CFD model of single-mode microwave – assisted biodiesel production via transesterification reaction of refined palm stearin using one-way coupling method in a batch stirred-tank reactor

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Abstract

Microwave can provide the direct heat without any losses to the chemical reaction. Chemicals having the dielectric property, especially the alcohol that used to produce the biodiesel, can absorb the microwave radiation and cause molecules to vibrate and generate the heat. As a result, the reaction rate can be accelerated by nearly 4.5 times. Refined palm stearin (RPS) as the by-product of crude palm oil (CPO) production is an alternative raw material to produce biodiesel due to its lower cost. However, the feasibility of biodiesel production from RPS with single-mode microwave assistance requires investigations into the influence of physical-chemical properties including operating conditions to the momentum, heat, and mass transport. Computational Fluid Dynamic (CFD) simulation of single-mode microwave – assisted biodiesel production from RPS and methanol in the presence of NaOH-derivative catalyst using ANSYS FLUENT and HFSS programs is presented in this study. The momentum profile of the stirrer was predicted by using Multi Reference Frame (MRF) method, while the temperature profile of the batch reactor due to microwave heating was also investigated by One-way Coupling method. Then, the concentration profile was obtained from the beginning of reaction until 600 s via the three-step reversible transesterification reaction mechanism.

Keywords: Biodiesel, transesterification, single-mode microwave, Refined Palm Stearin (RPS), Computational Fluid Dynamic (CFD)

1. Introduction

The invention of microwave brings the new breakthrough on chemistry application, that change the common way of heating on sample material. The conventional heating method happened from the heating source being transferred by conduction to the sample vessel, which can reach the sample either by conduction or convection depends on the phase of material [1]. This indirect heating provides slow reaction due to the long heating process. The microwave works on reverse by providing the direct heat on the sample by the electromagnetic field (EM) that transferred to its surrounding by convection and conduction. The dipole interaction due to the EM field on dielectric material cause the molecules rotate and start the intermolecular movement that make the sample to generate heat [1]. The reaction assisted by microwave heating is approximately 4-6 times faster [2-4].

The palm fruit derivatives product has great potential to be the biodiesel feedstock because of its characteristics as lipid [5]. Crude palm oil (CPO), refined-bleaching-degumming palm olein (RBDPO), palm fatty acid distillate (PFAD), palm olein, and palm stearin can be converted into biodiesel. Palm stearin came up as the attractive feedstock with lowest process cost [5].

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The biodiesel is produced via transesterification reaction in three-step reversible reaction mechanism and it is an endothermic reaction. In order to increase the product yield, its equilibrium condition needs to be shifted to the product side by increasing the reactant and the heat of reaction according to Le Chatelier principle [6]. Therefore, alcohol is typically in excess and the practical molar ratio of alcohol to oil is 6:1 or 7:1 which can increase the yield of biodiesel to be higher than 96.5% according to the standard EN 14214 [7-12]. By using the microwave as the heating source, the biodiesel product yield can be increased as shown in Table 1.

Table 1. Biodiesel yield via transesterification reaction of refined palm stearin with conventional heating vs. microwave heating

| Reference/Characteristics | Heating Method | Catalyst | Operating Conditions for Reaction | Yield (%) |
|---------------------------|----------------|----------|----------------------------------|-----------|
| Kriengyakul (2009) [8]    | Conventional Heating | NaOH catalyst | 60℃ heating stirred at 250 rpm 15 mins (900 s) | 98.3      |
| Theam (2015) [2]          | Conventional Heating | Double Metallic Methoxide | 70℃ heating without stirring 125 mins (7,500 s) | 98.0      |
| Songoen (2017) [9]        | Conventional Heating | Calcium Methoxide | 65℃ heating stirred at 750 rpm 125 mins (6,125 s) | 98.23     |
| Garnwityayee (2012) [10]  | Multi-mode Microwave Heating | NaOH catalyst | 60℃ heating stirred at 600 rpm 50 s | 97.5      |
| Kangsadan (2017) [11]     | Conventional Heating | NaOH-derivative catalyst | 60℃ heating stirred at 600 rpm 70 mins (3,600 s) | 96.8      |
| Wanking (2019) [12]       | Single-mode Microwave Heating | NaOH-derivative catalyst | 60℃ heating stirred at 600 rpm 400 s | 98.19     |

Computational Fluid Dynamics (CFD) as a reliable tool of numerical method to solve complex dimensional and time-dependent problems. ANSYS 18.2 program will be employed to solve the single-mode microwave-assisted biodiesel production with one-way coupling method between HFSS and FLUENT modules. Later, the simulated results will be validated with ones obtained from laboratory-scale experiments.

