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Original Research Article

Reaction Kinetics of Waste Cooking Oil Hydrocracking into Biofuel Using Ni-Impregnated Mesoporous Silica Catalyst

Siti Salamah*[1], Wega Trisunaryanti[2], Indriana Kartini[2], Suryo Purwono[3]

[1]Department of Chemical Engineering, Faculty of Industrial Technology, Universitas Ahmad Dahlan Kampus 4, Jl. Ringroad Selatan, Tamanan, Yogyakarta 55166, Indonesia
[2]Department of Chemistry, Faculty of Mathematics and Natural Sciences, Universitas Gadjah Mada, Sekip Utara, Yogyakarta 55281, Indonesia
[3]Department of Chemical Engineering, Faculty of Engineering, Universitas Gadjah Mada, Jl. Grafika No. 2, Yogyakarta 55281, Indonesia.

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Abstract

The growing demand for energy and the scarcity of fossil fuel resources have driven research into alternative fuels, one of which being the conversion of waste cooking oil into biofuel through hydrocracking. This study investigates the reaction kinetics of waste cooking oil hydrocracking using a Ni-impregnated mesoporous silica catalyst. The process was conducted at 450 °C with a hydrogen gas flow to produce products such as green naphtha, green gasoline, and green diesel. The proposed reaction kinetics model was the pseudo-first order, solved using differential and integral methods. The results showed that the first-order reaction provided a more representative outcome, with a reaction rate constant (k') of 0.276 h^{-1} at 450 °C. Additionally, the Arrhenius kinetic model revealed an activation energy of 37.8748 kJ/mol for this process. Thus, this study demonstrates a significant potential of using mesoporous silica catalysts in waste cooking oil hydrocracking to produce environmentally friendly and economically viable biofuels.

Keywords: Hydrocracking; Biofuel; Mesoporous Silica; Reaction Kinetics

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1. Introduction

The rapid increase in global energy demand, along with the depletion of fossil fuel reserves, has led to significant concerns regarding energy security and environmental sustainability. The growing gap between energy supply and demand has triggered interest in alternative fuels that are renewable, environmentally friendly, and economically viable [1]. Waste cooking oil (WCO) has been identified as a promising candidate due to its abundance and high triglyceride content. Statistically, WCO generated from restaurants reaches approximately 3 billion gallons per year, much of which is improperly disposed of, leading to serious environmental pollution [2]. Utilizing WCO as a raw material for biofuel production addresses two major issues simultaneously, namely waste management and the search for sustainable energy alternatives.

The main component of WCO, triglycerides, consists of long-chain fatty acid esters that are highly flammable and can be transformed into liquid fuels with energy content comparable to petroleum-based fuels [3,4]. In addition to its renewability and non-toxic nature, WCO offers significant economic advantages because of its low cost and widespread availability [5,6]. Various thermochemical processes such as pyrolysis, gasification, and hydrocracking have been studied to convert WCO into biofuel, among which

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* Corresponding Author.
Email: sitisalamah@che.uad.ac.id (S. Salamah)

hydrocracking has attracted considerable attention [7-9]. Hydrocracking is a process that breaks down heavy hydrocarbon molecules into lighter fractions like gasoline, diesel, and kerosene by cleaving C–C bonds, often under high pressure and temperature in the presence of hydrogen [10,11].

Compared to conventional catalytic processes, hydrocracking has several advantages, such as higher fuel quality and the potential to produce green fuels (green diesel, green naphtha, green gasoline) with better environmental performance [9,12]. The use of heterogeneous catalysts further enhances the process due to their ease of separation, reusability, and high selectivity [13]. Mesoporous silica, in particular, has gained interest as a catalyst support because of its high surface area and porous structure, which minimize diffusion limitations and increase catalytic efficiency [14,15]. Incorporating nickel into mesoporous silica can significantly enhance catalytic performance by increasing the number of active sites and improving hydrogenation capacity. Moreover, nickel is relatively inexpensive and environmentally benign, making it suitable for sustainable catalytic applications [16].

