Research Article

Modelling of Jatropha Oil Hydrocracking in a Trickle-Bed Reactor to Produce Green Fuel

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Trickle-bed reactor (TBR) modelling to produce green fuel via hydrocracking of jatropha oil using silica-alumina-supported Ni-W catalysts was performed in this research. The objectives of this study are to obtain a TBR with good heat transfer and the optimum condition for high purities of products. A two-dimensional axisymmetric model with a diameter of 0.1 m and a length of 10 m was used as a representative of the actual TBR system. Heterogeneous phenomenological models were developed considering mass, energy, and momentum transfers. The optimisation was conducted to obtain the highest green fuel purity by varying catalyst particle diameter, inlet gas velocity, feed molar ratio, and inlet temperature. The simulation shows that a TBR with an aspect ratio of 100 has achieved a good heat transfer. The diesel purity reaches 44.22% at 420°C, kerosene purity reaches 21.39% at 500°C, and naphtha purity reaches 25.30% at 500°C. The optimum condition is reached at the catalyst diameter of 1 mm, the inlet gas velocity of 1 cm/s, the feed molar ratio of 105.5, and the inlet temperature at 500°C with the green fuel purity of 69.4%.

1. Introduction

In recent years, the development of renewable fuel sources has become a focus in the science and energy sectors. Fossil fuels (such as gasoline and diesel fuel) have been the primary fuel sources in the industry and transportation sectors. However, fossil fuels are limited energy sources that can run out at any time, which could consequently induce a global energy crisis. Notably, the emission of combustion gases in the form of greenhouse gases and the price of fossil fuels are increasingly expensive. Therefore, it is necessary to develop renewable energy sources to tackle the energy crisis.

Interest in biodiesel and green fuel production has constantly increased over the past decades [1]. For example, Neste Oil in Europe and UOP Honeywell in America have built a hydrogenation-based plant to convert vegetable oil into biodiesel and green fuel [2]. The resulting green fuel should have properties similar to petroleum diesel that can be used in conventional machines.

Green fuel, which is a second-generation renewable fuel, can be produced from vegetable oils, one of which is jatropha oil. Jatropha oil is derived from *Jatropha curcas* Linn. The

plant is spread across several regions in Indonesia with different names such as kalekhe paghar, paku kase, and bindalo. Until now, in Indonesia, jatropha has been used as a traditional medicine and is planted as a living fence. Indonesian climate favours the growth of this plant; therefore, the use of jatropha as a source of renewable energy in Indonesia is possible [3].

Large-capacity green fuel production is performed using hydrotreating and hydrocracking lines, which involve three phases. The raw material is the liquid phase, the catalyst is the solid phase, and hydrogen is the gas phase. There are three types of reactors for three-phase processes: stirred slurry reactors, slurry bubble column reactors, and trickle-bed reactors (TBRs). In slurry reactors, the liquid and solid phases are mixed so that, at the end of the reaction, the separation of the products and catalyst is required as well as the additional cost for separation. Furthermore, abrasion of the catalyst may also occur in slurry reactors [4]. Slurry reactors are generally used for fermentation because the solid and liquid phases are mixed. The TBR is a three-phase reactor that uses a stream in trickle regime. In TBR, gas flows in the same or opposite direction with liquid over a packed

bed consisting of catalyst particles [5]. Therefore, TBRs are chosen based on their more efficient operation than slurry reactors. However, the problem often encountered in using TBRs is poor heat transfer. As a result, the optimisation of heat control is required [6].

The TBR model has been successfully used in various processes to predict the process variables and is still being developed till date. Giri and Majumder [7] reported the influences of concurrent downflow of air-Newtonian and non-Newtonian liquid systems on pressure drop and on its reduction in a downflow TBR. Observably, the reactor's efficiency may be influenced by the nonideality factor, which is higher than the pulse and trickle-flow regimes. Al-Naimi et al. [8] studied the hydrodynamics of TBRs in nonambient conditions for air-water and air-acetone systems, which represent pure organic liquids with low surface tension. Their study showed that the pressure drop increases with increasing superficial gas and liquid velocity, while it decreases with increasing bed temperature. Russo et al. [9] developed a dynamic model of a nonisothermal TBR for sugar hydrogenation. The resulting model can be applied to various reaction systems and tested to obtain both qualitatively and quantitatively promising results. Muharam et al. [10] modelled a TBR for the partial hydrogenation of polyunsaturated fatty acid methyl esters using a steady-state heterogeneous two-dimensional axisymmetric model comprising mass balances in each phase, energy balance, and two-phase Darcy equation-based momentum balance. Methyl linoleate was considered a polyunsaturated fatty acid methyl ester representative, and cis-methyl oleate, trans-methyl oleate, and methyl stearate were considered hydrogenated fatty acid methyl esters.