2. Materials and Methods

2.1. Model assumptions

The workflow of ANSYS 18.2 program is illustrated on Fig. 1. The momentum profile of the stirrer was predicted by using Multi Reference Frame (MRF) method, while the heat profile of the single-mode microwave batch reactor was also investigated by One-way Coupling method.

Fig. 1. Workflow of ANSYS 18.2 program

The mixture of RPS, methanol and catalyst reactants were assumed as a single phase during the reaction. The reaction temperatures to be investigated were at 55℃, 60℃, and 65℃ at 101.325 kPa. The model was defined at the transient state until the time reach 600 s with the time step size of 0.01 s. Since the batch process was considered, there was no inlet and outlet for the single-mode microwave reactor. The rotating impeller velocity was at 600 rpm with microwave at the maximum power of 800 W at frequency of 2.45 GHz. The six species to be monitored were triglycerides (TG), diglycerides (DG),
monoglycerides (MG), methanol (ROH), biodiesel (FAME), and glycerol (GLY). The flow regime was assumed to be laminar.

2.2. Model governing equations

The model governing equations are as follows:

Mass conservation equation

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{v}) = S_m$$

(1)

where $\rho$ is the liquid density, $\vec{v}$ is the velocity vector, and $S_m$ is the mass added to the continuous phase from the dispersed second phase and any user-defined sources [13].

Momentum conservation equation

$$\frac{\partial}{\partial t} (\rho \vec{v}) + \nabla \cdot (\rho \vec{v} \vec{v}) = -\nabla p + \nabla \cdot (\tau \vec{e}ff) + \rho \vec{g} + \vec{F}$$

(2)

where $p$ is the static pressure, $\tau \vec{e}ff$ is the stress tensor, $\rho \vec{g}$ is the gravitational body force, and $\vec{F}$ is the external body forces [13].

Energy conservation equation

$$\frac{\partial}{\partial t} (\rho E) + \nabla \cdot (\vec{v} (\rho E + p)) = \nabla \cdot \left( k_{eff} \nabla T - \sum_j h_{ff} \vec{j}_j + \left( \tau_{eff} \cdot \vec{v} \right) \right) + S_h$$

(3)

where $k_{eff}$ is the effective conductivity (thermal conductivity), $\vec{j}_j$ is the diffusion flux of species $j$, and $S_h$ is the energy generation [14].

Species conservation equation

$$\frac{\partial}{\partial t} (\rho Y_i) + \nabla \cdot (\rho \vec{v} Y_i) = -\nabla \cdot \vec{j}_i + R_i + S_i$$

(4)

where $Y_i$ is the concentration species $i$, $\vec{j}_i$ is the diffusion flux of species $i$, $R_i$ is the net rate of production of species $i$ by chemical reaction, and $S_i$ is the rate of creation by addition from the dispersed phase plus any user-defined sources. Since the flow regime is laminar, the laminar finite-rate model is used for this reaction mechanism [15].

$$R_i = M_{w,i} \sum_{r=1}^{N_R} \hat{R}_{i,r}$$

(5)

where $M_{w,i}$ is the molecular weight of species $i$ and $\hat{R}_{i,r}$ is the Arrhenius molar rate of creation/destruction of species $i$ in reaction $r$ and given as:

$$\hat{R}_{i,r} = (k_{f,r} \prod_{j=1}^{N} (C_{j,r})^{\eta_{j,r}} - k_{b,r} \prod_{j=1}^{N} (C_{j,r})^{\psi_{j,r}})$$

(6)

The forward reaction rate constant, as well as the reverse one, is calculated using the following form:

$$k_{f,r} = A_r T e^{-E_r/kT}$$

(7)
where $A_r$ is the pre-exponential factor in L·mol$^{-1}$·s$^{-1}$, $E_r$ is the activation energy for the reaction in J·kmol$^{-1}$, $T$ is the absolute temperature in K and $R$ is the universal gas constant in J·kmol$^{-1}$·K$^{-1}$ [15].

