

Article

Liquid–Liquid Equilibrium Behavior of Ternary Systems Comprising Biodiesel + Glycerol and Triglyceride + Methanol: Experimental Data and Modeling

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Abstract: Having a comprehensive knowledge of phase equilibrium is advantageous for industrial simulation and design of chemical processes. For further acquisition of primary data to facilitate the separation and purification of waste oil biodiesel systems, a liquid–liquid equilibrium (LLE) tank is deployed for the ternary system of waste oil biodiesel + methanol + glycerin, thereby enhancing the precision and efficiency of the process. The phase equilibrium system was constructed under the influence of atmospheric pressure at precise temperatures of 303.15 K, 313.15 K, and 323.15 K. The equilibrium components of each substance were analyzed by employing high-temperature gas chromatography, a sophisticated analytical method that enables the identification and quantification of individual components of a sample. Moreover, the ternary liquid–liquid equilibrium data were correlated by implementing the NRTL and UNIQUAC activity coefficient models. Subsequently, the binary interaction parameters of the ternary system were derived by conducting regression analysis. The experimental data demonstrated that the presence of lower methanol content in the system resulted in nearly immiscible biodiesel and glycerol phases, which ultimately facilitated the separation of biodiesel and glycerol. Conversely, with the increase in methanol content, the mutual solubility of biodiesel and glycerol was observed to increase gradually. The results showed that the calculated values of the NRTL and UNIQUAC models aligned well with the experimental values. The root-mean-square deviations of the NRTL and UNIQUAC models at 313.15 K were 2.76% and 3.56%, respectively.

Keywords: liquid–liquid equilibrium; waste oil biodiesel; NRTL modeling; UNIQUAC model



Citation: Yang, L.; Xing, S.; Teng, X.; Liu, R.; Wang, Z.; Lin, B.; Lv, P.; Al-Haimi, A.A.N.M.; Yehia, F.; Luo, W. Liquid–Liquid Equilibrium Behavior of Ternary Systems Comprising Biodiesel + Glycerol and Triglyceride + Methanol: Experimental Data and Modeling. *Catalysts* **2024**, *14*, 320. <https://doi.org/10.3390/catal14050320>

Academic Editor: Valeria La Parola

Received: 30 March 2024

Revised: 7 May 2024

Accepted: 8 May 2024

Published: 12 May 2024



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1. Introduction

In the contemporary era, biodiesel has drawn considerable attention around the globe as a substitute fuel for diesel engines. With an increasing awareness of the environmental impact of fossil fuels and the need for more sustainable and renewable sources of energy, biodiesel has emerged as a promising solution [1]. One of the critical benefits of biodiesel is that it is non-toxic and biodegradable, which makes it a much safer and cleaner fuel option. Additionally, biodiesel is free of aromatics and sulfur, toxic chemicals commonly found in diesel fuel, which contribute to air pollution and can have harmful health effects [2]. Typically, it is produced by converting vegetable or waste oil with homogeneous basic catalysts through transesterification and has similar combustion characteristics to fossil

materials [3]. Biodiesel is an excellent alternative to petroleum fuels, which can alleviate the scarcity of fossil energy resources and environmental problems [4,5]. As valuable feedstock in the production of biodiesel, waste oils hold considerable importance due to the majority of these oils possessing a high acid value, necessitating preliminary transformation through esterification via acid catalysts before the commencement of the alkali-catalyzed transesterification reaction, which ultimately results in the production of biodiesel [6–8]. The reaction occurs in short-chain alcohols, namely ethanol or methanol, and employs homogeneous or heterogeneous catalysts. The resultant by-product that ensues from the reaction is glycerol [9]. In the process of industrial biodiesel production, the mutual solubility of triglycerides, methanol, and biodiesel systems directly affects the efficiency of the reaction. In contrast, the miscibility of methanol, biodiesel, and glycerol is a determining factor in the separation and purification of transesterification [10]. The preparation of biodiesel encompasses the utilization of triglycerides and alcohol as reactants, which subsequently generate fatty acid esters and glycerol as products. Coupling the non-polar triglycerides with the polar nature of methanol to achieve mutual solubility between the two is a herculean feat. However, the reaction process is facilitated by studying the reaction system that encompasses these two substances. In the separation stage, the molecular discrepancies in size and polarity within the reaction mixture, constituting methyl ester and glycerol, impel the segregation of the separation of two phases. Nonetheless, the presence of methanol amplifies their solubility, thereby rendering the separation process more complex. Consequently, the separation and purification of these components are indispensable, as is the comprehensive study of the biodiesel + methanol + glycerol system.