Despite the growing body of research on biofuel production via hydrocracking, studies focusing on the kinetic modelling of WCO hydrocracking using Ni-impregnated mesoporous silica catalysts remain limited. Understanding the reaction kinetics is crucial for optimizing reactor design, scaling up processes, and improving overall system efficiency. This study aims to investigate the reaction kinetics of WCO hydrocracking using Ni/mesoporous silica as the catalyst, under the assumption of pseudo-first-order kinetics. The proposed kinetic model is expected to capture the reaction behaviour effectively, providing valuable insights for the development of sustainable biofuel production technologies.

2. Materials and Method

2.1. Materials

The material used in this study was waste cooking oil obtained from the Yogyakarta city area. The catalyst used was mesoporous silica impregnated with 1% Ni metal. The catalyst characterization results showed a surface area of 130.5 m^2/g, a total pore volume of 0.4 cm^3/g, and an average pore diameter of 12.3 nm. The equipment used was a hydrocracking reactor setup. The reactor was a semi-batch type made of stainless steel, with an outer diameter of 25 cm, an inner diameter of 5 cm, a reactor height of 36 cm, a catalyst bed height of 3.5 cm, and a feed bed height of 6.5 cm. The reactor setup is shown in Figure 1.

2.2. Hydrocracking Process

The used cooking oil sample was cleaned of impurities by filtering it with filter paper. The cracking process was conducted with a catalyst-to-feed ratio of 1:100. The catalyst and feed were weighed, with the catalyst placed in the catalyst bed and the feed placed in the feed bed. The sample and catalyst were inserted into the semi-batch reactor made of stainless steel. The cracking process was carried out at 450 °C with a hydrogen gas flow of 20 mL/min. The reaction temperature of 450 °C was selected based on preliminary

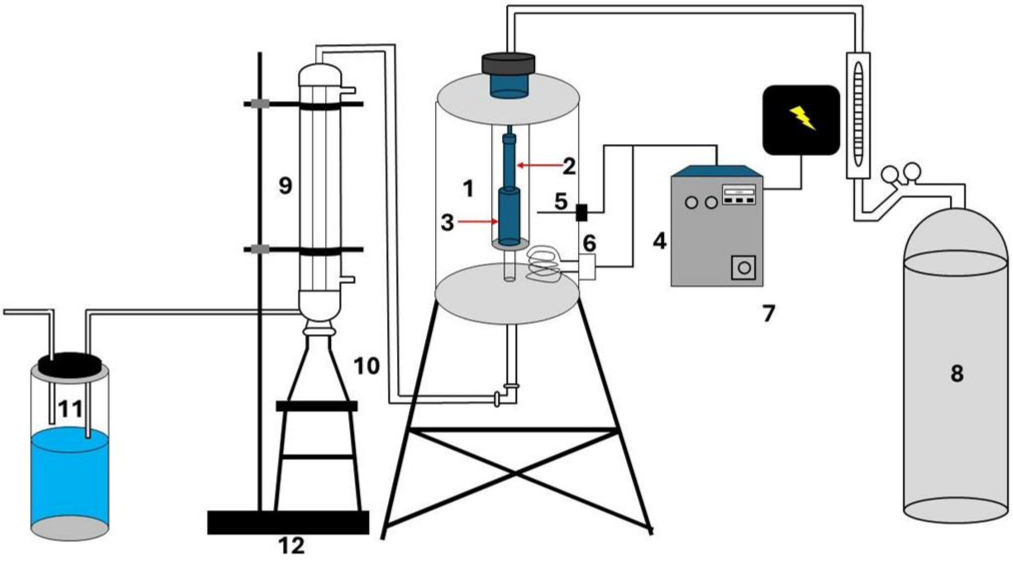

Figure 1. Hydrocracking reactor setup. (1). Combustion furnace, (2). Catalyst bed, (3). Feed bed, (4). Regulated power supply, (5). Thermocouple, (6). Heating plate, (7). Gas flow meter, (8). H_2 gas cylinder, (9). Condenser, (10). Liquid product collector, (11). Gas product collector, (12). Stand

studies and consistent with previous literature showing optimal liquid product yield at this temperature range for waste cooking oil hydrocracking [17-19]. Our preliminary experiments also indicated that 450 °C yielded the optimal liquid fraction [20]. The formed products were passed through a silicone hose via a condenser. The yield was calculated by Equation (1).