2.3 Reaction kinetics model

This investigation focused on the modelling and the validation of the simulation results using ANSYS 18.2 program with the laboratory-scale experiment. Therefore, the initial condition was set to the same optimal operating condition by Wanking [16] where the molar ratio of methanol to oil of 6:1 with the NaOH-derivative catalyst concentration of 2% w/v while the stirrer speed was fixed at 600 rpm were investigated varying the reaction times from 50 s to 600 s and the reaction temperatures of 55°C to 65°C. With the transesterification reaction, TG was converted into DG, MG, FAME and GLY in a three-step reaction mechanism given as follows:

\[
\begin{align*}
\text{TG} + \text{ROH} & \xrightleftharpoons[k_2]{k_1} \text{DG} + \text{FAME} \\
\text{DG} + \text{ROH} & \xrightleftharpoons[k_4]{k_3} \text{MG} + \text{FAME} \\
\text{MG} + \text{ROH} & \xrightleftharpoons[k_6]{k_5} \text{GLY} + \text{FAME}
\end{align*}
\]

(8)

(9)

(10)

Reaction rate constants ($k_1 - k_6$) can be calculated from the concentrations $[C]$ of all components and rates of reaction $\frac{d[C]}{dt}$ following second-order kinetic reactions [10, 12, 16]:

\[
\begin{align*}
\frac{d[TG]}{dt} &= -k_1[TG][ROH] + k_2[DG][FAME] \\
\frac{d[DG]}{dt} &= k_1[TG][ROH] - k_2[DG][FAME] \\
& \quad - k_3[DG][ROH] \\
& \quad + k_4[MG][FAME] \\
\frac{d[MG]}{dt} &= k_3[DG][ROH] - k_4[MG][FAME] \\
& \quad - k_5[MG][ROH] \\
& \quad + k_6[GLY][FAME] \\
\frac{d[GLY]}{dt} &= k_5[MG][ROH] - k_6[GLY][FAME] \\
\frac{d[FAME]}{dt} &= k_1[TG][ROH] - k_2[DG][FAME] \\
& \quad + k_3[DG][ROH] \\
& \quad - k_4[MG][FAME] \\
& \quad + k_5[MG][ROH] - k_6[GLY][FAME] \\
\frac{d[ROH]}{dt} &= - \frac{d[FAME]}{dt}
\end{align*}
\]

(11)

(12)

(13)

(14)

(15)

(16)

$A_r$ and $E_r$ parameters were taken from the laboratory-scale experiment by Wanking [16] and values are presented in Table 2. Then, the simulated concentration profiles were determined according to equations (5)-(7) using the ANSYS 18.2 program with one-way coupling method between HFSS and
Table 2. Activation energy and pre-exponential factor for six reaction rate constants of three-step second-order transesterification reaction mechanism for biodiesel production taken from wanking [16]

| Reaction Rate Constant | Activation energy, $E_a$ [J·kmol$^{-1}$] | Pre-exponential factor, $A$ [L·mol$^{-1}$·s$^{-1}$] |
|------------------------|----------------------------------------|---------------------------------------------|
| $k_1$                  | $3.7489\times10^7$                     | $3.03\times10^7$                           |
| $k_2$                  | $5.8627\times10^7$                     | $5.47\times10^7$                           |
| $k_3$                  | $3.7349\times10^7$                     | $4.23\times10^7$                           |
| $k_4$                  | $6.3727\times10^7$                     | $2.41\times10^7$                           |
| $k_5$                  | $4.7483\times10^7$                     | $1.87\times10^7$                           |
| $k_6$                  | $5.3306\times10^7$                     | $9.81\times10^7$                           |

2.4. Geometry

The customized laboratory-scale 2.45 GHz single-mode and continuous-type microwave-assisted reactor with maximum power of 800 W for biodiesel production was designed and developed by researchers in TGGS-CPE Novel Technologies Laboratory [11]. The microwave glass-reactor with the maximum capacity of 1 L was equipped with a mechanical rotating stirrer and the thermocouple and it located inside the microwave cavity which was made from aluminium. The geometry of microwave cavity (Fig. 2) was obtained from the previous investigation by Chawinnakorn [17]. The microwave irradiation was generated from magnetron and travelled through the waveguide which was controlled to continuously generate the single-mode microwave. Then, the microwave was absorbed by chemicals with high dielectric constant values, mainly the methanol in this reaction. Methanol has the dielectric constant value of 32.7 whereas RPS has the value in the range of 2.970 to 6.904 [1]. To prevent the leakage of microwave irradiation to the surrounding, the reflection rings were installed on the top of cavity along with the water chamber located between the reactor and the reflection ring.