Studies on green fuel production through hydrocracking, hydrotreating, and other pathways, such as biological treatment with the Fischer-Tropsch process, have been conducted. Milbrandt et al. [11] reported that green fuel could be produced from high-temperature biomass gasification, which converted biomass to syngas and then catalytically converted it to liquid and wax products through the Fischer-Tropsch process. Attanatho [12] conducted a study on the hydrotreatment of bio-oils using a microreactor, which could achieve the complete conversion of fatty acid hydrotreating reaction at a temperature of 325°C. Muharam and Soedarsono [13] observed the hydrodeoxygenation of vegetable oils to produce renewable diesel in a TBR. It has been found that the dominant reacting compounds at temperatures above 310°C and a pressure of 15 bar are C₁₈ hydrocarbons. Anand et al. [14] conducted a study on kinetics, thermodynamics, and the reaction mechanism of green fuel production via a hydrocracking system in a fixed-bed reactor. However, few studies have developed mathematical models by utilising experimental results to simulate the process on a large scale [12, 15, 16].

This study simulates the process of jatropha oil hydrocracking in a TBR comprising several tubes in which the solid particles of the catalyst in pellet form are arranged as a so-called fixed bed. The tubes are located in a shell in which the cooling media circulate. The reactions in the tube catalyst beds are exothermic, with the reaction rate heavily

influenced by the local temperature. A good design reduces the possibility of hotspots being formed within the reactor. Therefore, the purpose of this study is to obtain a TBR model for the hydrocracking process, a reactor size with good heat transfer, and optimum conditions for high purity green fuel.

2. Modelling

To study the jatropha oil hydrocracking process in a TBR, a model was developed to represent the actual condition of a reactor system. A simulation was performed to obtain the solution of the developed model. The use of a model and a simulation can avoid risks and reduce operational costs. Furthermore, this method allows for a detailed study of the most important variables to system performance.

The system modelled in this study is illustrated in Figure 1. A triglyceride solution and gaseous hydrogen enter from the top of the reactor and flow downwards over the packed bed of catalyst particles. In this model, a tube is selected as the representative of all the tubes within a shell based on the assumption that the other tubes undergo the same phenomena as the representative. This is achieved when the temperature of the cooling medium inside the shell is uniform.

The model comprises two parts: the reactor scale and the catalyst particle scale. The reactor scale model was developed based on 2D axisymmetric geometry, with a diameter of 0.1 m and a length of 10 m, using the concept of computational fluid dynamic (CFD). This CFD method allows for the evaluation of velocity, pressure, temperature, and the species concentration of fluid flow throughout the reactor domain. In addition, this model considers momentum, mass, and energy balances. The catalyst scale model considers radial mass transfer. The kinetic model used is adopted from the kinetic model for the hydrocracking reactions developed by Anand et al. [14], where the reactants are hydrogen and triglyceride. The main products are green fuels comprising diesel (C₁₅–C₁₈), kerosene (C₉–C₁₄), and naphtha (<C₉). Jatropha oil is represented by triglycerides, while the catalyst used is Ni-W/Si₂O₃-Al₂O₃.

2.1. Momentum Transfer. The momentum transfer is modelled using Darcy's law approach for two phases, which is a continuity equation combined with Darcy's law:

$$\frac{1}{r} \frac{\partial (rc_1 U_r)}{\partial r} + \frac{\partial (c_1 U_z)}{\partial z} = \frac{1}{r} \frac{\partial}{\partial r} \left(r D_c \frac{\partial c_1}{\partial r} \right) + \frac{\partial}{\partial z} \left(D_c \frac{\partial c_1}{\partial z} \right), \quad (1)$$

where D_c is the capillary diffusion coefficient. Axial superficial velocity U_z and radial superficial velocity U_r are described by

$$U_z = -\frac{\kappa}{\mu} \frac{\partial p}{\partial z}, \quad (2)$$

$$U_r = -\frac{\kappa}{\mu} \frac{\partial p}{\partial r}, \quad (3)$$

FIGURE 1: Trickle-bed reactor modelling strategy.

where κ is the bed permeability, p is the pressure, and μ is the average viscosity.

Liquid phase content c_1 is explained as $c_1 = \rho_1 s_1$, where ρ_1 is the liquid phase density and s_1 is the liquid phase saturation.

The relationship between the saturation of Phases 1 and 2 and the vacant volume in the bed are expressed in

$$s_1 = \frac{\varepsilon_1}{\varepsilon_p}, \quad (4)$$

$$s_2 = \frac{\varepsilon_2}{\varepsilon_p}, \quad (5)$$

$$s_1 + s_2 = 1, \quad (6)$$

where ε_p is the bed porosity and ε_1 and ε_2 are the liquid and gas holdups.