The purification process that follows the transesterification reactions occupies a significant role in endowing biofuel with requisite levels of quality, adhering to the specifications of international standards [11]. The purification stage can be optimized using a liquid–liquid extraction process, typically involving aliphatic alcohols such as methanol or, more recently, ethanol. In this vein, studies have been carried out on the different ternary systems (biodiesel + glycerol + ethanol) to garner insights into the liquid–liquid equilibrium (LLE) that characterizes the process [12]. Consequently, it is vital to conduct a comprehensive analysis to purify the products after the reaction. The optimal design of the transesterification process and the post-separation process is the acquisition of thermodynamic data, particularly pertaining to phase equilibrium, which is indispensable for achieving the desired quality levels of the final product. Examining the thermodynamic principles governing the prediction of the equilibrium (LLE) data and phase behavior of the liquid flow is beneficial for the purification of biodiesel [13–15].

For biodiesel phase equilibrium systems, the UNIQUAC and NRTL models, both derivatives of the Wilson model, are the most frequently utilized. These models are applicable to similar systems, specifically for calculating and predicting liquid–liquid phase equilibria, including systems composed of ketones, alcohols, ethers, nitriles, and lipids, as well as miscible systems with water, sulfur, and halides [16]. The NRTL model [17], noted for its simplicity, incorporates a non-random parameter that is typically considered independent of both temperature and solution composition, but is determined by the type of solution, as a characteristic parameter of the solution. The specific selection of the solution value directly affects the fitting effect of the NRTL equation. In comparison, UNIQUAC is more complex [18]. It is more theoretical and involves more parameters, but its range of applicability is broader. Specifically, in terms of volume fractions, the UNIQUAC equation considers the effects of planarity and three-dimensionality; it also introduces lattice coordination numbers (similar to those of solids), molecular contact numbers, and other parameters. Many studies use the UNIQUAC or NRTL models to conduct related predictions on the liquid–liquid phase equilibrium of biodiesel + methanol (ethanol) + glycerol (water), mainly focusing on ternary systems using vegetable oils like soybean oil biodiesel or rapeseed oil biodiesel.

Accordingly, it is also necessary for undertakers, but its range of applicability is in the liquid phase equilibrium of this system. Many researchers have focused on the study of

the liquid–liquid phase equilibrium of biodiesel + methanol (ethanol) + glycerol (water), primarily centered around the ternary systems utilized for the production of biodiesel from vegetable oils such as soybean oil biodiesel or rapeseed oil biodiesel [19–24]. Mhadessi [25] reported using the UNIQUAC and NRTL models to conduct related predictions of the liquid–liquid phase equilibrium of different biodiesel + glycerol + alcohol; therefore, the experimental date was the previous date in the literature. However, research pertaining to the phase equilibrium of waste oil biodiesel + methanol + glycerol has been relatively limited.

This work involves the study of the phase equilibrium of the ternary system comprising waste oil biodiesel + glycerol + methanol, employing a liquid–liquid phase equilibrium kettle. The NRTL and UNIQUAC activation coefficient models were utilized to correlate the acquired experimental data, and subsequently obtain the corresponding simulation parameters and relative errors. This analysis is intended to provide a reliable theoretical basis for the purification of industrial waste oil biodiesel and glycerol.

2. Results and Discussion

2.1. Phase Equilibrium of Biodiesel + Methanol + Triglyceride Ternary System

The ternary phase equilibrium solubility curve of biodiesel + methanol + triglyceride (palm oil) was determined at two different temperatures: 313.15 K and 333.15 K. The cloud point method was utilized to implement the ternary two-phase liquid–liquid equilibrium experiment, which was used to obtain the equilibrium mass fraction of each component. Based on the obtained results, the triangular phase diagram of the ternary system was drawn, as illustrated in [Figure 1]. Figure 1 demonstrates that triglyceride and methanol are partially miscible. Compared to the solubility of methanol in triglyceride, the triglyceride in methanol has very low solubility. Nevertheless, introducing biodiesel improves the mutual solubility of triglyceride and methanol. Once the biodiesel reached a specific concentration, triglyceride, methanol, and biodiesel demonstrated complete mutual solubility, indicating that biodiesel can serve as a solvent for the system. Comparing the solubility curves at temperatures of 313.15 K and 333.15 K, the saturation solubility curve shifted downward from 313.15 K to 333.15 K. This led to a reduction in the two-phase region and an increase in the single-phase region. Additionally, it is worth noting that the three-phase composition at intercalation varied at different temperatures. Higher biodiesel concentrations are compulsory to achieve complete intercalation under reaction conditions of lower temperatures. Otherwise, a minor quantity of biodiesel is required to assemble a fully intercalated system in the presence of a low concentration of methanol. In contrast, increasing the concentration of methanol accomplishes the separation of the system phases easily and then requires further biodiesel as a solvent to conduct the intercalation process.