Fig. 2. Microwave Cavity Geometry in ANSYS 18.2 Program

The reactor was located inside the microwave cavity without the inlet and the outlet to simulate the batch process. However, the apparatus was designed to be operated for both batch and continuous processes. The customized-glass impeller blade [11] was set in the middle of the reactor, where the rotating part surrounded it. To make the rotating flow succeed, the interface had to be defined between the rotating and stationary cell zones [17] as illustrated in Fig. 3.
2.5. Mesh generation

The mesh was generated using the curvature as the mesh size function with fine relevance center and the slow mesh transition with fine span angle center. The total number of nodes were 41,595 with 209,947 elements. With the academic license, the number of mesh must be less than 512,000 elements, however, it was sufficient for this investigation.

2.6. Boundary conditions

This reactor defined two cell zone conditions: stationary and rotating. Each cell zone has the boundary called the wall boundary. Since the wall stationary was in stationary cell zone, then the wall was set to have no motion. While the wall rotating was set to follow the motion of adjacent cell in the rotating cell zone.

3. Result and Discussion

3.1. Temperature profiles

The temperature contour profiles as illustrated in Table 3 varying the reaction times from 5 s to 600 s and at three reaction temperatures (55°C, 60°C and 65°C) demonstrated that the temperature was distributed from the center of the reactor where receiving the microwave irradiation resulting in heating and the colder temperature distribution was near the wall. This profile corresponds to the study by Chavoshani [1, 17]. This happened due to the volume loss density that affected by the electric and electromagnetic field distribution, in which the highest intensity located at the surface of the reactor in the center [17]. Thus, the microwave irradiation provided the selective heating into the mixture inside the reactor, in which can help to fasten the reaction compared to the conventional heating. As the temperature ramping to the setpoint reaction temperature, the reaction zone was approximately 50°C in which the reaction could proceed at 50 s. Then, the setpoint reaction temperature was reached and the heat was homogeneously distributed throughout the reactor after 100 s and remained at this temperature until the end of reaction (600 s).
Table 3. Temperature contour profiles in single-microwave reactor for biodiesel production using ANSYS 18.2 program with one-way coupling method between HFSS and FLUENT modules

| Time (s) | T = 55°C | T = 60°C | T = 65°C |
|---------|----------|----------|----------|
| 5       | ![Image](image1) | ![Image](image2) | ![Image](image3) |
| 25      | ![Image](image4) | ![Image](image5) | ![Image](image6) |
| 50      | ![Image](image7) | ![Image](image8) | ![Image](image9) |
| 100     | ![Image](image10) | ![Image](image11) | ![Image](image12) |
| 200     | ![Image](image13) | ![Image](image14) | ![Image](image15) |
| 300     | ![Image](image16) | ![Image](image17) | ![Image](image18) |
| 600     | ![Image](image19) | ![Image](image20) | ![Image](image21) |
3.2. Momentum profiles

The momentum vector profiles represented by the velocity profiles as illustrated in Table 4 varying the reaction times from 5 s to 600 s and at three reaction temperatures (55°C, 60°C, and 65°C) demonstrated that the mixing was homogenously distributed throughout the reactor with the intensity near the wall. Four circular loops were observed above and below the blades as typically observed with the mixing reactor [16].

Table 4. Momentum vector profiles in single-microwave reactor for biodiesel production using ANSYS 18.2 Program with one-way coupling method between HFSS and FLUENT Modules

| Time (s) | T = 55°C | T = 60°C | T = 65°C |
|---------|---------|---------|---------|
| 5       | ![Image](image1.png) | ![Image](image2.png) | ![Image](image3.png) |
| 25      | ![Image](image4.png) | ![Image](image5.png) | ![Image](image6.png) |
| 50      | ![Image](image7.png) | ![Image](image8.png) | ![Image](image9.png) |
| 100     | ![Image](image10.png) | ![Image](image11.png) | ![Image](image12.png) |
| 200     | ![Image](image13.png) | ![Image](image14.png) | ![Image](image15.png) |
| 300     | ![Image](image16.png) | ![Image](image17.png) | ![Image](image18.png) |
| 600     | ![Image](image19.png) | ![Image](image20.png) | ![Image](image21.png) |
3.3. Concentration profiles