$$\%\text{Yield} = \frac{liquid\ product\ weight}{WCO\ weight} \times 100\% \quad (1)$$

2.3. Kinetics Modelling

The hydrocracking process occurs between reactant A (Waste Cooking Oil - WCO) and hydrogen gas with the assistance of mesoporous silica catalyst. The reactions involved in this process are highly complex, involving triglyceride breakdown followed by reactions such as decarbonylation, decarboxylation, alkylation, aromatization, and others [21]. The products generated include diesel oil, alkanes, alkenes, and by-products such as CO, CO_2, and H_2O [22,23]. The variety of products produced leads to a multitude of kinetic reaction models being discussed. However, the main product is biofuel (liquid fraction), as indicated by the research data on the yield of liquid products (oil). Therefore, the primary focus of this kinetic review is the hydrocracking of waste cooking oil into biofuel. Several possible reactions during the hydrocracking of waste cooking oil can be seen in Table 1.

Due to this complexity and the difficulty of identifying and quantifying all possible intermediates and products, a lumped kinetic modeling approach is adopted in this study. In this context, Reaction 3 in Table 1 is not intended to fully represent the mechanistic details of hydrocracking, but rather to serve as a simplified stoichiometric reaction that captures the overall conversion of WCO into biofuel, particularly the liquid hydrocarbon fraction. This simplification allows for a practical and effective kinetic analysis using a pseudo-first-order approach, especially under conditions where hydrogen is in large excess and its concentration can be considered constant. Similar kinetic simplifications have also been used in prior studies such as Zhang *et al.* [22] and Hasanudin *et al.* [21].

Table 1. Representative reactions in the deoxygenation pathway during hydrocracking.

No	Reaction	Ref.
1	$R - COOH + H_2 \rightarrow R + H_2O$	[20]
2	$C_{17}H_{35}COOH + 2H_2 \rightarrow C_{18}H_{38} + 2H_2O$ $C_{17}H_{35}COOH + H_2 \rightarrow C_{17}H_{36} + 2H_2O + CO$ $C_{17}H_{35}COOH \rightarrow C_{17}H_{36} + CO_2$	[9]
3	$R - CH_2COOH + 3H_2 \rightarrow R - CH_2 - CH_3 + 2H_2O$	[24]

The scope of this research is focus on the hydrodeoxygenation reaction, as represented by Reaction Equation 3. Based on the experimental conditions, reactant A is WCO, with a liquid mass of 10 g. The reaction is initiated by heating at a temperature of 450 °C, and converting the WCO into a gas phase. Hydrogen gas is then continuously supplied, reacting with the WCO, and passing through a solid mesoporous silica catalyst. Considering the composition of WCO and hydrogen gas, the flow rate of hydrogen is relatively larger than the mass of WCO. This indicates that during the reaction, the reduction in hydrogen moles is relatively small compared to its initial amount. Therefore, it can be assumed that the moles of hydrogen remained approximately constant throughout the reaction, allowing for the application of a pseudo-first-order reaction approach in this case [25], following:

$$-r'_A = -\frac{V}{W}\frac{dC_A}{dt} = k'_A C_A^n \quad (2)$$

In this context, k_A' *W*/*V* can be assumed as *k*' so Equation (1) becomes:

$$\frac{dX_A}{dt} = \frac{k' C_A^n}{C_{A0}} \quad (3)$$

Since there is a volume change during the reaction, the concentration of A can be determined using the following formula:

$$C_A = \frac{C_{A0}(1-X_A)}{1+\varepsilon_A X_A} \quad (4)$$

The initial concentration of A (C_{A0}) can be calculated using the ideal gas law, with the following equation:

$$C_{A0} = \frac{y_{A0}P}{RT} \quad (5)$$

These equations are then used to determine the process parameters. The resulting process parameters include the hydrocracking rate constant and reaction order. Equation (2) can be solved using two methods: the differential method by determining the initial condition and the integral method by determining the boundary condition. The optimization model employed is to trial values of k' and n that yield the smallest SSE (Sum of Squares of Errors). The next step is to determine the reaction order (*n*) as 0, 1, 2, etc., and perform trials for *k*' and compare the resulting SSE. The reaction rate constant (k') is then used to determine the activation energy of the hydrocracking process of WCO. The activation energy is obtained by solving the Arrhenius equation, as follows:

$$k' = A.exp\left(-\frac{E_A}{RT}\right) \quad (6)$$

3. Results and Discussion

The hydrocracking process of waste cooking oil was carried out using mesoporous silica catalyst impregnated with Ni metal. The objective of this research is to investigate the reaction kinetics of hydrocracking waste cooking oil to produce biofuel. A kinetic model based on a pseudo-first order reaction approach was proposed and solved using both differential and integral methods. In this study, the differential method for hydrocracking reaction kinetics analysis was implemented using a numerical optimization approach. The *ode45* ordinary differential equation (ODE) solver in MATLAB was utilized to numerically solve the proposed differential kinetic model for various sets of reaction rate constant (k') and reaction order (n) values. The real-time reactant concentration (C_A) was calculated based on the product yield fraction (X_A). Optimization of k' and n was performed by minimizing the Sum of Squared Errors (SSE) between the simulated product yield fraction and the experimental data, using MATLAB's *lsqnonlin* function. The results are showed in Figure 2.

Based on Figure 2, the pattern of conversion over time demonstrates clear differences across various reaction orders (n = 0, 1, 2, 3). Table 2 shows that the Sum of Squared Errors (SSE) decreases as the reaction order increases, indicating that higher-order models provide a better statistical fit to the experimental data. However, model selection should not be based solely on the lowest SSE value. The appropriateness of a kinetic model must also

