Effect of Biodiesel Mixture Derived from Waste Frying-Corn, Frying-Canola-Corn and Canola-Corn Cooking Oils with Various Ages on Physicochemical Properties

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Received: 27 August 2019; Accepted: 27 September 2019; Published: 29 September 2019

Abstract: Waste frying, corn and canola cooking oil biodiesels were produced through the transesterification process and their properties were measured. Three different mixtures of biodiesel with the same blending ratio, namely, WCME1 (frying-corn biodiesel), WCME2 (frying-canola-corn biodiesel) and WCME3 (canola-corn biodiesel), were prepared. The effect of blending biodiesel with various ages (zero months (WCME3), eight months (WCME1), and 30 months (WCME2)) on kinematic viscosity and density was investigated under varying temperature and volume fraction. It was found that the kinematic viscosity of WCME2 remained within the ranges listed in ASTM D445 (1.9–6.0 mm²/s) and EN-14214 (3.5–5.0 mm²/s) at 30 months. It was also observed that both viscosity and density decreased as the temperature increased for each fuel sample. In order to improve the cold flow properties of the samples, the Computer-Aided Cooling Curve Analysis (CACCA) technique was used to explore the crystallization/melting profiles of pure methyl biodiesel as well their blends. The results show that pure WCME2 has the lowest cold flow properties compared to other samples. Furthermore, 10 correlations were developed, tested and compared with generalized correlations for the estimation of the viscosity and densities of pure biodiesels and their blends. These equations depend on the temperature and volume fraction of pure components as well as the properties of the fuel.

Keywords: biodiesel properties; computer aided cooling curve analysis; correlations; response surface methodology; various ages; waste vegetable cooking oils

1. Introduction

Due to the population growth and changes in lifestyle, the interest in waste cooking oil has increased rapidly. Therefore, environmental recycling has become important for effective waste management. The major source for energy comes from fossil fuels. In fact, global warming is causing environmental pollution, which is largely produced by fossil fuels [1]. Therefore, renewable and natural sources such as vegetable oil and fats are considered as an alternative sources to replace fossil fuels [2,3]. Biodiesel is recognized as an alternative fuel that can replace fossil fuels. Biodiesel is a renewable resource that is biodegradable and environmentally friendly [4]. Biodiesel is a fuel consists of mono-alkyl-esters of long-chain fatty acids, derived from vegetable oils or animal fats [5]. It has a higher viscosity [6], cloud point, and pour point temperatures [7] compared to diesel fuel. Various research studies have shown that biodiesel has received significant attention and it is a possible alternative fuel [8].
The most significant properties of biodiesel are viscosity and density, because they affect the atomization quality, the size of fuel droplets and jet penetration, all of which affect the quality of combustion and engine performance parameters. Several studies have been conducted to investigate the effects of biodiesel blends on the performance of engines [9–13].

Moreover, cold weather can affect the performance of engines because the crystallization of high melting saturated fatty acid methyl esters may lead to the plugging of filters and tubes [14,15]. Generally, the cloud point (CP), the cold filter plugging point (CFPP) and the pour point (PP) are the classical cold flow properties of biodiesel [16]. In fact, it has been well-established that a higher amount of saturated esters increases the CP and PP of biodiesel [17,18].

Recently, waste cooking/frying vegetable oils (WCVOs) have frequently been used to prepare biodiesel [19] fuels due to their low cost and high availability [20]. According to References [21–23], the cost of WCVOs is two or three times lower than refined vegetable oils. Utilizing WCVOs as a fuel could effectively reduce the cost of removing and treating residual oil [24–27]. In addition, due to chemical reactions during the food cooking process or raw food, the WCVOs contains other compounds such as water, free fatty acids (FFA), polar compounds and non-volatile compounds in addition to triacylglycerols, which mainly affect homogeneous catalytic transesterification reactions [28,29].

Therefore, the aim of this study was to expand on the previous analyses on the subject [30,31] specifically: a) the effect of a long-term storage period of eight months for WCME1 and 30 months for WCME2 at a constant temperature on biodiesel properties was explored, b) the influence of the blending of WCME1, WCME 2 and WCME3 on biodiesel properties including viscosity, density and cold flow properties was investigated, c) the accuracy of generalized correlations in the prediction of kinematic viscosity and density for wide ranges of temperature and volume fraction was improved and d) the predictive capability of obtained correlations in estimating the properties of biodiesel blends in terms of kinematic viscosity and density were examined. To the best of the authors’ knowledge, this is the first study to investigate the properties of a biodiesel mixture derived from waste frying-corn, frying-canola-corn and canola-corn cooking oils with various ages.

2. Materials and Methods

The objective of the study was to highlight fuel-aging behavior by measuring the fuel properties such as kinematic viscosity, density and cold flow properties. Section 2.1 describes the preparation process of pure biodiesel samples and their properties according to ASTM standards. Section 2.2 discusses the storage test procedures for WCME1 and WCME2. The procedure for measuring the biodiesel samples, including kinematic viscosity, density and cold flow properties, is explained in Sections 2.3 and 2.4. Moreover, the solidification characteristics of biodiesel samples through computer-aided cooling curve analysis (CA-CCA) are described in Section 2.5. Section 2.6 presents the empirical models that were used to estimate the kinematic viscosity and the density of fuel samples. The schematic flow of this research work is depicted in Figure 1.
2.1. Biodiesel Sample Preparation

Waste frying (WF), canola (WCA) and corn (WCO) cooking oils were used in this study and were collected from different domestic sources like cafés and restaurants. Three different biodiesel fuels, namely, waste frying methyl ester (WFME), waste canola methyl ester (WCAME) and waste corn methyl ester (WCOME), were prepared by a methanol-based transesterification process. The production of biodiesel was discussed in Reference [30]. The detailed specifications of the three pure biodiesels including the fatty acid composition and most important properties are summarized in Tables 1 and 2. In this experiment work, three different mixtures of waste cooking methyl ester (WCME) with significantly different compositions (WCME1, WCME2 and WCME3) were prepared. These mixtures were prepared as follows:

![Flowchart of research works in this study](image-url)
WCME1 was prepared by mixing 50 vol% WFME and 50 vol% WCAME.