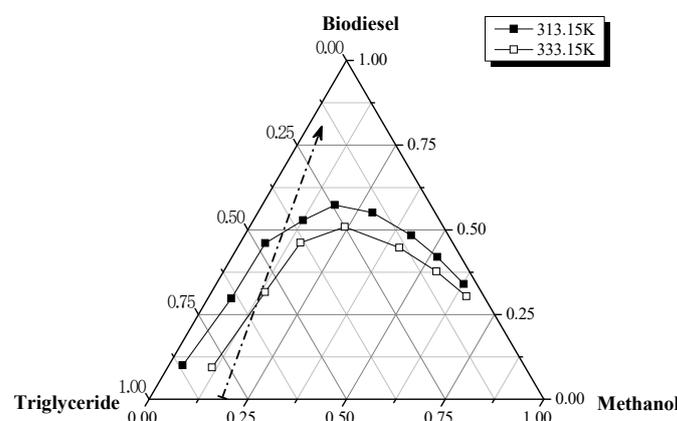


Figure 1. Ternary phase diagram of methanol + biodiesel + triglyceride at different temperatures.

2.2. Phase Equilibrium of Pure Methyl Ester + Methanol + Glycerol Ternary System

A comprehensive comparison of the fatty acid compositions of 11 oils, including salad oil, soybean oil, and vegetable oil, was investigated; the results are shown in Table S1.

The findings revealed that, except for canola oil, which was postulated to be primarily composed of erucic acid, all the others included palmitic acid, oleic acid, and linoleic acid as the predominant components. Therefore, the ternary liquid–liquid phase equilibrium performance of methyl ester (biodiesel) + methanol + glycerol was subsequently examined using methyl palmitate, methyl oleate, and methyl linoleate as pure substances to represent methyl esters. Figure 2 shows the phase diagram of the pure methyl ester + methanol + glycerol ternary system. The phase diagram has two regions separated by the saturation curve. The first in the two-phase region is the mixed liquid phase, which is stratified after standing, with the biodiesel phase in the upper layer and the glycerol phase in the lower layer. Methanol is distributed in the two phases with a specific distribution coefficient; due to the similar polarity of methanol and glycerol, most of the methanol is enriched in the glycerol phase. The other area is a single-phase region where the three substances are dissolved in one phase. It is considerably challenging to separate the phases and substances in this region. However, if the reaction conditions can reach this homogeneous phase condition, the reaction rate will be increased by orders of magnitude [19,25].

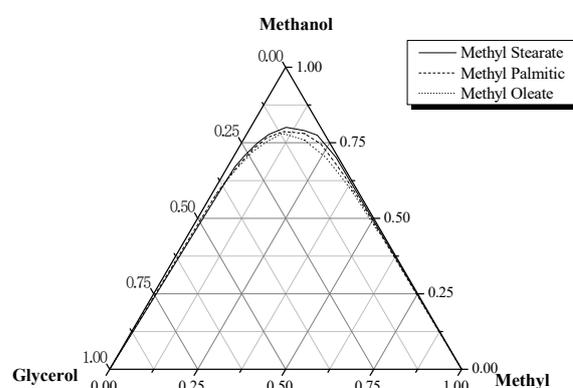


Figure 2. Phase diagram of pure methyl ester + methanol + glycerol ternary system at 313.15 K.

Biodiesel and glycerol are not mutually soluble. The mutual solubility of biodiesel and glycerol increases with increasing methanol content, i.e., methanol can be employed as a solvent to promote the mutual solubility of biodiesel and glycerol in the presence of a certain ratio of biodiesel and glycerol mixture. Conversely, if the methanol content is continuously decreased in the biodiesel + methanol + glycerol mixture, the mutual solubility of biodiesel and glycerol is continuously decreased, i.e., it is more favorable for the separation of biodiesel and glycerol. In the pure methyl ester + methanol + glycerol ternary system, also highlighted in Figure 2, the solubility properties of methyl palmitate, methyl oleate, and methyl linoleate are remarkably similar. Among them, methyl oleate has the best solvent properties, followed by methyl palmitate and, finally, methyl stearate. This phenomenon may be related to the viscosity of the three methyl, where the lower viscosity facilitates the movement between molecules and increases the mutual solubility [26].