The average physical properties of the mixture are given by

$$\rho = s_1 \rho_1 + s_2 \rho_2, \quad (7)$$

$$\frac{1}{\mu} = s_1 \frac{\kappa_{r1}}{\mu_1} + s_2 \frac{\kappa_{r2}}{\mu_2}. \quad (8)$$

The superficial fluid velocity is the sum of the superficial velocities of each phase, U_1 and U_2 , which are correlated using the saturation of the two fluids, as in

$$U = U_1 + U_2, \quad (9)$$

$$U_1 = s_1 U, \quad (10)$$

$$U_2 = s_2 U. \quad (11)$$

The relative permeabilities of (8) are based on the research results of Sáez and Carbonell [17]:

$$\kappa_{r1} = \delta_1^{2.43}, \quad (12)$$

$$\kappa_{r2} = s_2^{4.80}. \quad (13)$$

Parameter δ_1 is the reduced saturation of Phase 1 (liquid), which is mathematically defined by

$$\delta_1 = \frac{\varepsilon_p s_1 - \varepsilon_1^0}{\varepsilon_p - \varepsilon_1^0}. \quad (14)$$

In (14), ε_1^0 is the static holdup of Phase 1. The equation for calculating ε_1^0 follows Sáez and Carbonell [17]:

$$\varepsilon_1^0 = \frac{1}{(20 + 0.9 E \ddot{o}^*)}, \quad (15)$$

where $E \ddot{o}^*$ is the modified Eotvos number, which is expressed as follows:

$$E \ddot{o}^* = \frac{\rho_1 g d_p^2 \varepsilon_p^2}{\sigma_1 (1 - \varepsilon_p)^2}, \quad (16)$$

where d_p is the catalyst diameter and g is the gravity constant.

Bed permeability (κ) can be calculated using a slightly modified Ergun equation, as suggested by Propp et al. [18]:

$$\kappa = \left(\frac{180(1 - \varepsilon_p)^2}{d_p^2 \varepsilon_p^3} + \frac{1.8(1 - \varepsilon_p) \rho U_0}{\mu d_p \varepsilon_p^3} \right)^{-1}, \quad (17)$$

where U_0 is the inlet mixed superficial velocity.

The capillary diffusion coefficient (D_c) in (1) is a function of capillary pressure:

$$D_c = \frac{\kappa_{r1} \kappa}{\mu_1} (s_1 - 1) \frac{\partial p_c}{\partial s_1}, \quad (18)$$

where p_c is the capillary pressure, that is, the difference between the gas phase and liquid phase pressure. Capillary pressure is calculated using the equation proposed by Leverett [19]:

$$p_c = \frac{\varepsilon_p}{\kappa} \sigma J(s_1), \quad (19)$$

where σ is the gas-liquid surface tension and $J(s_1)$ is a function of capillary pressure on the liquid phase saturation (s_1). The function J is as suggested by Grosser et al. [20]:

$$J = 0.48 + 0.036 \ln \left(\frac{s_2}{s_1} \right). \quad (20)$$

2.2. Mass Transfer. The mass transfer in the gas phase uses the following dispersion-convection equation:

$$\left(\frac{1}{r} \frac{\partial}{\partial r} \left(r D_{Gr} \frac{\partial C_{G,i}}{\partial r} \right) + \frac{\partial}{\partial z} \left(D_{Gz} \frac{\partial C_{G,i}}{\partial z} \right) \right) - \left(u_{Gr} \frac{\partial C_{G,i}}{\partial r} + u_{Gz} \frac{\partial C_{G,i}}{\partial z} \right) = R_{GL,i}, \quad (21)$$

where $C_{G,i}$ is the species i concentration in the gas phase, D_{Gr} and D_{Gz} are the gas phase dispersion coefficients in the radial and axial directions, u_{Gr} and u_{Gz} are the gas phase

interstitial velocities in the radial and axial directions, and $R_{GL,i}$ is the gas-liquid mass transfer rate of species i .

The gas-phase interstitial velocities are correlated with the Darcy total velocities in

$$u_G = \frac{U s_2}{\varepsilon_p}, \quad (22)$$

$$u_{Gr} = \frac{U_r s_2}{\varepsilon_p}, \quad (23)$$

$$u_{Gz} = \frac{U_z s_2}{\varepsilon_p}. \quad (24)$$

The gas-liquid mass transfer rate is expressed in

$$R_{GL,i} = k_{GL,i} a \left(\frac{p_{G,i}}{H_{c,i}} - C_i \right), \quad (25)$$

where $k_{GL,i}$ is the gas-liquid mass transfer coefficient, a is the gas-liquid interphase specific surface area, $p_{G,i}$ is the partial pressure of species i in the gas phase, and $H_{c,i}$ is Henry's constant of species i . C_i is the concentration of species i in the liquid phase.