WCME2 was obtained by blending 35 vol% WFME, 35 vol% WCAME and 35 vol% WCOME.

WCME3 was the blended using 50 vol% CAME and 50 vol% WCOME.

These biodiesels (WCME1, WCME2 and WCME3) had different ages, i.e., the ages of WCME1, WCME2 and WCME3 were eight months, 30 months, and zero months, respectively. Nine blends of three biodiesel were tested in this study, possessing 0%, 25%, 50%, 75% and 100% biodiesel by volume basis. The chemical composition of the biodiesel samples is presented in Table 1. The analysis of biodiesel characteristics by gas chromatography is discussed in Reference [30]. The properties of the biodiesels are shown in Table 2 and compared with biodiesel specifications according to ASTM D6751.

Table 1. Acid methyl ester composition (wt%) of pure biodiesel and their mixture fuels.

| wt%     | Pure Biodiesel | Mixture of Biodiesel |
|---------|----------------|----------------------|
|         | WFME (0 months) | WCAME (0 months) | WCOME (0 months) | WCME1 (8 months) | WCME2 (30 months) | WCME3 (0 months) |
| C8:0    | 0.05           | 0.00                | 0.29              | 0.00             | 0.00              | 0.15              |
| C10:0   | 0.33           | 0.00                | 0.32              | 0.00             | 0.00              | 0.16              |
| C12:0   | 1.18           | 0.08                | 4.03              | 0.10             | 0.18              | 2.06              |
| C14:0   | 0.10           | 0.00                | 2.10              | 0.70             | 0.67              | 1.05              |
| C16:0   | 36.29          | 13.50               | 13.73             | 24.80            | 28.53             | 13.62             |
| C16:1   | 0.00           | 0.00                | 0.90              | 1.00             | 0.32              | 0.45              |
| C16:2   | 0.00           | 0.00                | 0.00              | 0.20             | 0.00              | 0.00              |
| C17:0   | 0.00           | 0.00                | 0.00              | 0.10             | 0.00              | 0.00              |
| C18:0   | 4.04           | 2.19                | 5.10              | 5.10             | 3.91              | 3.65              |
| C18:1   | 40.30          | 57.33               | 46.17             | 41.60            | 44.36             | 51.75             |
| C18:2   | 17.53          | 20.47               | 20.10             | 22.30            | 18.58             | 20.28             |
| C18:3   | 0.18           | 5.29                | 5.10              | 2.90             | 2.70              | 5.20              |
| C20:0   | 0.00           | 0.35                | 0.53              | 0.40             | 0.35              | 0.44              |
| C20:1   | 0.00           | 0.78                | 0.76              | 0.40             | 0.40              | 0.77              |
| C22:0   | 0.00           | 0.00                | 0.00              | 0.10             | 0.00              | 0.00              |
| MUFAMEs | 40.30          | 58.11               | 47.83             | 43.00            | 45.08             | 52.97             |
| PUFAMEs | 17.71          | 25.76               | 25.20             | 25.20            | 21.28             | 25.48             |
| SFAMEs  | 41.61          | 16.12               | 25.49             | 31.20            | 33.64             | 20.82             |

MUFAMEs: Monounsaturated FAMEs; \( \sum MUFAMs = C_{16} : 1 + C_{18} : 1 + C_{20} : 1 \)

PUFAMEs: Polyunsaturated FAMEs; \( \sum PUFAMs = C_{18} : 2 + C_{18} : 3 \)

SFAMEs: Saturated FAMEs; \( \sum SFAMs = C_{12} : 0 + C_{14} : 0 + C_{18} : 0 + C_{20} : 0 + C_{22} : 0 \)

Table 2. Pure biodiesel and their mixtures according to the ASTM D6751 biodiesel standard.

| Property                        | Unit    | Test Method | Limits     | WCME1 | WCME2 | WCME3 |
|---------------------------------|---------|-------------|------------|-------|-------|-------|
| Kinematic viscosity at 40 °C    | mm²/s   | ASTM D445   | 1.9–6.0    | 4.67  | 4.86  | 4.35  |
| Density at 15 °C                | kg/m³   | ASTM D854   | 867 min.   | 876.4 | 895.8 | 912.7 |
| Cold Point                      | °C      | ASTM D2500  | Report     | 16.5  | −1.0  | −7.5  |
| Pour Point                      | °C      | ASTM D97    | Report     | 10.5  | −11.0 | −12.0 |
| Acid value                      | mg KOH/g| ASTM D664   | 0.5 max.   | 0.37  | 0.42  | 0.41  |
| Oxidation Stability (at 110 °C) | h       | EN 14112    | 3.0 min.   | 7.56  | 7.25  | 9.45  |

| Property                        | Unit    | Test Method | Limits     | WCME1 | WCME2 | WCME3 |
|---------------------------------|---------|-------------|------------|-------|-------|-------|
| Kinematic viscosity at 40 °C    | mm²/s   | ASTM D445   | 1.9–6.0    | 4.46  | 4.46  | 4.53  |
| Density at 15 °C                | kg/m³   | ASTM D854   | 867 min.   | 917.10| 918.03| 910.88|
| Cold Point                      | °C      | ASTM D2500  | Report     | 5.2   | −2.0  | 4.5   |
| Pour Point                      | °C      | ASTM D97    | Report     | 4.6   | −2.3  | 4.2   |
| Acid value                      | mg KOH/g| ASTM D664   | 0.5 max.   | 0.51  | 0.90  | 0.30  |
| Oxidation Stability (at 110 °C) | h       | EN 14112    | 3.0 min.   | 5.0   | 2.2   | 14.0  |
2.2. Storage Test Procedures