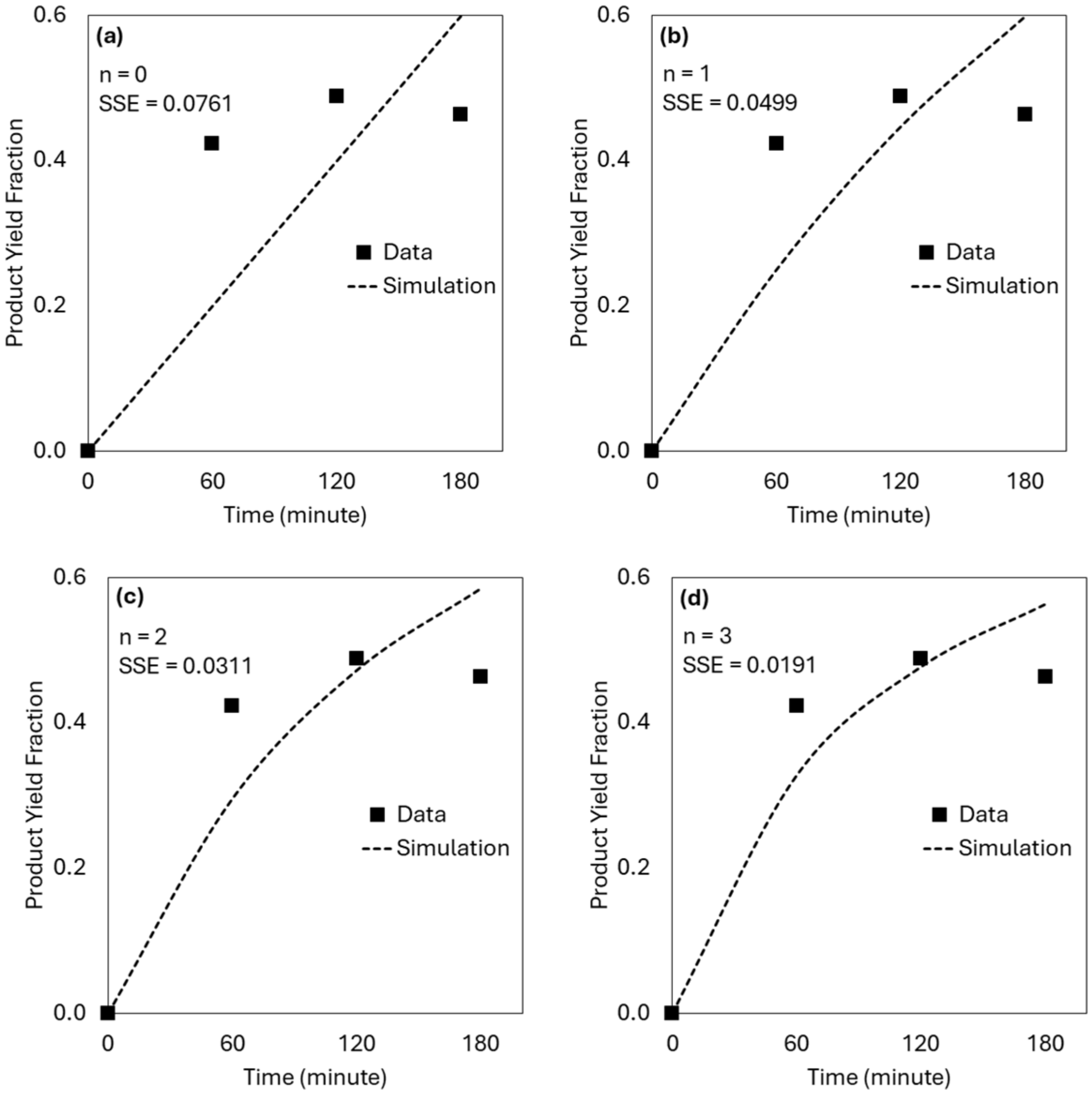

Figure 2. Hydrocracking kinetics plot with differential solution at various reaction orders.

account for the underlying reaction phenomena, including the reaction mechanism, operating conditions, and theoretical limitations of the system. Theoretically, the maximum conversion achievable is 1.0, meaning all reactants are transformed into products. Therefore, the selected model must be physically realistic and chemically justifiable. Among the tested models, the first-order reaction provides both a reasonable fit (SSE = 0.0499) and aligns well with the physical context of the process. Specifically, the pseudo-first-order assumption is supported by the excess hydrogen flow relative to the amount of waste cooking oil (WCO), allowing the hydrogen concentration to be considered constant. This supports a reaction rate primarily dependent on the WCO concentration, justifying the application of a first-order kinetic model.

To further assess the predictive capability of the models, conversion versus time curves were re-plotted using the fitted values of k' and n, as shown in Figure 3. These simulations were extended up to 5000 minutes to evaluate long-term behaviour. It is important to note that due to technical limitations and the semi-batch nature of the reactor setup, experimental data collection was limited to a shorter duration. The extended simulation is, therefore, a theoretical representation to demonstrate the long-term trends and physical realism of the kinetic model, such as its approach towards the theoretical maximum conversion of 1.0. Figures 3a and 3b illustrate the conversion profiles for n = 1 and n = 3, respectively. The first-order model (Figure 3a) shows a realistic approach toward the theoretical maximum conversion of 1.0, with a gradual decline in reaction rate. In contrast, the third-order model (Figure 3b), despite a lower SSE (0.0191), demonstrates a slower and potentially less realistic approach to maximum conversion. Moreover, higher-order models may fit short-term data well but can exhibit mechanistically unjustified behaviour at longer time scales, such as instability or nonphysical conversion trends. The visual alignment of the first-order curve with expected reaction behaviour, coupled with theoretical justification and reasonable predictive performance, supports the conclusion that the first-order kinetic model is the most representative and appropriate choice for describing the hydrocracking process under the given experimental conditions.