The liquid phase mass transfer uses the following convection-dispersion equation:

$$\left(\frac{1}{r} \frac{\partial}{\partial r} \left(r D_{Lr} \frac{\partial C_{L,i}}{\partial r} \right) + \frac{\partial}{\partial z} \left(D_{Lz} \frac{\partial C_{L,i}}{\partial z} \right) \right) - \left(u_{Lr} \frac{\partial C_{L,i}}{\partial r} + u_{Lz} \frac{\partial C_{L,i}}{\partial z} \right) = R_{LS,i} - R_{GL,i}, \quad (26)$$

where $C_{L,i}$ is the species i concentration in the liquid phase, D_{Lr} and D_{Lz} are the liquid-phase dispersion coefficients in the radial and axial directions, u_{Lr} and u_{Lz} are the liquid-phase interstitial velocities in the radial and axial directions, and $R_{LS,i}$ is the liquid-solid mass transfer of species i .

The liquid-solid mass transfer rate is expressed in

$$R_{LS,i} = k_{LS,i} a_s (C_i - C_{pe,i}), \quad (27)$$

where $k_{LS,i}$ is the liquid-solid mass transfer coefficient and a_s is the catalyst specific surface area.

Since spherical catalyst particles are used, their specific surface area is as in

$$a_s = \frac{6}{d_p} (1 - \varepsilon_p). \quad (28)$$

The liquid-phase interstitial velocities are calculated using

$$u_L = \frac{U s_1}{\varepsilon_p}, \quad (29)$$

$$u_{Lr} = \frac{U_r s_1}{\varepsilon_p}, \quad (30)$$

$$u_{Lz} = \frac{U_z s_1}{\varepsilon_p}. \quad (31)$$

The mass transfer in the catalyst particles uses the following reaction-diffusion equation:

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left(-r^2 D_{pe,i} \frac{\partial C_{pe,i}}{\partial r} \right) = R_{pe,i}, \quad (32)$$

where $C_{pe,i}$ is the species i concentration in the catalyst particles, $D_{pe,i}$ is the effective diffusion coefficient of species i in the catalyst particles, and $R_{pe,i}$ is the surface reaction rate of species i in the catalyst particles.

The effective diffusion coefficient is calculated based on the Millington and Quirk model, as shown in

$$D_{pe,i} = \varepsilon_{pe}^{4/3} D_{L,i}. \quad (33)$$

The correlations for calculating the mass transfer coefficients are listed in Table 1.

2.3. Energy Transfer. The energy transfer is calculated on the reactor scale by assuming that the thermal equilibrium in all phases is present so that the temperature difference between the phases is negligible. For the energy balance, the equation is given as follows:

$$\rho C_p \left(u_r \frac{\partial T}{\partial r} + u_z \frac{\partial T}{\partial z} \right) - \left(\frac{1}{r} \frac{\partial}{\partial r} \left(r k_{er} \frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial z} \left(k_{ez} \frac{\partial T}{\partial z} \right) \right) = Q_{bed}, \quad (34)$$

where T is the temperature, ρ is the mixed fluid density, C_p is the effective heat capacity, k_{er} and k_{ez} are the thermal conductivities of the mixed fluid in the radial and axial directions, u_r and u_z are the mixed interstitial velocities in the radial and axial directions, and Q_{bed} is the reaction heat.

The axial heat capacity and the axial effective thermal conductivity are calculated based on the average volume of each phase using

$$(\rho C_p)_{eff} = (1 - \varepsilon_p) \rho_s C_{ps} + \varepsilon_G \rho_G C_{pG} + \varepsilon_L \rho_L C_{pL}, \quad (35)$$

$$k_{ez} = (1 - \varepsilon_p) k_p + \varepsilon_L k_L + \varepsilon_G k_G, \quad (36)$$

where C_{ps} , C_{pG} , and C_{pL} are the solid, gas, and liquid heat capacities, ρ_s , ρ_G , and ρ_L are the solid, gas, and liquid densities, and k_p , k_L , and k_G are the solid, liquid, and gas thermal conductivities.

The mixed interstitial velocities for the heat transfer are calculated using

$$u_r = \frac{U_r}{\varepsilon_p}, \quad (37)$$

$$u_z = \frac{U_z}{\varepsilon_p}. \quad (38)$$

The correlations for calculating the heat transfer coefficients, the thermal conductivity, and the formulas for physical properties are listed in Table 1.

2.4. Kinetic Models. The kinetics used in this study are from the kinetic model developed by Anand et al. [14] based on

TABLE 1: Correlations for mass and energy transfer parameters.