Figure 2 shows a conceptual drawing of the storage tank used to keep the biodiesel samples in the current study. The tank was thermally isolated from any heat transfer by 11 cm thick Styrofoam layers. It was used for long-term storage of the biodiesel samples under an appropriate constant temperature (24 ± 1 °C). For storage of the biodiesel, 1000 mL glass bottles, because this material would not react with the biodiesel and they can be used for storage under different conditions. The bottles were left open for one day and cleaned with acetone before being filled with fuel sample. Additionally, the biodiesel samples were filled up to half of the total volume of the bottle and kept in a dark place to avoid contact with the metal lid closures of the bottle and with any light. The samples were stored for various time periods and the kinematic viscosity, density and cold flow properties were periodically measured.

![Figure 2. Glass bottles in test apparatus.](image)

2.3. Kinematic Viscosity and Density Measurement

The kinematic viscosity of biodiesel was measured using two Ubbelohde viscometers with various technical specifications following the ASTM D445 standard [32]. Furthermore, the density of the samples was measured using a Pycnometer with a bulb capacity of 25 mL. The measurement was made according to ASTM D854 [33]. The properties were measured at various temperature ranges, i.e., high temperature ranges between 20–80 °C in intervals of 10 °C and low-temperature ranges of −10–20 °C in intervals of 2 °C. The weighing was performed with an electronic balance with a precision of ± 0.1 mg. To reduce the experimental error, the test was repeated three–four times, and then the average was calculated. The experimental setup used to measure the biodiesel and density at various temperature ranges is described in References [30,31].

2.4. Cold Flow Measurement

The measurements of Cloud point (CP), Pour Point (PP) and Cold Filter Plugging Point (CFPP) were made according to ASTM D2500 [34], ASTM D97 [35] and ASTM D6371 [36], respectively. For a greater degree of accuracy, PP measurements were made with a resolution of 1 °C instead of the specified increment of 3 °C. The description of the CP and PP measurement process is explained in Reference [30]. Additionally, the description of the CFPP measurement procedure is explained in Reference [37]. In this study, the cold flow properties were manually measured, i.e., the solidification process was followed visually. To increase the accuracy of the results, the measurement was repeated two to three times for the biodiesel sample (each time, the used biodiesel was disposed and replaced with new sample).
2.5. Computer-Aided Cooling Curve Thermal Analysis (CA-CCA)

The major solidification parameters, including latent heat, critical points of phase transformation and fraction solid during the solidification process can be presented through CA-CCA [38]. CA-CCA is categorized into Newtonian and Fourier analysis [39].

Figure 3 schematically presents the experimental setup for CA-CCA. An Ordel Data Logger with five channels was used to record the temperatures (the temperature of the biodiesel sample ($T_1$) and cooling bath temperature ($T_2$)), as shown in Figure 3. Ethyl alcohol was used as the coolant liquid in this study due to its low freezing temperature (−114.1 °C). The cooling bath was insulated with 11 cm thick Styrofoam to prevent heat transfer between the coolant and the environment. Additionally, two T-type thermocouples were used to measure the temperature of the sample and the cooling bath. It should be noted that both thermocouples were calibrated with reference to the standard thermometer. The difference between the temperatures of both thermocouples was found to be ±0.1 °C. Moreover, a thermostat was used to control the temperature of the bath. To ensure the temperature of the bath was uniform and homogenous, a thermostat wire was utilized to measure the temperature of the bath and compare it with $T_2$. As shown in Figure 3, the compression unit consisted of a compressor, temperature display and control unit. The compressor cooled down the refrigerator gas and the control unit (thermostat) was adjusted to the required temperature for this study. A stirrer was used for thermal homogeneity of the ethanol in the cooling bath. In the current study, the cooling bath was cooled down to −20 °C and the biodiesel sample was heated up to 65 °C. The Newtonian analysis was performed with a sample size of 45 mL biodiesel in a glass test jar. An aluminum cylinder jacket was placed in the middle of the cooling bath. A 6 mm thick cork disk was placed at the bottom of the jacket as a thermal insulator. The glass test jar was filled with the biodiesel sample to a level of 54 mm corresponding to a sample volume of about 45 mL. Two gaskets were placed into the test jar to ensure the jacket fitted jacket and there was a uniform air gap of 5 mm in the radial direction between the test jar and the jacket. The Newtonian technique based on CA-CCA used to characterize properties of the material is discussed in detail in References [37–39].

The heat balance equation for the solidifying can be rewritten as:

$$\frac{dQ}{dt} - MC_p \frac{dT}{dt} = UA(T - T_0) \quad (1)$$

where $M$ is the mass, $C_p$ is the specific heat of the sample, $U$ is the overall heat transfer coefficient, $A$ is the sample surface area, $T_0$ is the temperature of the cooling bath, $T$ is the temperature of the sample, $t$ is the time and $Q$ is the latent heat of solidification.

If no phase transformation occurs, $\frac{dQ}{dt} = 0$, then the cooling rate of the test sample (first derivative of the CC) can be written as:

$$\frac{dT}{dt} = - \frac{UA}{MC_p} (T - T_0) = Z_c \quad (2)$$

where $Z_c$ is the zero curve or baseline.