Table 2. Comparison of optimization results for k and n with differential and integral methods.

n	Differential Method			Integral Method		
	SSE	k'	Unit	SSE	k'	Unit
0	0.0761	0.000056	mol.L^{-1}.min^{-1}	0.0761	0.000056	mol.L^{-1}.min^{-1}
1	0.0499	0.0046	min^{-1}	0.1154	0.0042	min^{-1}
2	0.0311	0.3820	L.mol^{-1}.min^{-1}	0.1822	0.3204	L.mol^{-1}.min^{-1}
3	0.0191	30.7122	L^{2}.mol^{-2}.min^{-1}	0.3016	25.0273	L^{2}.mol^{-2}.min^{-1}

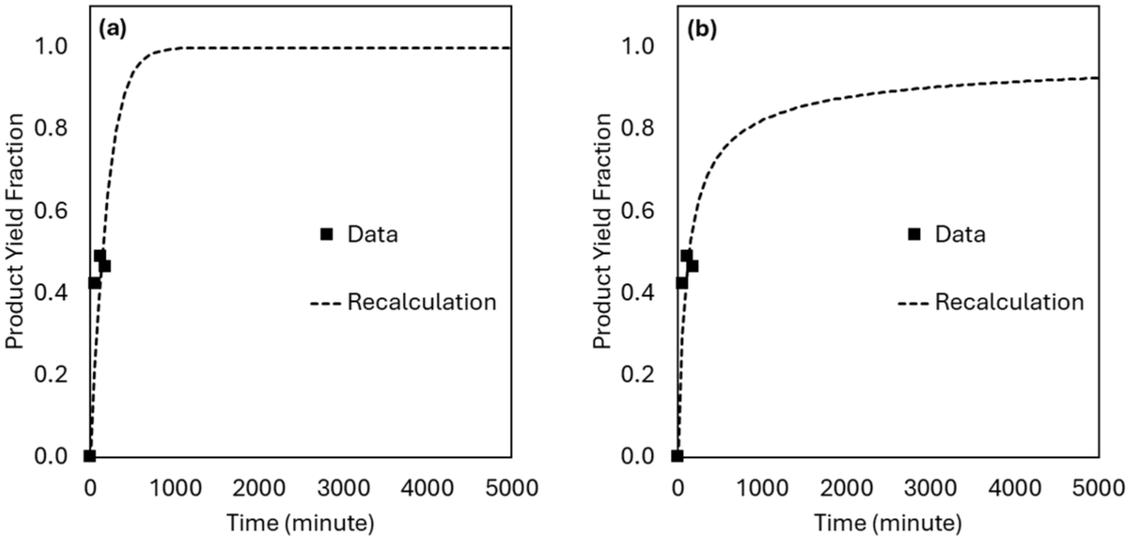

Figure 3. Re-plotting of hydrocracking kinetics with differential method (a) order 1 and (b) order 3.

The underlying reaction conditions, particularly the high hydrogen excess, justify the application of a pseudo-first-order model. Since the concentration of hydrogen can be considered constant, the reaction rate is primarily dependent on the concentration of the waste cooking oil. This approach is consistent with previous studies on similar systems [21,22]. By focusing on the overall conversion of WCO to liquid biofuel through a lumped kinetic model, this approach effectively bypasses the complexities of modelling each individual mechanistic step. It's important to note that the kinetic model proposed here provides a global kinetic representation of WCO hydrocracking rather than describing elementary steps like pure deoxygenation or cracking mechanisms. This is a common and accepted practice in modelling complex catalytic systems where product distributions are broad and difficult to analyse precisely. The agreement of the obtained kinetic parameters with published literature further corroborates the reliability of this modelling approach.

In addition to the differential method, this study also solved the model using the integral method. In the integral method, the values of k' and n can be optimized to produce the smallest SSE value. The optimization results for k' and n using the integral method are k' =0.0036 and n = 0.9776 with an SSE of 0.1160. This reaction order can be classified as a first-order reaction with a low SSE value. The graph plot of the optimal results can be seen in Figure 4. Based on Figure 4, we can observe that the integration model is linear, resulting in a straight-line output. However, considering the phenomena occurring, the differential model is more appropriate as it can better represent the observed phenomena.

