction rate and reaction time is shortened. However, the yield of biodiesel is possible to decrease when temperature increase because of the saponification reaction of triglycerides appearing. Then temperature is considered to be a control parameter to improve biodiesel yield [7].

In the batch transesterification process, the dependence of the reaction kinetics and temperature can be categorized by nonlinear behavior and time-varying system and non-stationary. As a result that, control of transesterification process is more challenging.

Optimal temperature control by model based control in batch processes have been used in many cases. A Generic model control (GMC) is one of model based control that is effective to control. Moreover, the algorithm of GMC is simpler than that the other model based control do [8-10]. On the other hand, batch transesterification process has not been much attended [11-13]. However, model based control needs the state variables and/or parameters that some of variables cannot know exactly or sometime the measured variables with time delay will be introduced. One of several estimation techniques that has been widespread is an extended Kalman filter (EKF) [8-10, 14].

In this work, to modify optimal set point profile, an optimization technique based on a sequential optimization approach is applied. The optimization problem to improve biodiesel in the batch tranesterification reactor is maximum product in minimum batch time. The EKF has been designed to estimate the uncertain parameters and unmeasurable states variable. GMC is applied to tracking the desire profile. However, the control performances of GMC are compared with a conventional control technique under the nominal and mismatch case.

## II. BATCH TRANSESTERIFICATION REACTOR

A reactor system is presented in Fig.1 which consists of a reactor and jacket system [8-10]. Biodiesel is produced by the transesterification reaction between lipids (animal fats or plants), and short chain of alcohol (methanol or ethanol) in a base or acid catalyst. In this study, vegetable oil, methanol and a homogenous alkaline catalyst [15] are studied.

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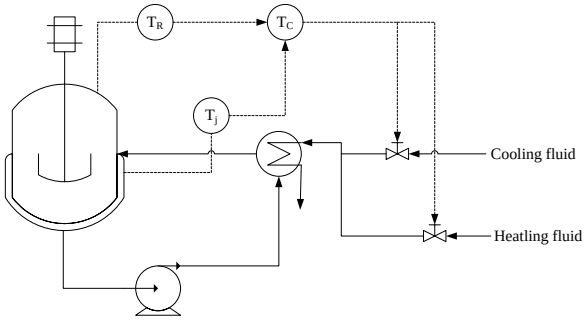


Fig. 1. Batch reactor system

Biodiesel, free fatty acid methyl ester (FAME), can be formed by the transesterification reaction of triglyceride (TG) with methanol (A) that consist of three sequential and reversible reactions.Free fatty acid methyl ester (RiCOOCH3) is produced in each step. The sequence converting of substances in this process consists of triglyceride to diglycerides (DG), diglycerides to monoglycerides (MG), and monoglycerides to glycerol (GL).

The reaction steps are given;

$$TG + CH_3OH \underset{k_2}{\overset{k_1}{\Leftrightarrow}} DG + R_1COOCH_3$$

$$DG + CH_3OH \underset{k_4}{\overset{k_3}{\Leftrightarrow}} MG + R_2COOCH_3$$

$$MG + CH_3OH \underset{k_6}{\overset{k_5}{\Leftrightarrow}} GL + R_3COOCH_3$$

Overall reaction;

$$TG + 3CH_3OH \Leftrightarrow GL + 3RCOOCH_3$$

The assumption for the mathematical model can be obtained by mass balance for each components in batch reactor following by Noureddini and Zhu [16] and Richard et al. [17]; the reactions are first-order, reactor is operated under atmospheric pressure.
Material balances

$$\frac{dC_{TG}}{dt} = -k_1 C_{TG} C_A + k_2 C_{DG} C_E \quad (1)$$

$$\frac{dC_{DG}}{dt} = k_1 C_{TG} C_A - k_2 C_{DG} C_E - k_3 C_{DG} C_A + k_4 C_{MG} C_E \quad (2)$$

$$\frac{dC_{MG}}{dt} = k_3 C_{DG} C_A - k_4 C_{MG} C_E - k_5 C_{MG} C_A + k_6 C_{GL} C_E \quad (3)$$

$$\frac{dC_E}{dt} = k_1 C_{TG} C_A - k_2 C_{DG} C_E + k_3 C_{DG} C_A - k_4 C_{MG} C_E + k_5 C_{MG} C_A - k_6 C_{GL} C_E \quad (4)$$