The difference between the integral areas of these two curves represents the latent heat ($L$) of solidification, as shown in Equation (3). The solid fraction ($f_s$) can be obtained from Equation (4):

$$L = C_p \int_{T_{solidus}}^{T_{liquidus}} (\frac{dT}{dt} - Z_c) dt \quad (3)$$

$$f_s = \frac{C_p \int_{T_{liquidus}}^{T} (\frac{dT}{dt} - Z_c) dt}{L} \quad (4)$$
2.6. Oxidative Stability and Acid Value

The induction periods (IP) and acid values (AV) of the biodiesel samples were measured according to EN 14112 and ASTM D664, respectively, and performed as described in Reference [30]. The IP and AV of the biodiesel samples were measured on a bimonthly basis.

2.7. Empirical Models

In the present research, evaluations and comparisons of the empirical models have been conducted for predicting the kinematic viscosity and density of biodiesel blends. The performances of the models were compared using statistical criteria including determination coefficient ($R^2$). Several empirical equations have been proposed in the literature to predict the kinematic viscosity and density of biodiesel blends, which are summarized in Table 3. As shown in Table 3, the correlations can be divided as follows:

- Equations (5), (6) and (17)–(21): viscosity ($\nu$) correlations as a function of temperature ($T$) only.
- Equations (12) and (14)–(16): viscosity ($\nu$) correlations as a function of volume fraction ($VF$) only.
- Equations (11), (22) and (27)–(31): viscosity ($\nu$) correlations as a function of temperature and volume fraction.
- Equations (7) and (9): viscosity ($\nu$) correlations as a function of temperature and viscosity of pure fuels.
- Equations (13) and (26): viscosity ($\nu$) correlations as a function of volume fraction and viscosity of pure fuels.
- Equations (32) and (35)–(37): density ($\rho$) correlations as a function of temperature ($T$) only.
- Equations (34) and (38)–(40): density ($\rho$) correlations as a function of temperature and volume fraction.
- Equation (33): density ($\rho$) correlations as a function of volume fraction and viscosity of pure fuels.
- Equations (41) and (42): viscosity as a function of density.
- Equations (43) and (44): density as a function of viscosity.
Table 3. Proposed correlations by scientific researchers.

| Equation Number | Investigators | Equation |
|-----------------|--------------|----------|
| 5               | Tate et al. [40], Andrade’s equation [41] | ln(ν) = A + B ln(T) |
| 6               | Yuan et al. [42] | ln(ν) = A + \frac{B}{T} |
| 7               | Moradi et al. [43] | \nu = \nu_1 \nu_2 \exp(A + \frac{B}{T}) |
| 8               | Mejia et al. [44] | ln(\nu) = A + B VF + C |
| 9               | Kassem and Çamur [30] | \nu = \nu_1 \nu_2 \exp(A + \frac{B}{T}) |
| 10              | Tesfa et al. [50] | ln(\nu) = A + B VF |
| 11              | Tat and Van Gerpen [53] | \nu = \nu_1 \nu_2 \exp(A + \frac{B}{T}) |
| 12              | Ramírez-Verduzco et al. [52] | \nu = \nu_1 \nu_2 \exp(A + \frac{B}{T}) |
| 13              | Kay mixing rule [54] | \rho = A + B/VF |
| 14              | Ramírez-Verduzco et al. [52] | \nu = \nu_1 \nu_2 \exp(A + \frac{B}{T}) |
| 15              | Gümüş and Bilgin [47] | \nu = A + B VF |
| 16              | Kassem and Çamur [30] | \nu = \nu_1 \nu_2 \exp(A + \frac{B}{T}) |
| 17              | Tat and Van Gerpen [53] | \nu = \nu_1 \nu_2 \exp(A + \frac{B}{T}) |
| 18              | Kay mixing rule [54] | \rho = A + B/VF |
| 19              | Ramírez-Verduzco et al. [52] | \nu = \nu_1 \nu_2 \exp(A + \frac{B}{T}) |
| 20              | Gümüş and Bilgin [47] | \nu = A + B VF |
| 21              | Kassem and Çamur [30] | \nu = \nu_1 \nu_2 \exp(A + \frac{B}{T}) |
| 22              | Tat and Van Gerpen [53] | \nu = \nu_1 \nu_2 \exp(A + \frac{B}{T}) |
| 23              | Kay mixing rule [54] | \rho = A + B/VF |
| 24              | Ramírez-Verduzco et al. [52] | \nu = \nu_1 \nu_2 \exp(A + \frac{B}{T}) |
| 25              | Gümüş and Bilgin [47] | \nu = A + B VF |
| 26              | Kassem and Çamur [30] | \nu = \nu_1 \nu_2 \exp(A + \frac{B}{T}) |
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| 42              | Tat and Van Gerpen [53] | \nu = \nu_1 \nu_2 \exp(A + \frac{B}{T}) |
| 43              | Kay mixing rule [54] | \rho = A + B/VF |
| 44              | Ramírez-Verduzco et al. [52] | \nu = \nu_1 \nu_2 \exp(A + \frac{B}{T}) |

A, B, C, D, E and F are constants

3. Results and Discussion

3.1. Effect of Storage Period on WCME1 and WCME2 Properties

This section explains the effects of the storage period on the properties (the kinematic viscosity, density, CP, CFPP, PP, induction period and acid value) of WCME1 and WCME2, as discussed below.