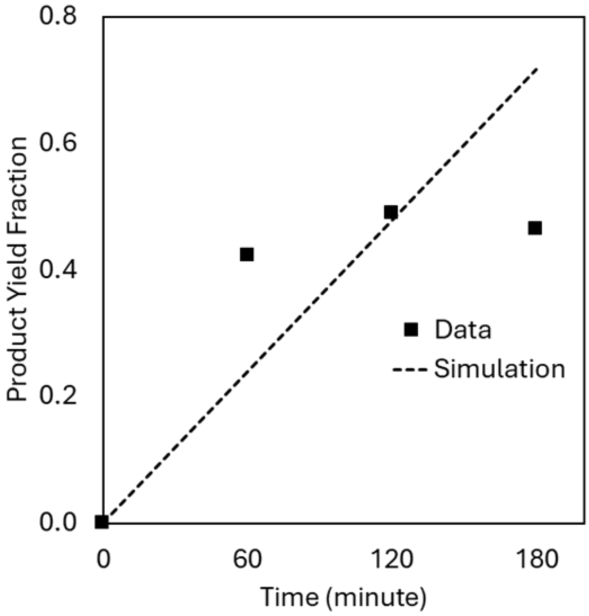

Figure 4. Plot of hydrocracking kinetics model optimization for k and n using the integral method.

Based on Table 2, it can be observed that both models yield nearly identical values for the apparent rate constant (k'). The main difference between the two methods lies in the SSE values. Among the tested models, the first-order reaction model shows a good fit to the experimental data, as indicated by its minimum SSE. Accordingly, the first-order differential model appears to adequately represent the hydrocracking reaction behaviour, with a calculated rate constant of k' = 0.0046 min^{-1}, equivalent to 0.276 h^{-1} at a reaction temperature of 450 °C. For comparison, Zhang *et al.* [22] reported a hydrocracking rate constant of 0.3007 h^{-1} at 300 °C, while Hasanudin *et al.* [21] reported a value of 1.2610 at 400 °C. It is important to note that Zhang *et al.* expressed their kinetic parameters in h^{-1} for deoxygenation pathways, although the specific reaction orders and lumping schemes in their model differ from the pseudo-first-order approach used in this study. Similarly, Hasanudin *et al.* assumed a second-order reaction for the main reactant and reported the rate constant without specifying explicit units. Despite these methodological differences in reaction order, model complexity, and parameter definition, the pseudo-first-order rate constant obtained in this study (0.276 h^{-1}) is of the same order of magnitude as those reported by Zhang *et al.* and Hasanudin *et al.*, thus supporting the validity and relevance of the proposed model under similar hydrocracking conditions [21,22].

In addition to solving the model for the reaction rate constant, this study also involved estimating the activation energy (E) and the pre-exponential factor (A) in the Arrhenius equation. The known value of the reaction rate constant k at 450 °C was used as a reference to solve the Arrhenius equation. Using MATLAB's fzero solver, the values of A and E were calculated by finding the root of the nonlinear equation that equates the left-hand side and right-hand side of the Arrhenius expression at that specific temperature. Once the values of A and E were obtained, the Arrhenius equation was then used to simulate the temperature dependence of the reaction rate constant over the range of 420–500 °C, using the ideal gas constant R = 8.314 J.mol^{-1}.K^{-1}. This approach yielded values of A = 145.4027 h^{-1} and E = 37.8748 kJ.mol^{-1}. These values are within the expected range compared to previous studies on the hydrocracking of used cooking oil, which reported activation energies of 31.79 kJ.mol^{-1} using a Ru/Al_2O_3 catalyst [24], and 56 kJ.mol^{-1} using a NiW/SiO_2–Al_2O_3 catalyst [26].