Energy balances

$$\frac{dT_R}{dt} = \frac{M_R (Q_R + Q_j)}{V \rho_R c_{m,R}} \quad (5)$$

$$\frac{dT_j}{dt} = \frac{F_j \rho_j c_w (T_{jsp} - T_j) - Q_j}{V_j \rho_j c_w} \quad (6)$$

$$Q_R = -V \Delta H_R r \quad (7)$$

$$Q_j = UA(T_j - T_R) \quad (8)$$

$$r = \frac{dC_E}{dt} \quad (9)$$

where $C_{TG}$, $C_{DG}$, $C_{MG}$, $C_E$, $C_A$, $C_{GL}$ are concentrations of triglycerides, diglycerides, monoglycerides, methyl ester, methanol, and glycerol, respectively and $T_R, T_j$ and $T_{jsp}$ are the reactor, jacket, and set point value of the jacket temperatures, respectively.
The reaction rate constant ($k_i$) can be expressed by the Arrhenius equation;

$$k_i = k_{0,i} \exp\left(\frac{E_{a,i}}{RT_R}\right) \qquad for\ i=1,\ldots,6 \quad (10)$$

where $k_{0,i}$ is pre-exponential factor and $E_{a,i}$ is activation energy of component "i". Moreover, R is universal gas constant. The process parameters and initial condition values in this work are presented in Table I, and Table II.

TABLE I
THE PARAMETERS IN ARRHENIUS EQUATION

| $k_{0,i}$ | m³/(kmol.s) | $E_{a,i}/R$ | K |
|---|---|---|---|
| $k_{0,1}$ | $3.92 \times 10^7$ | $E_{a,1}/R$ | 6614.83 |
| $k_{0,2}$ | $5.77 \times 10^5$ | $E_{a,2}/R$ | 4997.98 |
| $k_{0,3}$ | $5.88 \times 10^{12}$ | $E_{a,3}/R$ | 9993.98 |
| $k_{0,4}$ | $9.80 \times 10^9$ | $E_{a,4}/R$ | 7366.64 |
| $k_{0,5}$ | $5.35 \times 10^3$ | $E_{a,5}/R$ | 3231.18 |
| $k_{0,6}$ | $2.15 \times 10^4$ | $E_{a,6}/R$ | 4824.87 |

TABLE II
PROCESS PARAMETER AND INITIAL CONDITIONS VALUES

| | |
|---|---|
| V = 1 m³ | $c_w$ = 4.12 kJ/kg.K |
| $\rho_R$ = 860 kg/m³ | $C_{TG}$ = 0.3226 mol/L |
| $M_R$ = 391.40 kg/kmol | $C_{DG}$ = 0 mol/L |
| $c_{m,R}$ = 1277 kJ/kmol.K | $C_{MG}$ = 0 mol/L |
| $\Delta H_R$ = -1850 kJ/kmol | $C_E$ = 0 mol/L |
| UA = 450 kJ/min.K | $C_A$ = 1.9356 mol/L |
| $F_j$ = 0.348 kg/min | $C_{GL}$ = 0 mol/L |
| $V_j$ = 0.6812 m³ | |
| $\rho_j$ = 1000 kg/m³ | |

## III. OPTIMIZATION PROBLEM FORMULATION

For batch transesterification process, an optimization problem is transformed into a nonlinear programming (NLP) problem by a sequential approach and uses the control vector parameterization (CVP) technique to ask this problem which is solved using a SQP-based optimization technique and process model is integrated by using the explicit Runge-Kutta Fehlberg (RKF) method. The optimal rector temperature profile is determined by following the objective function; maximum the amount of product of $C_E$ and minimum batch time subject to the process model that

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described by the system of differential algebraic equations (DAEs) presented in (1) - (10). Moreover, specified product is greater than 0.83 mol/L. Reactor temperature is selected to be the decision variable into a finite set in which a piecewise constant function is utilized. The optimization problem formulation can be written as:

$$\underset{T_R, t_f}{Max} \quad \left(C_E / t_f\right) \tag{11}$$

Subject to

$f(t, \dot{x}, x, u, v) = 0$ (Model equation)
$f(t = 0, x(0), u, v) = 0$ (Equality constraints)
$C_E(t_f) > 0.83$ (Inequality constraints)
$t_f < 100$ (Inequality constraints)
$293.15 \le T_R \le 363.15$ (Inequality constraints)