3.1.1. Kinematic Viscosity and Density of WCME1 and WCME2

The kinematic viscosity and density were determined at different temperatures over various periods at regular intervals. In this study, the biodiesel properties of WCME1 and WCME2 were measured at different temperature ranges (−5 °C–80 °C) and storage periods. The results demonstrated that viscosity and density decreased as the temperature increased and the kinematic viscosity and density increased when the storage period increased. Moreover, the kinematic viscosity and density
values of biodiesel samples (Table 4) were within the ranges specified in ASTM D445 and ASTM D854, respectively (Table 2). This shows that the properties of the biodiesel, samples increased when the storage period is changed. Moreover, for storage of 30 months, it is observed that the kinematic viscosities of WCME2 remained below the maximum limit specified in the biodiesel standards (EN 14214 [59], 3.5–5.0 mm²/s, and ASTM D445 [32], 1.9–6.0 mm²/s). Furthermore, to show the effects of the storage period on the properties of biodiesel, the increase ratio ($I_r$) was calculated using Equation (45) and summarized in Table 4. It was found that all $I_r$ values were greater than 1.00.

\[
I_r = \frac{\text{Biodiesel properties of a given sample at last month of storage}}{\text{Biodiesel properties of a given sample at zero month of storage}}
\]  

(45)

| Months | WCME 1 | WCME 2 |
|--------|--------|--------|
|        | Kinematic Viscosity | Density | Kinematic Viscosity | Density |
| 0      | 4.44   | 914.90 | 4.42    | 909.53 |
| 2      | 4.44   | 915.57 | 4.43    | 910.09 |
| 4      | 4.45   | 916.07 | 4.44    | 910.65 |
| 6      | 4.46   | 916.64 | 4.44    | 911.22 |
| 8      | 4.46   | 917.10 | 4.45    | 911.78 |
| 10     | -      | -      | 4.46    | 912.35 |
| 12     | -      | -      | 4.46    | 912.92 |
| 14     | -      | -      | 4.47    | 913.48 |
| 16     | -      | -      | 4.48    | 914.05 |
| 18     | -      | -      | 4.49    | 914.62 |
| 20     | -      | -      | 4.49    | 915.18 |
| 22     | -      | -      | 4.50    | 915.75 |
| 24     | -      | -      | 4.51    | 916.32 |
| 26     | -      | -      | 4.52    | 916.89 |
| 28     | -      | -      | 4.53    | 917.46 |
| 30     | -      | -      | 4.53    | 918.03 |
| $I_r$ (8 months) | 1.007  | 1.002  | 1.007  | 1.002 |
| $I_r$ (10 months) | -      | -      | 1.010  | 1.004 |
| $I_r$ (20 months) | -      | -      | 1.017  | 1.006 |
| $I_r$ (30 months) | -      | -      | 1.026  | 1.009 |

3.1.2. Cold Flow Properties of WCME1 and WCME2

The low-temperature properties including CP, CFPP and PP of WCME1 and WCME2 were measured bimonthly to determine the effects of cold-weather performance and storage period ($t$). Table 5 shows the values for the cold flow properties for two pure biodiesels over eight months for WCME1 and 30 months for WCME2. To show the effect of storage period on the cold flow properties of biodiesel values, Equation (46) was used to calculate the temperature differentials:

\[
\Delta T = T[t = 0] - T[t = 8 \text{ for WCME1 or } t = 30 \text{ for WCME2}]
\]  

(46)

where $T$ is the temperature value of CP, CFPP or PP and $t$ is the storage time in a month.

It was observed that storage for eight months or 30 months did not significantly influence the CP, CFPP or PP values of WCME1 and WCME2, respectively.
Table 5. Influence of storage period (months) on cold flow properties of WCME1 and WCME2.

| Month | WCME1 | WCME2 | ΔT [°C] |
|-------|-------|-------|---------|
|       | CP (°C) | CFPP (°C) | PP (°C) | CP (°C) | CFPP (°C) | PP (°C) | CP (°C) | CFPP (°C) | PP (°C) |
| 0     | 4.8    | 4.3   | −1.9    | −1.4   | −1.9   | −4.5    | −1.5    | −1.9    | −4.6    |
| 2     | 4.9    | 4.4   | −1.5    | −1.5   | −2.0   | −4.6    | −1.5    | −1.9    | −4.6    |
| 4     | 5.0    | 4.5   | −1.6    | −1.6   | −2.0   | −4.6    | −1.6    | −1.9    | −4.6    |
| 6     | 5.1    | 4.5   | −1.6    | −1.6   | −2.0   | −4.6    | −1.6    | −1.9    | −4.6    |
| 8     | 5.2    | 4.6   | −1.7    | −1.7   | −2.1   | −4.8    | −1.7    | −1.9    | −4.6    |
| 10    | −     | −     | −1.8    | −1.8   | −2.1   | −4.8    | −1.8    | −1.9    | −4.6    |
| 12    | −     | −     | −1.9    | −1.9   | −2.1   | −4.9    | −1.9    | −2.0    | −4.6    |
| 14    | −     | −     | −2.0    | −2.0   | −2.2   | −5.0    | −2.0    | −2.0    | −4.6    |
| 16    | −     | −     | −2.1    | −2.1   | −2.2   | −5.1    | −2.1    | −2.0    | −4.6    |
| 18    | −     | −     | −2.1    | −2.1   | −2.2   | −5.1    | −2.1    | −2.0    | −4.6    |
| 20    | −     | −     | −2.2    | −2.2   | −2.2   | −5.2    | −2.2    | −2.0    | −4.6    |
| 22    | −     | −     | −2.3    | −2.3   | −2.2   | −5.3    | −2.3    | −2.0    | −4.6    |
| 24    | −     | −     | −2.3    | −2.3   | −2.2   | −5.3    | −2.3    | −2.0    | −4.6    |
| 26    | −     | −     | −2.4    | −2.4   | −2.2   | −5.4    | −2.4    | −2.0    | −4.6    |
| 28    | −     | −     | −2.5    | −2.5   | −2.2   | −5.5    | −2.5    | −2.0    | −4.6    |
| 30    | −     | −     | −2.5    | −2.5   | −2.2   | −5.5    | −2.5    | −2.0    | −4.6    |
3.1.3. Oxidative Stability and an Acid Value of WCME1 and WCME2

The induction periods (IP) of WCME1 and WCME2 at different storage periods, which were measured according to EN-14112 standard [60], are summarized in Table 6. It was observed that with an increase in the storage period, the IP decreases for all the fuel samples. In general, the oxidative stability of biodiesel decreases when the polyunsaturation was increased [61,62].