Figure 5 visualizes the temperature dependence of the hydrocracking rate constant based on the Arrhenius equation, with the complete set of k' values and corresponding temperatures summarized in Table 3. The Figure 5(a) plot displays the natural logarithm of the rate constant as a function of the reciprocal of temperature, while the Figure 5(b) plot shows the direct relationship between k' and absolute temperature. These curves are generated using the Arrhenius model and illustrate the expected trend, that the rate constant increases exponentially with temperature. This is due to the fact that an increase in temperature can enhance molecular kinetic energy, which implies an increase in the reactions occurring. This aligns with previous research related to hydrocracking [27].

4. Conclusions

In this study, the hydrocracking process of used cooking oil was conducted using mesoporous silica catalyst impregnated with Ni metal. The proposed kinetic model was solved using differential and integral methods. From the kinetic reaction analysis conducted, the differential method provided results that were more representative of the reaction phenomena occurring. The reaction proceeded as a pseudo first-order reaction with a heterogeneous kinetic reaction model. The simulation results show that the first-order reaction has a reaction rate constant (k') of 0.276 h^{-1} at a temperature of 450 °C. Furthermore, the Arrhenius kinetic model yields an activation energy for the hydrocracking process of used cooking oil of 37.8748 kJ.mol^{-1}.

Table 3. Comparison of hydrocracking rate constant values at various temperatures.

T (°C)	k' (h^{-1})
420	0.2034
440	0.2445
460	0.2911
480	0.3434
500	0.4014

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CRedit Author Statement

Author Contributions: S. Salamah: Methodology, Investigation, Formal Analysis, Data Curation, Writing Draft Preparation, Visualization, Review and Editing. W. Trisunaryanti: Methodology, Validation ,Review, Supervision. I. Kartini: Review, Supervision. S. Purwono: Conceptualization, Review, Supervision. All authors have read and agreed to the published version of the manuscript.

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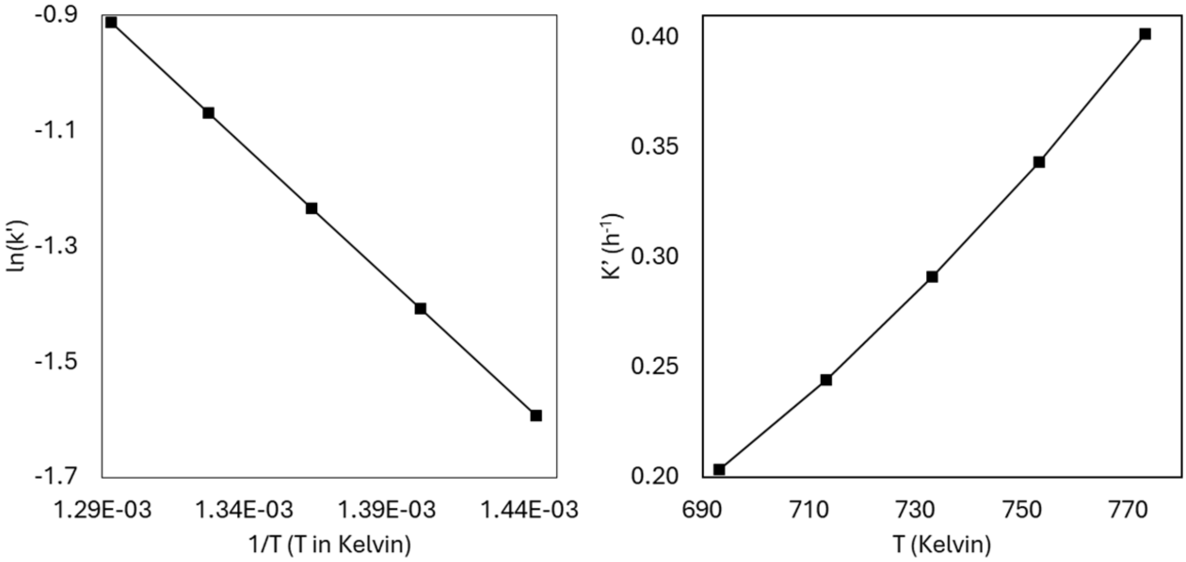

Figure 5. Arrhenius plot for waste cooking oil hydrocracking (a) and effect of temperature on reaction rate constant (b).

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