## IV. Generic model control (GMC) with Extended Kalman Filter (EKF) Estimator

The general formula of GMC algorithm are

$$\frac{dx}{dt} = f(x, p, t) + g(x, t)u \tag{12}$$

$$y = h(x) \tag{13}$$

where *x*, *y* are the vectors of state output variable, u is input variables, *p* is a process parameters, and *f*, *g*, and *h* are generally nonlinear functions. The general form of GMC algorithm can be written as

$$\frac{dy}{dt} = K_1(y_{sp} - y) + K_2 \int_0^{t_f} (y_{sp} - y)dt \tag{14}$$

where $K_1$ and $K_2$ are the tuning parameters for the GMC [18]. The GMC control law is represented by

$$u = \frac{\left[K_1(y_{sp} - y) + K_2 \int (y_{sp} - y)dt - \left(dh/dt\right) f(x, p, t)\right]}{\left(dh/dt\right) g(x, t)} \tag{15}$$

Application GMC controller for temperature control in the batch reactor, the energy balance around the reactor is required (the reactor temperature and the jacket temperature). The assumption that the amount of the heat accumulated in the walls of the reactor is negligible compared with the heat transferred in the reactor. In this study, the reactor temperature is the controlled variable and the jacket temperature is the manipulated variable, so we obtain the GMC control algorithm as

$$T_j = T_R + \frac{M_R c_{m,R}}{UA}(K_1(T_{R,sp} - T_R) + K_2 \int_0^t (T_{R,sp} - T_R)dt - \frac{Q_R}{UA} \tag{16}$$

The integral term in (16) can be approximated by numerical integration. This leads to the discrete-time form of the GMC algorithm as given

$$T_j(k) = T_R(k) + \frac{M_R c_{m,R}}{UA}(K_1(T_{R,sp} - T_R(k)) + K_2 \sum_0^k (T_{R,sp} - T_R(k))\Delta t - \frac{Q_R(k)}{UA} \tag{17}$$

where Δt is the sampling time to be equal to the frequency of the temperature measurement. The actual jacket temperature $T_j(k)$ in (17) is not a set point or inlet jacket temperature. The effect of a dynamic of the jacket control system is accommodated by approximation that the first order model with time constant, $\tau_j$. The jacket temperature set point can be computed by:

$$T_{jsp}(k) = T_j(k-1) + \tau_j \left( \frac{T_j(k) - T_j(k-1)}{\Delta t} \right) \tag{18}$$

In this work, the jacket temperature is bounded between 293.15 K and 363.15 K. The tuning parameters of GMC are given in Table III.

In (17), the heat released ($Q_R(k)$) that cannot be measured directly is needed in the GMC algorithm. As a result, the EKF estimator is applied to estimated the heat released, $Q_R(k)$. Moreover, the EKF estimator is also used to provide the estimates of process parameters to handle process-model mismatches [8-10, 14] . In this work, the heat released and the overall heat transfer coefficient are studied. Other parameters can be also considered in the future work. The design of the state estimation bases on simplified mathematical models are given by

$$\frac{dT_R}{dt} = \frac{M_R Q_R + UA(T_j - T_R)}{V \rho_R c_{m,R}} \tag{19}$$

$$\frac{dT_j}{dt} = \frac{F_j \rho_j c_j (T_{jsp} - T_j) - UA(T_j - T_R)}{V_j \rho_j c_j} \tag{20}$$

$$\frac{dN}{dt} = -bNT_R \tag{21}$$

$$\frac{dQ_R}{dt} = N\frac{dT_R}{dt} + T_R \frac{dN}{dt} \tag{22}$$

$$\frac{db}{dt} = 0 \tag{23}$$

$$\frac{dUA}{dt} = 0 \tag{24}$$

The initial condition and parameters to support in the EKF estimator is presented in Table III. Detailed regarding the EKF algorithm is given in [8-10, 14].

The control structure proposed in this work is shown in Fig. 2 which consists of the optimization to carry out to compute the optimal reactor temperature. And then, GMC controller with EKF estimator is used to control the reactor temperature to following the desired profile using a jacket temperature as a manipulated variable.