The ternary liquid–liquid phase equilibrium data for methyl oleate + methanol + glycerol was analyzed at different temperatures: 313.15 K, 323.15 K, and 333.15 K, and the corresponding phase diagrams of the ternary system were plotted (Figures 3–5). The temperature affected the mutual solubility of the three substances. Specifically, the overall saturation solubility curve shifted downward, while the two-phase region shrank and the single-phase region increased with increasing temperature. The ternary system of methyl palmitate/methyl linoleate + methanol + glycerol reflects the same pattern (albeit not illustrated in the figure). The reason may be that the increase in temperature is favorable for the movement of molecules, which results in reducing the viscosity of the three substances, especially glycerol viscosity, which decreases by an order of magnitude. Moreover, the increase in temperature increases the average kinetic energy of the molecules of the three substances in the liquid phase, leading to more molecules participating in the hydrogen bonding of the molecules in this state, thereby increasing the mutual solubility [27,28].

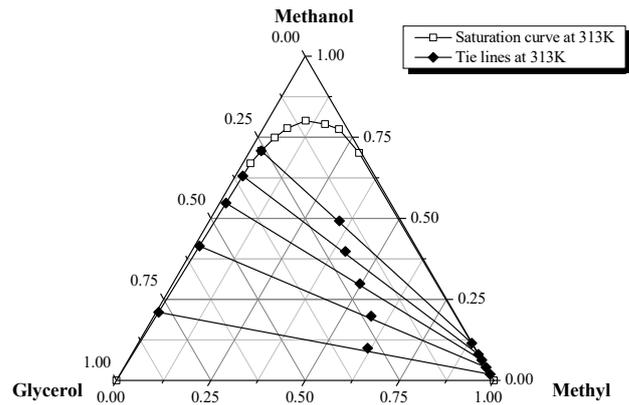


Figure 3. Solubility curve and linkage line of ternary system of methyl oleate + methanol + glycerol at 313.15 K.

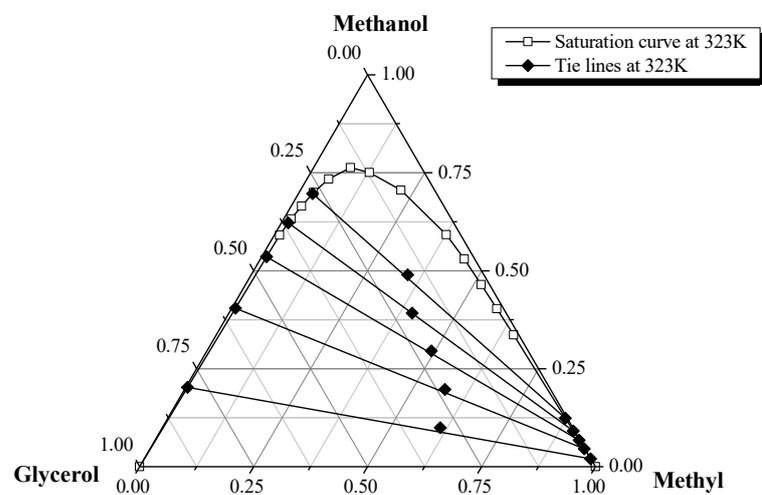


Figure 4. Solubility curve and linkage line of ternary system of methyl oleate + methanol + glycerol at 323.15 K.

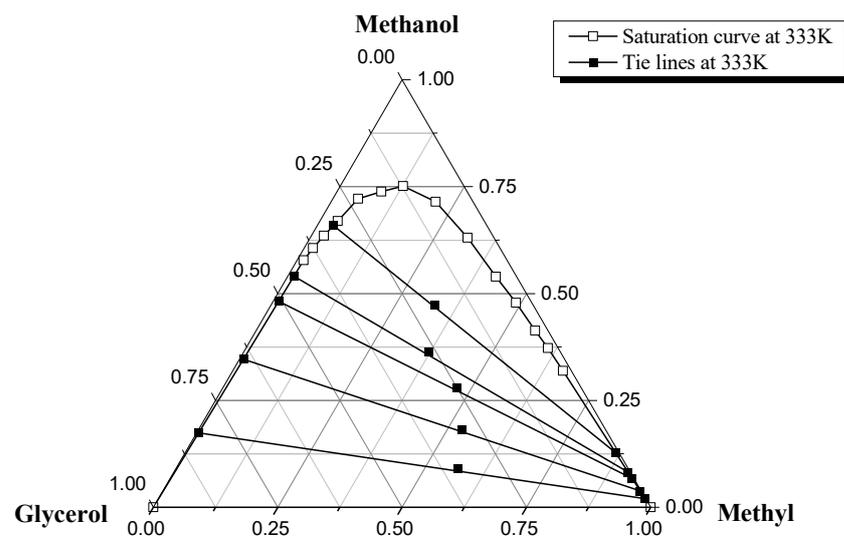


Figure 5. Solubility curve and linkage line of ternary system of methyl oleate + methanol + glycerol at 333.15 K.