Parameters	Source	Unit	Correlation	No.
Gas-liquid mass transfer coefficient ($k_{GL,i}$)	[21]	1/s	$Sh_{GL} = (k_{GL,i} d_p / D_{L,i}) = 6 (G_L / \mu_L)^{0.41} (\mu_L / \rho_L D_{L,i})^{1/2}$	(41)
Liquid-solid mass transfer coefficient ($k_{LS,i}$)	[22]	cm/s	$Sh_{LS} = (k_{LS,i} a_s / D_{L,i}) = 1.8 (G_L / \mu_L a_s)^{1/2} (\mu_L / \rho_L D_{L,i})^{1/3}$	(42)
Liquid phase diffusion coefficient ($D_{L,i}$)	[23]	m ² /s	$D_{L,i} = 117.3 \times 10^{-18} (x M_i)^{1/2} T / (\mu_L \nu_i^{0.6})$	(43)
Gas phase diffusion coefficient ($D_{G,i}$)	[24]	cm ² /s	$D_{G,i} = ([3.03 - (0.98 / \sqrt{M_i})] (10^{-3}) T^{3/2} / p \sqrt{M_i} \sigma_i^2 \Omega_D)$	(44)
Diffusion coefficient of gas in liquid phase ($D_{L,i}$); $i = H_2, CO, CO_2$	[25]	m ² /s	$(D_{L,i} \times 10^9) = \beta (V - V_D) T^{1/2}$	(45)
The bed-to-wall heat transfer coefficient (h_T)	[26]	W/(m ² K)	$Nu_T = (h_T d_p / k_L) = 2.51 [1 - \exp(-(4.71 d_p / D_R))] Re_L^{0.68}$	(46)
Liquid phase axial dispersion coefficient (D_{Lz})	[27]	m ² /s	$Pe_{Lz} = (u_{Lz} d_p / D_{L,i}) = 0.042 Re_L^{0.5}$	(47)
Gas phase axial dispersion coefficient (D_{Gz})	[27]	m ² /s	$Pe_{Gz} = (u_{Gz} d_p / D_{G,i}) = 1.8 Re^{-0.7} \times 10^{-0.005 Re_L}$	(48)
The effective thermal conductivity in the radial direction (k_{er})	[26]	W/(m.K)	$k_{er} = k_{e0} + 0.093 (G_L / s_1) d_p C_{pL}$ $k_{e0} = (1 - \sqrt{1 - \varepsilon_p}) k_G + \sqrt{1 - \varepsilon_p} \Theta k_L$ $\Theta = (2/N) \{ (B(k_{rad} - 1)/N^2 k_{rad}) \ln(k_{rad}/B) - (B + 1/2) - (B - 1/N) \}$ $B = 1.25 (1 - \varepsilon_p / \varepsilon_p)^{10/9}$ $N = 1 - (B/k_{rad})$ $k_{rad} = (k_i / k_L)$	(49)
Density of gas (ρ_G)	[28]	kg/m ³	$\rho_G = (0.002016 p / 8.314 T)$	(50)
Viscosity of gas (μ_G)	[28]	Pa.s	$\mu_G = 2.145 \times 10^{-6} + 2.543 \times 10^{-8} T^1 - 1.024 \times 10^{-11} T^2 + 2.809 \times 10^{-15} T^3$	(51)
Thermal conductivity of liquid (k_L)	[28]	W/(m.K)	$k_L = -4.654 \times 10^{-13} T^6 + 1.639 \times 10^{-9} T^5 - 2.402 \times 10^{-6} T^4 + 1.876 \times 10^{-3} T^3 - 8.234 \times 10^{-1} T^2 + 1.926 \times 10^2 T - 1.875 \times 10^4$	(52)
Thermal conductivity of gas (k_G)	[28]	W/(m.K)	$k_G = 5.180 \times 10^{-3} + 6.728 \times 10^{-4} T^1 - 3.039 \times 10^{-7} T^2 + 6.589 \times 10^{-11} T^3$	(53)
Heat capacity of gas (C_{pG})	[28]	J/(kg.K)	$C_{pG} = 10808.501 + 21.580 T^1 - 0.045 T^2 + 3.854 \times 10^{-5} T^3 - 1.150 \times 10^{-8} T^4$	(54)
Heat capacity of liquid (C_{pL})	[28]	J/(kg.K)	$C_{pL} = 1.982 \times 10^{-5} T^2 - 1.772 \times 10^{-2} T + 6.883$	(55)
Gas-liquid surface tension (σ)	[28]	N/m	$\sigma = 6.101 \times 10^{-10} T^3 - 9.703 \times 10^{-7} T^2 + 4.503 \times 10^{-4} T - 5.000 \times 10^{-2}$	(56)