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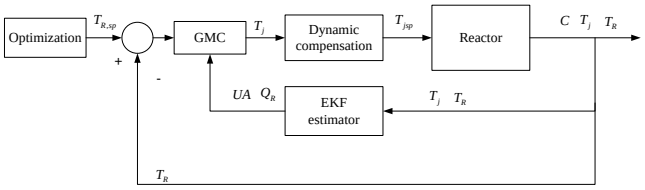


Fig. 2. Control structure: GMC controller with EKF estimator

## V. Simulation Results

In simulation study, model equations for transesterification batch reactor are integrated using the explicit Runge-Kutta Fehlberg (RKF) method with a step size (6 seconds). The optimal temperature at 347 K can be achieve by solving an optimization problem with an objective function for maximum product in minimum batch time; 0.8481 mol/L in 51 minutes. The measured variables (reactor and jacket temperatures) at every sampling time (24 seconds) are introduced to estimator and controller to control the reactor temperature tracking the desired values by manipulating the jacket temperature. Tuning parameters of the GMC controller ($K_1$, and $K_2$) are 0.005, and $1\times 10^{-7}$, respectively and tuning parameters of the PID controller is tuned to provide identical control response of $K_c$ = 10, $\tau_i$ = 60, and $\tau_D = 1\times10^{-5}$.

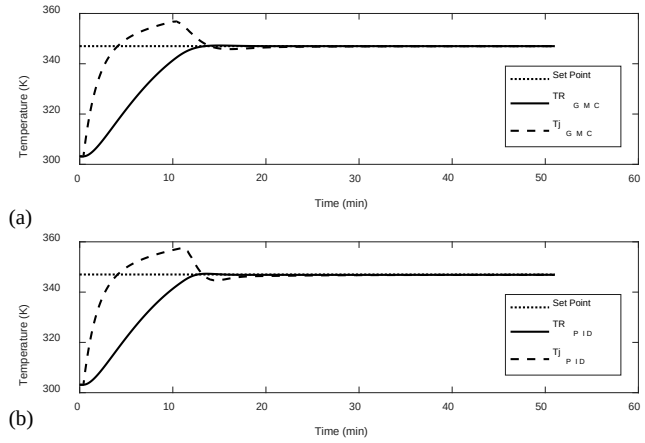


Fig. 3. Control response in nominal case: (a) GMC and (b) PID control

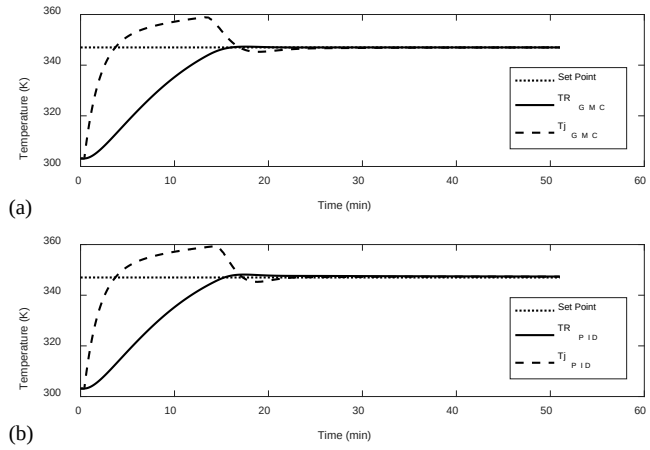


Fig. 4. Control response in mismatch case of UA -30%: (a) GMC and (b) PID control