The selectivity coefficient (referring to the difference in the solubility of biodiesel to methanol and glycerol) of biodiesel as a solvent in the system with pure methyl ester +

methanol + glycerol was analyzed. The results as shown in Figure 6 indicate that the increase in temperature improves the selectivity of biodiesel for methanol. Increasing the temperature is favorable for increasing the amount of methanol dissolved in biodiesel, thus facilitating the biodiesel reaction.

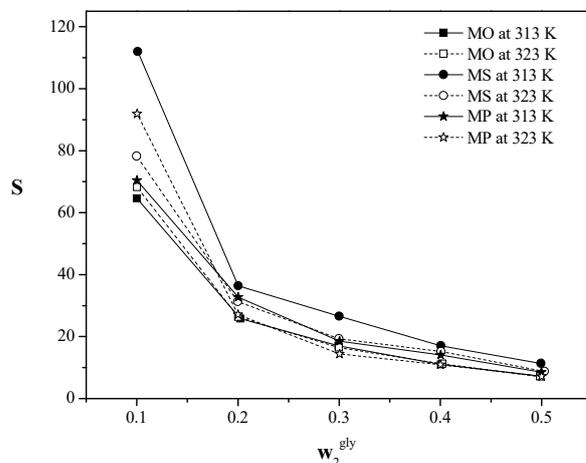


Figure 6. Comparison of biodiesel selectivity for pure methyl ester + methanol + glycerol system at different temperatures.

2.3. Different Biodiesel + Methanol + Glycerol Ternary Systems

The phase equilibrium data of the ternary system, including palm oil biodiesel/waste oil/soybean oil/blend of 70% soybean oil and 30% palm oil in conjunction with methanol and glycerol, were investigated and evaluated, charted, and visualized in [Figure 7]. The size of single-phase regions was classified from small to large in the following sequence: the palm oil system had the smallest one-phase zone, followed by the waste oil system, and the soybean oil system, with the blended oil system showing noticeably larger zones. It is significant to mention that the latter three are remarkably similar in the one-phase region in terms of size. This could be associated with the composition of the fatty acid contained in different oil sources, for example, palm oil containing 40.1% palmitic acid. On the other hand, the waste oil was mainly composed of methyl linoleate and methyl oleate; therefore, the viscosity of palm oil biodiesel was higher than that of waste oil biodiesel and was not conducive to the movement of molecules. As a result, miscibility was reduced. This is consistent with the results of the previously mentioned pure methyl ester system.

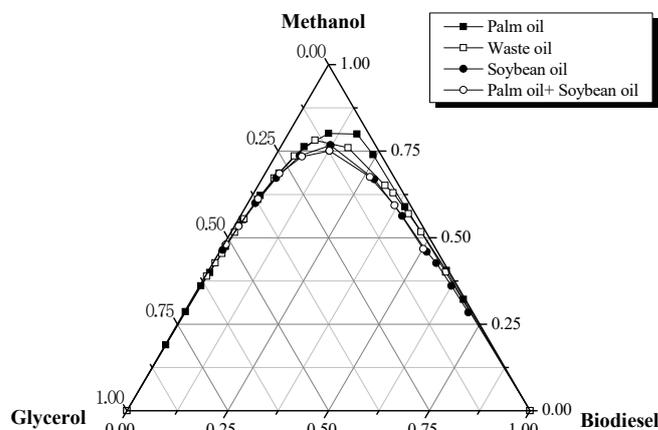


Figure 7. Ternary system phase diagram of biodiesel + methanol + glycerol from different oil sources.

2.4. Analysis of Liquid Phase Equilibrium Data for Waste Oil Biodiesel + Methanol + Glycerol Ternary Liquid

The main methyl ester components of waste oil biodiesel were determined using gas chromatography, and the obtained results are reported in Table S1 in the Supporting Information, revealing that the waste oil biodiesel included saturated fatty acids and unsaturated fatty acids with 24.14% and 74.81% of the methyl ester, respectively. The component of saturated fatty acid methyl ester was mainly methyl palmitate, which made up 16.83 wt% of the biodiesel. On the other hand, the majority of the unsaturated fatty acid methyl ester was methyl oleate (C18:1) and methyl linoleate (C18:2), accounting for 45.07 wt% and 24.82 wt% of the biodiesel, respectively. According to the aforementioned experimental procedure, the liquid–liquid phase equilibrium data for the ternary system of waste oil biodiesel–methanol–glycerol at 303.15–323.15 K were analyzed under atmospheric pressure. The resulting data are presented in Tables S2–S4 of the Supporting Information.