Table 6. Influence of storage time (months) and temperature on the induction period and acid value of WCME1 and WCME2.

| Months | IP [h]  | AV [mg KOH/g] |
|--------|--------|---------------|
|        | WCME1  | WCME2        | WCME1  | WCME2 |
| 0      | 8.0    | 12.0         | 0.35   | 0.19  |
| 2      | 7.0    | 10.8         | 0.39   | 0.21  |
| 4      | 6.2    | 9.6          | 0.42   | 0.23  |
| 6      | 5.5    | 8.6          | 0.47   | 0.25  |
| 8      | 5.0    | 7.7          | 0.51   | 0.28  |
| 10     | -      | 6.8          | -      | 0.31  |
| 12     | -      | 6.1          | -      | 0.35  |
| 14     | -      | 5.4          | -      | 0.39  |
| 16     | -      | 4.9          | -      | 0.43  |
| 18     | -      | 4.3          | -      | 0.48  |
| 20     | -      | 3.9          | -      | 0.53  |
| 22     | -      | 3.5          | -      | 0.59  |
| 24     | -      | 3.1          | -      | 0.66  |
| 26     | -      | 2.8          | -      | 0.73  |
| 28     | -      | 2.5          | -      | 0.81  |
| 30     | -      | 2.2          | -      | 0.90  |

A simple ratio (retention factor \( R_f \)) was defined to compare the IP results. \( R_f \) is expressed in Equation (47):

\[
R_f = \frac{IP \ (zero \ - \ month)}{IP \ (overextended \ storage \ period)}
\] (47)

For example, it was found that for eight months of storage, \( R_f \) values were 0.627 and 0.636 for WCME1 and WCME2, respectively.

The initial acid values (AV) of two pure biodiesels (WCME1 and WCME2) were significantly below the maximum allowable limit of 0.50 mg KOH/g specified in ASTM D664 [63] (Table 2), as shown in Table 6. It was found that storage over an extended period (eight months for WCME1 and 30 months for WCME2) resulted in an increase in acid value for all biodiesel samples. According to Bouaid et al. [64], the acid values of biodiesel samples are increased with an increase in storage period for all fuel samples because of the hydrolysis of fatty acid methyl esters to fatty acids.

Generally, the acid value increases with an increase in peroxide formation because the esters are first oxidized to form peroxides, which then undergo complex reactions including a split into more reactive aldehydes, which further oxidize into acids [65]. The oxidation process is followed by a polymerization reaction, whereby the smaller molecules are combined to form larger molecules, and thus has the tendency to increase the densities of the fuel samples when the storage period is increased (Table 4). The higher molecular weight compounds thus formed also increased the kinematic viscosities of the fuel samples (Table 4).
3.2. Analyzing the Properties of Biodiesel Mixture

As mentioned previously, the aim of the current study is to analyze the properties of biodiesel mixtures derived from three waste cooking oil biodiesels, namely, WCME1, WCME2 and WCME3, with different ages and compositions. The experimental results of the fuel samples were compared through figures and tables.

3.2.1. Kinematic Viscosities

To ensure the accuracy of the results, the repeatability test was carried out for each sample. The measurement of flow time was repeated three to four times for each specific temperature; then, the average was determined to calculate the viscosity. Based on the results, the repeatability error was less than 1%, which indicates that the results to be discussed are 99% accurate and precise.

The kinematic viscosities of 12 biodiesel fuels were experimentally measured. It was observed that the kinematic viscosity of the biodiesel samples decreased when the testing temperature increased. Additionally, it was found that the kinematic viscosities of the biodiesel blends varied in the range of 2.15–13.42 mm²/s at the temperature range of 0–80 °C.

To establish the quality of the used fuels, the results of the biodiesel properties including the kinematic viscosity at 40 °C, which was determined by ASTM D455, are shown in Figure 4. The results indicate that all biodiesel samples are within the accepted range, as shown in Table 2. The variation in kinematic viscosity of biodiesel blends at 40 °C (Figure 4) can be explained as follows:

- For WCME1-WCME2; the kinematic viscosities of pure WCME1 and WCME2 were higher than the viscosity of the blends. Additionally, WCME1-WCME2-50% had the highest value of viscosity compared to WCME1-WCME2-75% and WCME1-WCME2-25%.
- For WCME1-WCME3; based on the results, it was found that the increasing percentage amount of WCME2 WCME1-WCME3 blend (from 0% to 50%) increases the kinematic viscosity of biodiesel blends almost linearly, i.e., WCME1-WCME3-50% had the highest maximum kinematic viscosity compared to the other samples.
- For WCME2-WCME3; it was observed that increasing the amount of WCME3 leads to a decrease in the kinematic viscosity of WCME2-WCME3, i.e., WCME2-WCME3-25% had the minimum kinematic viscosity compared to the other samples.