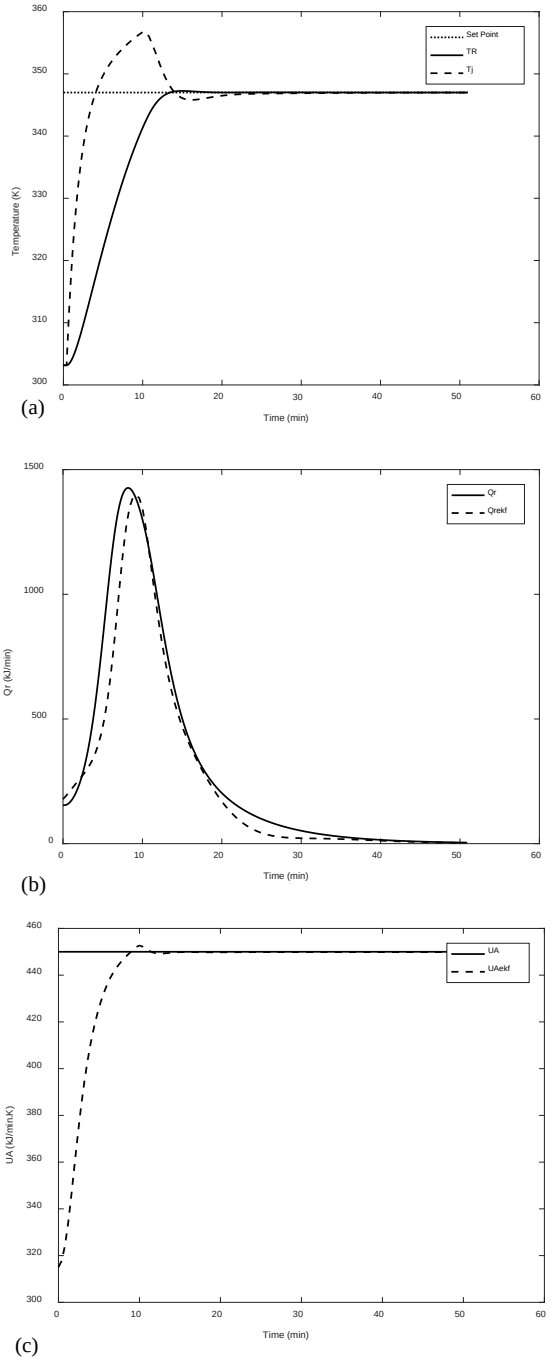


Fig. 5. (a) Control response of GMC with EKF in mismatch case of UA -30% . (b) Estimate of Qr. (c) Estimate of UA

TABLE III
PARAMETER AND INITIAL CONDITIONS IN EKF

| | |
|---|---|
| $N(0)$ =0.5539 | $P$= diag[100 100 $2\times10^5$ $2.5\times10^7$ $1.5\times10^{-5}$ $4.5\times10^4$] |
| $b(0)$ = $5.5157\times10^{-7}$ | $Q$ = diag[100 100 1000 100 1.5 $\times10^{-5}$ $4.5\times10^4$] |
| $Q_R(0)$ = 0 | $R$ = diag([1 1] |

Comparing the control performance of GMC and PID controllers are illustrated in Fig.3 to track the desired profile in the nominal case. The control performance of controllers are summarized in the terms of integrated absolute error (IAE) as presented in Table IV. It has been observed that the control performance of GMC and PID controllers are slightly different.

The presence of plant/model mismatch in the overall heat transfer coefficient decreased 30 % from its real value. The

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control responses of the PID and MPC controllers are illustrated in Fig.4. Although the GMC and PID controllers can track the desired profile in the mismatch cases, the control performance of GMC is greater than that PID does. To improve GMC, the EKF needs to estimate the heat released and the overall heat transfer coefficient that is practiced in the GMC formular. Fig. 5 (a) shows the control response of the GMC with EKF in the mismatch case of the overall heat transfer coefficient. And the estimated response of the state and parameter are showed in Fig. 5 (b) for the heat released, and Fig. 5 (c) for the overall heat transfer coefficient. The control performances of the GMC with the EKF for state and parameter estimation are presented in Table IV.

TABLE IV
THE CONTROL PERFORMANCE OF CONTROLLERS

| Case studies | IAE |
|---|---|
| GMC in nominal case | 254.66 |
| PID in nominal case | 258.16 |
| GMC in missed math case | 311.58 |
| PID in missed math case | 329.77 |
| GMC with EKF in missed math case | 254.97 |

## VI. CONCLUSION

In this work, production of biodiesel in the batch transesterification improved by a generic model control (GMC) with EKF has been proposed. The optimal reactor temperature computed by optimization is the set point in the controllers. The EKF is incorporated into the GMC algorithm in order to estimate the heat released by measurement of reactor and jacket temperatures. The control performance of the GMC is better than that of PID. In the presence of unknown/uncertain parameters the estimator is still able to provide accurate value. As a result, the GMC with the EKF is still robust and applicable in real plants.

*Nomenclature*:

$c_{m,R}$ Molar capacity of the reactor content,

$\Delta H_R$ Heat of reaction,

$M_R$ Molar mass of the reactor,

$V$ Reactor volume,

$\rho_R$ Density of the reactor content,

$UA$ Product of the heat exchanger surface of $A$ and the overall heat transfer coefficient $U$,

$c_w$ Specific heat capacity of water,

$r$ Rate of reaction

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