Accordingly, the system consisting of waste oil biodiesel and glycerol was almost insoluble in waste oil biodiesel + methanol + glycerol. However, the increase in methanol content enhanced the mutual solubility of glycerol and waste oil biodiesel. The content of methanol in the glycerol phase was significantly higher than that of methanol in the waste oil biodiesel phase. This occurred because the strong hydrogen bonding between methanol and glycerol molecules made methanol dissolve mostly in the glycerol. As a result, the process of separating glycerol from biodiesel as a by-product was more complicated than that of biodiesel as the main product. Therefore, the effect of temperature on the phase equilibrium of this liquid–liquid system was insignificant at a temperature range of 303.15–323.15 K.

2.5. Liquid Phase Equilibrium Modeling of Waste Oil Biodiesel + Methanol + Glycerol Ternary Liquid

The model parameters and RMSD obtained from regression analysis of experimental data of the ternary liquid system using UNIQUAC and NRTL models at different temperatures are presented in Table 1. The obtained results show that the NRTL model is better than the UNIQUAC model at regression estimation of the system. The RSMD values of the NRTL and UNIQUAC models at 313.15 K were 2.76% and 3.56%, respectively. Figure 8 illustrates the solubility curves at 313.15 K. After the simulated temperature reached 323.15 K, the relative accuracies of the results of the UNIQUAC and NRTL models were larger than 5% due to the volatility of methanol at the high temperature.

Table 1. Model parameters and RSMD of waste oil biodiesel (1) + methanol (2) + glycerol (3) Ternary liquid–liquid system at different temperatures.

Temperature (K)	i–j	UNIQUAC		RSMD (%)	α	NRTL		RSMD (%)
		τ_{ij}	τ_{ji}			τ_{ij}	τ_{ji}	
303.15	1–2	0.389	1.235	4.98	0.2	−1.054	4.341	3.95
	1–3	0.146	1.176		0.2	5.754	4.736	
	2–3	3.543	0.647		0.2	1.789	−1.987	
313.15	1–2	0.305	1.103	3.56	0.2	−3.146	4.975	2.76
	1–3	0.102	0.968		0.2	6.459	6.341	
	2–3	3.561	0.075		0.2	2.769	−0.916	
323.15	1–2	0.891	0.755	5.34	0.2	−6.785	5.246	5.67
	1–3	0.156	1.235		0.2	5.679	5.976	
	2–3	2.316	1.479		0.2	1.598	−8.137	