![Kinematic viscosity versus volume fraction at a temperature of 40 °C (WCME1-WCME2: WCME 1 and WCME2 are considered as fuel 1 and fuel 2; WCME1-WCME3: WCME 1 and WCME3 are considered as fuel 1 and fuel 2; WCME2-WCME3: WCME 2 and WCME3 are considered as fuel 1 and fuel 2).](image-url)

Figure 4. Kinematic viscosity versus volume fraction at a temperature of 40 °C (WCME1-WCME2: WCME 1 and WCME2 are considered as fuel 1 and fuel 2; WCME1-WCME3: WCME 1 and WCME3 are considered as fuel 1 and fuel 2; WCME2-WCME3: WCME 2 and WCME3 are considered as fuel 1 and fuel 2).
The kinematic viscosity of fatty compounds is significantly influenced by the compound structure as indicated by the present data obtained at 40 °C (Figure 4). Influencing factors are chain length, position, number and nature of the double bonds, as well as the nature of the oxygenated moieties. Based on the fatty acid methyl ester composition of the biodiesels, the WCME1 contained large amounts of methyl oleate (C18:1) at 41.6 wt.%, methyl palmitate (C16:0) at 24.8 wt.%, and methyl linoleate (C18:2) at 22.3 wt.%. Similarly, the WCME2 principally consisted of methyl oleate (C18:1) at 44.36 wt.% and methyl palmitate (C16:0) at 28.53 wt.%. The primary FAMEs in the WCME3 were methyl oleate (C18:1) at 51.75 wt.% and methyl linoleate (C18:2) at 20.28 wt.%, representing a relatively high fraction of polyunsaturated FAMEs. Because the fuel properties of biodiesels are mainly determined by their major components, particularly alkyl esters [66,67], biodiesels produced from different feedstocks have distinct fuel properties due to their dissimilar FAME profiles. In addition, WCME1 and WCME2 are mostly unsaturated and thus have a low cloud point with respect to WCME3, which contains significantly more saturated esters. By adding WCME2 in the WCME1/WCME2 or WCME2/WCME3 mixture, the net effect progressively increases the saturated nature of the mixture.

3.2.2. Density of Biodiesel

Figure 5 demonstrates the variations of densities of the biodiesel samples measured by the authors with respect to testing temperature. The results indicated that the densities of all biodiesel samples were in the range of 867–900 kg/m³. It was observed that the density of the samples decreased when the temperatures of all tested fuels are increased. Moreover, it was observed that the densities of the WCME2-WCME3 blends were lower than the density of the pure biodiesels (WCME2 and WCME3).
Figure 5. The density of WCME1-WCME2 versus temperature for the different volume fraction of WCME2.

Figure 6 shows the effect of blending ratio on the density of the pure biodiesels at 15 °C. It can be seen that the density of the biodiesel blends increased or decreased according to fuel 2 (WCME2 or WCME3) concentrations in the blend.

3.2.3. Kinematic Viscosity and Density Phenomenon

According to Kassem et al. [30], the behavior of the kinematic viscosity and density of two waste oil biodiesel blends at various volume fractions cannot be explained by the variations in the fatty acid concentration profiles in the mixture according to the standard of mixing rule, which can be considered as a complex phenomenon.

In order to verify that the density and kinematic viscosity of the biodiesel samples have the same phenomenon, the dynamic viscosity has been illustrated for 40 °C and 15 °C, as shown in Figure 7. The reason for choosing these temperatures is that kinematic viscosity and density should be measured at 40 °C and 15 °C, respectively, according to the ASTM standard, to ensure that the biodiesel could be used as diesel fuel for a diesel engine. Generally, the dynamic viscosity value is obtained from the measurements of density and kinematic viscosity. Thus, it was observed that dynamic viscosity had the same characteristics as the kinematic viscosity.
3.2.4. Cold Flow Properties of Biodiesel Samples

Cold flow properties of the fuel samples including CP, CFPP and PP are summarized in Table 7. It can be seen that increasing the percentage of WCME2 in the mixture of WCME1-WCME2 had a beneficial effect on decreasing the CP and increasing the PP of the blends. It was concluded that WCME2 could be used to improve the cold flow properties of WCME1. Additionally, it was observed that the CP of WCME1-WCME3 and WCME2-WCME3 increased with the increasing percentage of WCME3 in the blend. It was also found that increasing the blend ratios of WCME1-WCME3 and WCME2-WCME3 led to a decrease in the PP of the blends.

Table 7. Cold flow properties of all biodiesels.

| Properties | WCME 1 | WCME1-WCME2-75% | WCME1-WCME2-50% | WCME1-WCME2-25% | WCME 2 |
|------------|--------|------------------|------------------|------------------|--------|
| CP (°C)    | 5.2    | 3.4              | 1.6              | −0.2             | −2.0   |
| CFPP (°C)  | 4.6    | 2.9              | 1.3              | −0.4             | −2.3   |
| PP (°C)    | −2.0   | −2.46            | −3.2             | −4.2             | −5.5   |
| Properties | WCME 1 | WCME1-WCME3-75% | WCME1-WCME3-50% | WCME1-WCME3-25% | WCME 3 |
| CP (°C)    | 5.2    | 5.0              | 4.8              | 4.6              | 4.5    |
| CFPP (°C)  | 4.6    | 4.5              | 4.4              | 4.3              | 4.2    |
| PP (°C)    | −2.0   | −0.1             | 1                | 1.3              | 1.4    |
| Properties | WCME 2 | WCME2-WCME3-75% | WCME2-WCME3-50% | WCME2-WCME3-25% | WCME 3 |
| CP (°C)    | −2.0   | 0.2              | 2.0              | 3.4              | 4.5    |
| CFPP (°C)  | −2.3   | −2.1             | 1.6              | 3.1              | 4.2    |
| PP (°C)    | −5.5   | −4.1             | −2.5             | −0.9             | 1.4    |

Figure 7. Viscosities versus volume fraction at; (a) the testing temperature of 40 °C and (b) the testing temperature of 15 °C (Dynamic viscosity: solid line; kinematic viscosity: dashed line).
3.2.5. Computer-Aided Cooling Curve Analysis of Biodiesel Samples

Computer-aided cooling curve thermal analysis presents useful information about the solidification latent heat, fraction of solid during solidification and the amount of different phases. The cooling curve recorded in thermal analysis is a temperature versus time (T versus t) graph of a melt during freezing; hence, it keeps the whole solidification history.