the reactions and the reaction rate coefficients' formulas, as listed in Table 2. The preexponential factors and activation energies for each reaction rate coefficient are shown in Table 3. The vegetable oil is represented by a model compound, i.e., triglycerides. The reactions involve ten components: triglyceride (TG), diesel ($C_{17}H_{36}$), undecane ($C_{11}H_{24}$), nonane (C_9H_{20}), octane (C_8H_{18}), hexane (C_6H_{14}), propane (C_3H_8), wax ($C_{19}H_{40}$), CO_2 , and H_2 . The reactions took place on Ni-W/ Si_2O_3 - Al_2O_3 catalyst. In the reaction mechanism, triglycerides react with hydrogen to produce hexane, nonane, diesel, propane, and wax. Some resulting diesel undergoes cracking into kerosene (nonane and undecane) and naphtha (hexane and octane). Wax is a by-product of the hydrocracking reaction.

2.5. Boundary Conditions. The flow rate at the reactor inlet and the pressure at the reactor outlet were specified for the momentum transfer. For mass transfer, the boundary condition has a mass flux at the reactor inlet and an outflow at the reactor outlet. For heat transfer, the boundary condition has an inlet temperature at the reactor inlet and an outflow at the reactor outlet. At the reactor wall, all boundary conditions have no flux, except that of the energy transfer, where energy flux due to heat transfer from the catalyst bed to the reactor wall was considered.

The boundary conditions for the catalyst pellet model are as follows:

At $r = 0$,

$$\frac{\partial C_{pe,i}}{\partial r} = 0. \quad (39)$$

At $r = R_p$,

$$-D_{pe,i} \frac{\partial C_{pe,i}}{\partial r} = R_{LS,i}. \quad (40)$$

3. Simulation and Discussion

One of the problems with TBRs is the high heat transfer resistance in the radial direction of the reactor bed. Therefore, a high ratio of the length to the diameter of the reactor bed or aspect ratio helps reduce the problem. In this study, an aspect ratio of 100 was used to observe the temperature distribution in the reactor bed. The hydrocracking process was simulated in excess hydrogen. To prevent catalyst deactivation, a 5% *w/w* triglyceride (TG) in diesel was fed to the reactor. The feed is eventually a mixture of gaseous hydrogen and TG solution and enters the reactor at 420°C. Since the reaction is exothermic, the reactor bed is cooled by a cooling medium, providing the reactor wall with a temperature of 420°C. The process parameters used in this simulation are listed in Table 4. The simulation was performed numerically using COMSOL Multiphysics 5.3.

3.1. Temperature Profile. Figure 2 shows the temperature profile in the bed reactor. The increasing temperature in the reactor bed towards the bed outlet indicates that the

hydrocracking reaction is exothermic. The heat generated along the reactor is shown in Figure 3. The highest reaction heat occurs in the reactor inlet due to the high concentrations of the reactants in this location. Towards the reactor outlet, the triglyceride concentration decreases as the reactant is converted. The heat generated in the reactor is transported by the flowing fluid to the reactor outlet so that although the reaction heat in the downstream reactor is lower, its temperature is higher. The temperature gradient in the radial direction is shown in Figure 4. The temperature difference from the symmetry axis to the reactor wall increases towards the reactor outlet. Near the reactor inlet, there is a negative gradient. This is because the wall temperature near the reactor inlet is higher than that at the reactor symmetry axis. The temperature gradient is due to resistance to conduction and convection heat transfers in the radial direction. Heat carried downstream of the reactor by the fluid flow accumulates at the reactor outlet so that the temperature at the symmetry axis is higher than that at the reactor wall. The temperature difference between the wall and the symmetry axis at the reactor outlet is 0.47 K/m or 0.0235 K, along with a radial distance of 5 cm. It can be seen that the temperature is almost unchanged radially. This indicates that an aspect ratio of 100 provides effective radial heat transfer.

3.2. Concentration Profiles. Figure 5 shows the concentration profile of triglyceride, diesel, and hydrogen. Figure 5(a) shows that triglyceride concentration decreases towards the reactor because it reacts in the reactor to become products. Conversely, the diesel concentration increases towards the reactor outlet (Figure 5(b)). The triglyceride and diesel concentrations are evenly distributed in the radial direction. This proves that the mixing and cooling processes are very effective. The hydrogen concentration increases towards the outlet (Figure 5(c)). This is due to the mass transfer of hydrogen from the gas phase into the liquid phase. This hydrogen situation causes hydrogen molecules to accumulate towards the outlet. Therefore, its concentration is the highest at the reactor outlet. However, the hydrogen transfer to the liquid phase insignificantly reduces its concentration in the gas phase. The hydrogen concentration profile shows that the hydrogen concentration in the liquid phase is constant from the 3 m position at the reactor inlet to the reactor outlet. This indicates that the hydrogen concentration in the liquid phase is saturated from the 3 m position.