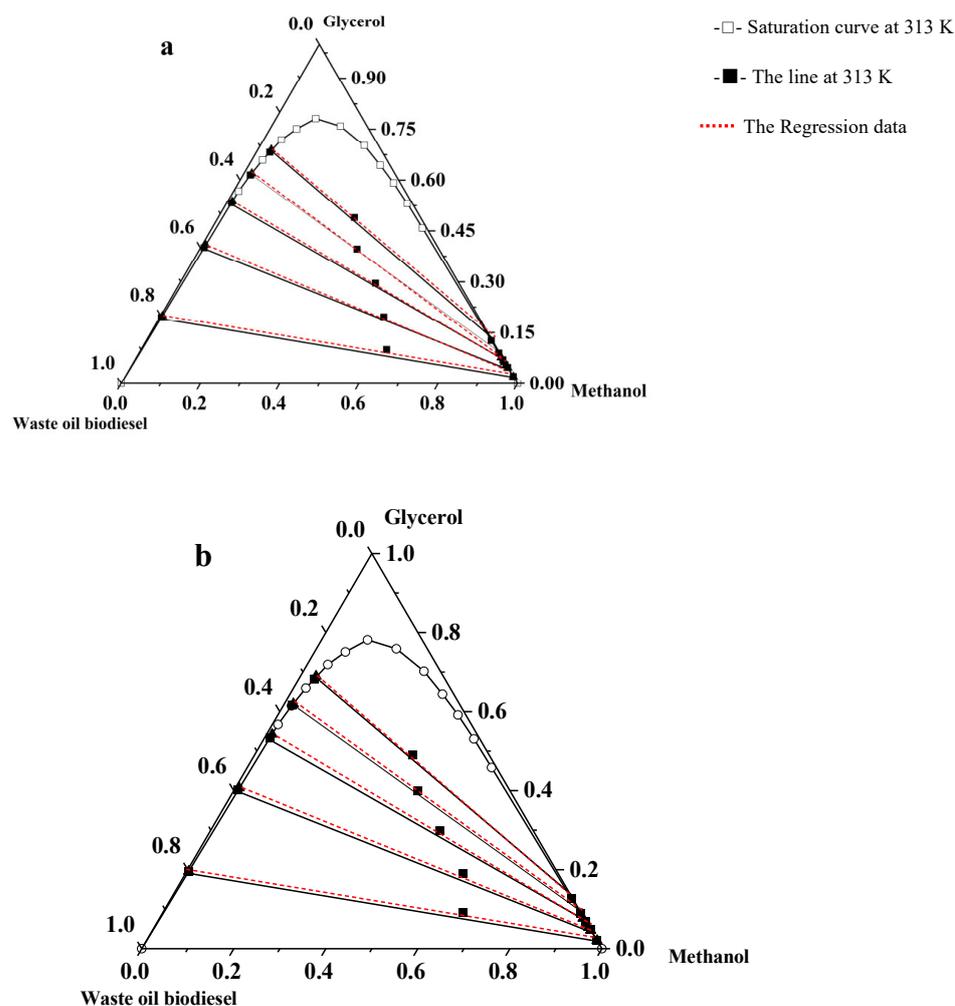


Figure 8. Liquid–liquid equilibria for the system containing waste oil biodiesel + methanol + glycerol at 313.15 K. (a) NRTL model; (b) UNIQUAC model.

3. Materials and Methods

3.1. Materials and Device

The main component used in this work consists of waste cooking oil as a feedstock; its composition is enumerated in Table S1 of the Supporting Information. It was purchased from Maoming Hongyu New Energy Co., Ltd. (Maoming, China). Other chemicals, including glycerol (99.9%), methanol (99.9%), methyl oleate (96%), methyl stearate (96%), methyl palmitic (99%), glycerol (99%), methanol (99%), sulfuric acid(99%), and sodium hydroxide (96%) were procured from Shanghai Aladdin Biochemical Technology Co., Ltd. (Shanghai, China). To reduce the impact of water, glycerol was subjected to a moderate temperature of 353.15 K for several days. No additional purification steps were performed for methanol and the three methyl types that were previously mentioned.

Intelligent thermostatic oscillator (Jiangsu Jinyi Instrument Technology Co., Ltd., Jintan, China, Made in 2021), digital display temperature controller (Shanghai Jiapai Technology Co., Ltd., Shanghai, China, Made in 2022), analytical balance (Beijing Sedolis Instrument Co., Ltd., Beijing, China, Made in 2019), Miniature injector, conical bottle, sampling bottles (Sigma Aldrich Shanghai Trading Co., Ltd., Shanghai, China, Made in 2022), and GC-2010Plus gas chromatograph (Shimadzu Instruments, Tokyo, Japan, Made in 2018) were also used.

3.2. Saturation Curves of the Glycerol + Methanol + Methyl Ester System

The phase boundaries for each ternary system were determined at three different temperatures (303.15, 313.15, and 323.15 K) by employing the titration approach under isothermal conditions. Typically, glycerol is favored as a titration reagent for obtaining the glycerol saturation curve in the methyl-rich phase [29]. However, its high viscosity presents a challenge to performing the titration. Hence, turbidimetric analysis is employed to determine the methyl-rich phase. A prescribed amount of methyl-containing glycerol was added to a flask immersed in a water bath of constant temperature and subjected to an uncertainty temperature of ± 0.2 K. Subsequently, this mixture was titrated with methanol and simultaneously stirred using a magnetic stirrer until a perceptible transformation from turbid to transparent was discernible. The discernible transformation from a turbid two-phase region to a transparent one-phase region is conventionally regarded as the saturation point of glycerol in the methyl + methanol mixture. Similarly, in the case of the glycerol-rich phase, a combination of glycerol-containing methyl was titrated with methanol until the mixture turned transparent. The corresponding solubility was subsequently ascertained based on the addition of each component. Notably, uncertainty in the mass fraction of each component was estimated to be less than 10^{-3} .

3.3. Tie Lines of the Glycerol + Methanol + Methyl System

In order to simulate the actual process, glycerol, methanol, and methyl were mixed at a certain proportion and vigorously stirred for 3 h in an isothermal water bath kettle. Then, the resulting mixture was subjected to a constant temperature for 24 h to provide an adequate time to facilitate the complete separation of the solution into two phases. Samples were carefully removed from the methyl-rich and glycerol-rich phases using syringes. The samples were weighed and then placed in an oven at a temperature of 348.15 K for 5 h to evaporate the methanol. According to the phase rule, at a specified temperature and pressure, only the independent variation of one component can be determined. Consequently, the points representing the composition of each phase were determined by measuring the quantity of removed methanol with uncertainties less than 10^{-3} g and employing the bimodal curves obtained in the previous section. In a similar vein, to obtain the other tie lines, the procedure was duplicated by changing the relative proportion of the three compounds.