The biodiesel concentration profiles from both experiment and simulation are shown in Fig. 4. From the laboratory-scale experiments [16], the optimal condition for the biodiesel production using the transesterification reaction in a single-mode microwave reactor was determined to be at 60°C for 400 s with the energy consumption of 0.481 kJ·g⁻¹ of biodiesel while using the conventional heating, the energy consumption was nearly 10 times higher (4.61 kJ·g⁻¹ of biodiesel). Even though the reaction was completed at 50 s in the experiment, nevertheless, the reaction was proceeded further to 400 s to ensure its purity and met the standard. On inspecting the results from simulations and the measured concentration obtained from experiments, there seems to be a “delay” in the concentration from experiments by 20 s. This cannot be confirmed since there were limitations in conducting the experiments at shorter reaction time less than 50 s. However, the simulation results were relatively closed with the overestimation by 8.33%.

Fig. 4. Concentration profiles of biodiesel product obtained from ANSYS 18.2 simulation compared with experimental data by wanking [16].

From the simulation results, final TG concentrations at 600 s were 1.68, 2.18 and 2.80 mol·m⁻³ at 55°C, 60°C and 65°C, respectively. These contrasted with the measured concentrations from experiments in which TG was expected to deplete with increasing in temperature. It should be pointed out that the measured concentrations of TG from experiments were 1.19, 0.68 and 0.59 mol·m⁻³ at 55°C, 60°C and 65°C, respectively. This might be the result of unaccounted for excess methanol in the simulation.

Fig. 5. Concentration profiles of triglycerides obtained from ANSYS 18.2 simulation compared with experimental data by wanking [16].
As the reaction proceeded, large amount of DG had been produced within the first 5 s (First Phase) and then rapidly depleted by approximately 50 s (Second Phase) and reproduced to a relatively constant value after 100 s (Third Phase) as illustrated in Fig. 6. Remaining DG concentrations at 600 s were 2.12, 2.47 and 2.81 mol·m⁻³ at 55°C, 60°C and 65°C, respectively. These trends in the first and second phases of reaction were similar to the study of Tran et al. [17]. However, the third phase was not observed in this study and in experiments by Wanking [16].

![Figure 6](image_url)

**Fig. 6.** Concentration profiles of diglycerides obtained from ANSYS 18.2 simulation compared with experimental data by Wanking [16].

The same phenomena with three phases were observed for the concentration profile of MG as illustrated in Fig. 7. As expected, higher remaining MG concentrations at 600 s were obtained as 5.80, 5.86 and 5.75 mol·m⁻³ at 55°C, 60°C and 65°C, respectively. These were in corresponding to the findings of Tran et al. [17]. However, the third phase was not observed in the experiment and final measured concentrations were much higher in the experiment as 8.0, 5.97 and 5.25 mol·m⁻³ at 55°C, 60°C and 65°C, respectively.

![Figure 7](image_url)

**Fig. 7.** Concentration profiles of monoglycerides obtained from ANSYS 18.2 simulation compared with experimental data by Wanking [16].

### 4. Conclusion

The CFD model of single-mode and continuous-type microwave reactor had been developed using the ANSYS program with one-way coupling method between HFSS and FLUENT modules to investigate the
influence of microwave irradiation with a three-step transesterification reaction mechanism for biodiesel production. The momentum profile of the mixture in the reactor showed the similarity mixing behavior and characteristic with the observation during the experiment. Moreover, the temperature distribution generated by the microwave heating confirmed that the highest value was in the center of the reactor due to the occurrence of volume loss density maximum in the surface center. Importantly, the concentration profiles of all chemical species obtained from the simulation were validated with the experimental data and showed the similar trends. The results from both laboratory-scale experiments and CFD numerical simulations will provide insight to the design of the single-mode microwave reactor for the biodiesel production with the optimal operating condition at the pilot and even the commercial scales.

Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

Ahmad Fathur Muhtadin conducted the CFD simulation and analyzed the data and wrote the paper. Phanida Wanking conducted the laboratory-scale experiment, analyzed the concentration of all species and determined the kinetics parameters. Tawiwan Kangsadan supervised the research, wrote and edited this paper. All authors had approved the final version.

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