The concentration profiles in the catalyst particles are shown in Figure 6. These profiles were captured from the particles in the middle of the reactor length and in the symmetry axis of the reactor. Overall, the product concentrations decrease from the core of the catalyst particle towards the external surface of the catalyst particle. The diesel concentration increases until it peaks at the 0.8 m dimensionless positions of the particles and then decreases towards the external surface of the catalyst particle.

3.3. Optimisation. In this study, optimisation was conducted with the purity of green fuel products as an objective function (i.e., the highest purity). Decision variables are

TABLE 2: Hydrocracking reactions and their reaction rate coefficients' formula.

Reaction rate	Reaction stoichiometry	Reaction rate coefficient	Reaction rate formula
r_1	$TG + H_2 \longrightarrow \text{hexane} + CO_2$	k_1	$k_1 C_{TG}$
r_2	$TG + H_2 \longrightarrow \text{nonane} + CO_2$	k_2	$k_2 C_{TG}$
r_3	$TG + H_2 \longrightarrow \text{diesel} + \text{propane} + CO_2$	k_3	$k_3 C_{TG}$
r_4	$TG + H_2 \longrightarrow \text{wax} + CO_2$	k_4	$k_4 C_{TG}$
r_5	$\text{Diesel} + H_2 \longrightarrow \text{nonane} + \text{octane}$	k_5	$k_5 C_{C17}$
r_6	$\text{Nonane} + H_2 \longrightarrow \text{hexane} + \text{propane}$	k_6	$k_6 C_{C9}$
r_7	$\text{Diesel} + H_2 \longrightarrow \text{hexane} + \text{undecane}$	k_7	$k_7 C_{C17}$

TABLE 3: Preexponential factors and activation energies for each reaction rate coefficient.

Reaction rate coefficient	Preexponential factor (1/h)	Activation energy (kJ/mol)
k_1	4.65×10^9	130
k_2	9.48×10^8	122
k_3	1.00×10^{10}	116
k_4	1.02×10^{10}	115
k_5	9.48×10^8	122
k_6	4.65×10^9	130
k_7	4.65×10^9	130

TABLE 4: Process parameters.

Reactor outlet pressure	80 bars
Inlet temperature	420°C
Feed molar ratio H_2/TG	105.5
Inlet gas velocity	5 cm/s
TG fraction in the feed	5% w/w
Wall temperature	420°C

FIGURE 2: Temperature profile.

catalyst pellet diameter, inlet gas velocity, feed molar ratio, and inlet temperature. Based on the base case simulation, the reactor geometry provided good heat transfer so that to accelerate the convergence of the numerical calculations, the optimisation was conducted in isothermal conditions. The range of each parameter is listed in Table 5. The optimisation results are shown in Figure 7.

FIGURE 3: Heat of the reaction.

The increasing pellet diameter results in decreasing product purity. The phenomenon occurs because the catalyst mass decreases with increasing catalyst size. This reduces the active sites of the catalyst so that the conversion decreases. More reactants presented in the reacting mixture decrease the product purity. However, within the low range of the pellet diameter, the increase in the pellet diameter provides better mixing in the bed interstice. The reactant molecules in the liquid phase reach the external surface of the catalyst particles more easily as the bed porosity increases. The highest product purity was achieved with the catalyst pellet diameter at 1 mm.

Based on simulations with various inlet gas velocities, the highest product purity was achieved at 0.5 cm/s. When the velocity increases, the residence time reduces, which results

FIGURE 4: Radial temperature gradient along the reactor.

FIGURE 5: Concentration profiles of (a) triglyceride, (b) diesel, and (c) hydrogen.

FIGURE 6: Concentration profiles in the catalyst pellet.

TABLE 5: Process parameter ranges.

Parameter	Range
Particle diameter (mm)	0.5–10
Inlet gas velocity (cm/s)	0.5–9
Feed molar ratio	21.1–105.5
Inlet temperature (°C)	340–500

in reduced conversion. Thus, the product purity also decreases. Although the highest purity was obtained at the lowest velocity in the simulation range, the production rate was low at a low velocity. Therefore, the best velocity is not at 0.5 cm/s but at 1 cm/s. At this velocity, the product purity remains high, and the production rate is not too low.

Changing the feed molar ratio changes the feed triglyceride concentration since the simulation was conducted with excess hydrogen. Therefore, a higher feed molar ratio means less triglyceride in the feed and more hydrogen in the feed. Since triglyceride is the limiting reactant, the increase in the hydrogen concentration induces an increase in the converted triglyceride; thus, the product purity increases. The best feed molar ratio is 105.5, which is the highest ratio in the simulation range.