3.4. Analysis of Fatty Acid Methyl Esters

The fatty acid methyl esters and methanol biodiesel components were evaluated qualitatively and quantitatively using a Shimadzu GC2010 gas chromatograph from Tokyo, Japan. The temperature control accuracy of chromatography gave a significant degree of precision, with an error margin of 0.1 K. The internal standard approach was the analytical technique that was utilized, and the correlation of the internal standard curve revealed a remarkable coefficient of correlation exceeding 0.999. The operating conditions of the gas chromatograph were as follows: GC-2010Plus gas chromatograph from Shimadzu Instruments, Japan, with an FID hydrogen flame ion detector and a DB-5HT capillary column with the dimensions $15\text{ m} \times 0.25\text{ mm} \times 0.10\text{ }\mu\text{m}$. The flow rates of nitrogen, air, and hydrogen were 15 mL/min, 20 mL/min, and 20 mL/min, respectively. The column temperature was set at 423.15 K and maintained for 1 min. Subsequently, a ramp rate of 5 K/min was initiated to reach a temperature of 463.15 K, where it was maintained for 1 min. The inlet temperature was set to 623.15 K while the detector temperature was set to 648.15 K. For constituents with insignificant quantities, the method of averaging multiple measurements was implemented to minimize the error.

3.5. Liquid–Liquid Equilibrium Calculation

In this study, Aspen Plus V.12 software was used for data fitting and regression with the parameters of the UNIQUAC and NRTL coefficient models. The NRTL and UNIQUAC activity coefficient models are widely employed in chemical thermodynamic studies for phase equilibria calculations and materials behavior prediction and/or correlation [30–32].

In this study, those equations were utilized to correct the ternary liquid–liquid equilibrium data of waste oil biodiesel + methanol + glycerol. Additionally, the correctness of the experimental results was checked using the root-mean-square deviation (RSMD) computation using the following formula.

$$\text{RSMD} = \left[\frac{\sum_{i=1}^3 \sum_{j=1}^2 \sum_{k=1}^M (x_{ik}^j - \hat{x}_{ik}^j)^2}{6M} \right]^{1/2} \quad (1)$$

4. Conclusions

This research entailed a comparative and analytical evaluation of the liquid–liquid equilibrium (LLE) behavior of the distinct ternary systems of biodiesel + methanol + triglyceride. In addition, the UNIQUAC and NRTL models were used to correlate phase equilibrium data for waste oil biodiesel + methanol + glycerol ternary system at different temperatures within the range of 303.15 K–323.15 K. The miscibility of the system was affected by the contents of methanol and biodiesel; increasing the amount of biodiesel promoted the miscibility of glycerol and methanol. Thus, in the biodiesel production system, a judicious amount of biodiesel was ultimately required to improve the degree of system miscibility and strategically enhance the reaction efficiency. Moreover, methanol played a vital role in achieving the targeted miscibility of biodiesel and glycerol. The separation of biodiesel from a mixture of reactant and product should prioritize the removal of methanol as a preliminary step, resulting in the successful separation of biodiesel and glycerol. The use of the UNIQUAC and NRTL models for LLE characterization demonstrated commendable congruity between the experimental data and the calculated values for biodiesel + glycerol + methanol. Notably, the NRTL model gave a superior description, with a maximum RSMD of 5.26%.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/catal14050320/s1>, Table S1: The FAME composition of biodiesel produced from different feedstock. Table S2: Experimental liquid–liquid equilibrium data for the ternary systems containing waste cooking oil biodiesel + methanol + glycerol at 303.15 K. Table S3: liquid–liquid equilibria data for the ternary systems containing waste oil biodiesel + methanol + glycerol at 313.15 K. Table S4: Experimental liquid–liquid equilibria data for the ternary systems containing waste oil biodiesel + methanol + glycerol at 323.15 K.

Author Contributions: Conceptualization, L.Y.; methodology, S.X.; software, B.L.; validation, X.T., and P.L.; formal analysis, L.Y. and W.L.; investigation, Z.W.; resources, L.Y. and S.X.; data curation, L.Y. and W.L.; writing—original draft preparation, L.Y., F.Y. and W.L.; writing—review and editing, L.Y. and W.L.; visualization, A.A.N.M.A.-H.; supervision, R.L.; project administration, R.L.; funding acquisition, R.L. and W.L. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the National Natural Science Foundation of China (Nos. 32271822, 51606201).

Data Availability Statement: Data are contained within the article and Supplementary Materials.

Conflicts of Interest: The authors declare no conflict of interest.

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