Since the reaction kinetics are affected by the temperature, the reaction temperature has an interesting effect on the product purity. Generally, an increase in the reaction temperature causes an increase in product purity. After 420°C, however, the purity of diesel decreases, while the purities of kerosene and naphtha increase exponentially. This is because diesel is cracked to become kerosene and naphtha at higher reaction rates above 420°C. The diesel purity reached 43.26% at 420°C, with the green fuel purity being 51%. The green fuel

purity increases up to 69% at 500°C. The purities of individual diesel, kerosene, and naphtha at 500°C are shown in Table 6. The hydrocracking reaction leads to hydrocarbon products with shorter chains at higher temperatures.

The optimisation results are resumed in Table 7. The table shows that if diesel purity is emphasised, then the inlet gas velocity must be 1 cm/s, the catalyst diameter must be 1 mm, the feed molar ratio must be 105.5, and the inlet temperature must be 420°C. This gives the 43.26% diesel purity and 44.57% yield. This table also provides information on when the purities of kerosene or naphtha are emphasised.

These optimisation results were then compared to the optimisation results of the triglyceride hydrotreating process to produce green diesel in a TBR through simulations conducted by Muharam et al. [16]. These results were also compared to the experimental results obtained by Anand et al. [14] for the kinetics study, whose models were used in this study. The comparative information on the three studies is shown in Table 8.

The table shows that the conversion and yield of the hydrocracking process are higher than those of the hydrotreating process. This is because the hydrocracking process occurs at a higher temperature. In addition, the energy supplied to the reacting mixture breaks down more chemical bonds of reactants in hydrocracking than in hydrotreating. In the hydrotreating process, many bond breakings occur in double bonds or other weak bonds.

The product purity in this study cannot be compared to that in the hydrotreating process because of differences in purity calculations. In the hydrotreating process, the purity was calculated by involving a solvent, which was assumed to have the same properties as diesel products.

FIGURE 7: Process parameters of optimisation results: (a) particle diameter, (b) gas inlet velocity, (c) feed molar ratio, and (d) inlet temperature.

TABLE 6: Purity of products at 500°C.

Product	Purity (%)
Diesel	28.37
Kerosene	18.8
Naphtha	22.24
Total	69.41

TABLE 7: Optimisation results based on the purity of each product.

Parameter	Diesel product	Kerosene product	Naphtha product
Catalyst diameter (mm)	1	1	1
Inlet gas velocity (cm/s)	1	1	1
Feed molar ratio	105.5	105.5	105.6
Inlet temperature (°C)	420	500	500
Yield (%)	44.57	21.52	25.46
Purity (%)	43.26	18.80	22.24

TABLE 8: Comparison of this study's results with Muharam et al. [11] and Anand et al. [14].

Parameter	This research	Muharam et al. [11]	Anand et al. [14]
Process	Hydrocracking	Hydrotreating	Hydrocracking
Reactor diameter (cm)	10	150	0.65
Reactor height (cm)	1000	800	3.5
Catalyst diameter (mm)	1	1.6	Powder
Inlet gas velocity (cm/s)	1	1.5	8.00E-04
Feed molar ratio	105.5	188	63.3
TG fraction in feed	5% w/w	5% w/w	5% w/w
Inlet temperature (°C)	500	375	420
Pressure (bar)	80	34.5	80
Triglyceride conversion (%)	99.97	88.3	100
Yield (%)	79.46	58.5	87.43
Purity (%)	69.4	97.8*	N/A

*Calculation involved solvent.

When compared to the experimental results in the small reactor with a powder catalyst performed by Anand et al. [14], the yield calculated in this study is lower, with a difference of 5.54%. In fact, the purity in this study is the highest purity, which is the result of optimisation. The higher energy and mass transfer resistances in the larger reactor are believed to be responsible for this difference. Advantageously, both the model and the experimental study achieved a high triglyceride conversion, 99.97% and 100%, respectively.

4. Conclusion

The hydrocracking model of jatropha oil in a TBR was successfully developed in this study. Good heat transfer in the reactor was achieved with an aspect ratio of 100. The diesel purity reaches 44.22% at 420°C, kerosene purity reaches 21.39% at 500°C, and naphtha purity reaches 25.30% at 500°C. With varying the inlet temperature, feed molar ratio, gas inlet velocity, and catalyst diameter, it is observed that the green fuel purity increases with the increase in inlet temperature and feed molar ratio, while the green fuel purity decreases with the increase in catalyst diameter and gas inlet velocity. The optimum condition is achieved at the catalyst diameter of 1 mm, the inlet gas velocity of 1 cm/s, the feed molar ratio of 105.5, and the inlet temperature of 500°C. The green fuel purity achieved at this optimum condition is 69.4% with a triglyceride conversion of 99.97%.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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