As mentioned before, the cooling bath was cooled down to −20 °C and the fuel sample was heated up to 65 °C. Temperature readings from thermocouples were recorded using a data logger with 30-second intervals and the data was stored for analysis. The cooling curves and their rate of change with respect to time (dT/dt) using second-order approaches (Equation (48)) are plotted for pure biodiesel samples, as shown in Figures 8 and 9.

\[
T'_n = \left( \frac{dT}{dt} \right)_n = \frac{T_{n+1} - T_{n-1}}{t_{n+1} - t_{n-1}}, \quad (t'_n)_n = \frac{t_{n+1} + t_{n-1}}{2}
\]  

(48)

The average cooling bath temperature was fixed at −20.3 °C with a minimum of −20.7 °C and a maximum of −19.8 °C. A plateau is observed on the cooling curve (T versus t) at about 4.4 °C, −2.2 °C and 4.5 °C, which correspond to the experimentally determined CFPP values for WCME1, WCME2 and WCME3, respectively (Table 7). The CP (6.1 °C for WCME1, 1.4 °C for WCME2 and 9.4 °C for WCME3) was located before the plateau, which corresponds to a sharp change in the slope of \( \frac{dT}{dt} \) versus the t curve, as shown in Figure 8. Moreover, the thermal analysis for the blends was analyzed. For instance, Figure 9 shows the cooling curve and \( \frac{dT}{dt} \) for some selected blends including WCME1-WCME2-50%, WCME1-WCME3-50% and WCME2-WCME3-50%. It was found that the CFPP values for WCME1-WCME2-50%, WCME1-WCME3-50% and WCME2-WCME3-50% are 1.4 °C, 4.5 °C and 1.7 °C, respectively, which are close to the experimental values, as shown in Table 7.

![Image](https://via.placeholder.com/150)

**Figure 8.** Cooling curve analysis and rate of change of temperatures with respect to time for all pure biodiesel.
The primary information obtained from the cooling curves is the phase transition temperature. Transition temperatures appear as a kink followed by a considerable change in slope in the second derivative curve. The plotting of the rate change of temperature with respect to time versus \((T - T_0)\) can be helpful to identify the regions of the liquid phase, solid phase and two phases (liquid and solid phases). Changes in the \(\frac{\Delta A}{\Delta C_p} \) value in the liquid and solid states depend on the composition of fatty acid methyl esters and their crystallization [37]. The term \(\frac{\Delta A}{\Delta C_p} \) in Equation (2) remained constant and was derived from the slope of the straight line as 0.000744 s\(^{-1}\), 0.000721 s\(^{-1}\) and 0.000763 s\(^{-1}\) for WCME1, WCME2 and WCME3, respectively (see Figure S1 as supplementary material). It should be noted that the \(\frac{\Delta A}{\Delta C_p} \) term can still be defined as a function of temperature and is inserted into Equation (2).

Moreover, the examination of different phases (liquid phase, solid phase, and two phases) for biodiesel blends is investigated. The \(\frac{\Delta A}{\Delta C_p} \) values are found to be 0.000817 s\(^{-1}\), 0.000721 s\(^{-1}\) and 0.000744 s\(^{-1}\) for WCME1-WCME2-50%, WCME1-WCME3-50% and WCME2-WCME3-50%, respectively (see Figure S2 as supplementary material). Furthermore, the Newtonian zero curve plots \((Z_c)\) for six selected samples along with cooling curves and \(\frac{dT}{dt}\) are also given in Figures 9 and 10.

The area between \((Z_c)\) and the \(\frac{dT}{dt}\) curves from the start to the end of freezing of the biodiesel is directly related to the total latent heat for solidification [37]. The ratio of the incremental cumulative area, \(A_n\), to the total area, \(A_{Total}\), gives the incremental solid fraction \(f_s\) of the sample during solidification [37]. The Trapezoidal Rule was used for the area calculations. Equations (49) and (50) were used to estimate the solid fraction using Newtonian thermal analysis for any data point \(n\) [36]. \(A_{Total}\) can also be calculated from Equation (49) by substituting \(n = n_{Total} - 1\), where \(n_{Total}\) is the total number of data points. The new corresponding temperature \((T'_{f_s})_n\) was calculated by averaging the two successive temperatures, as given in Equation (51) [37]:

\[
(f_s)_n = \frac{A_n}{A_{Total}}
\]  

\[
A_n = \sum_{i=1}^{n}\left\{\frac{1}{2}(T'_{i+1} + T'_i) - \frac{1}{2}(Z_{Ni+1} + Z_{Ni})\right\} \times (t_{i+1} - t_i)
\]
\[
(T_k)_n = \frac{T_n + T_{n+1}}{2}
\]  

From the obtained cooling curve, the first derivative graph was calculated. The zero-curve of the graph was determined after the first derivative of the graph. The baseline of the graph was obtained from the differential temperature of liquidus and solidus from the first derivative graph using a linear equation. The baseline can be defined as a hypothetical path that the first derivative curve would follow if there were no latent heat releases. A baseline may also be isothermal in experiments where temperature is held constant. The \( Z_c \) was estimated with Equation (2) using all the data points and is incorporated into Figure S3 as supplementary material. The \( Z_c \) and \( \frac{dT}{dt} \) curves overlap before and after solidification but deviate from each other during solidification, i.e., in the two phase regions, since \( Z_c \) does not include latent heat during freezing. The solid fraction was calculated based on the first derivative and the baseline graph (zero curves). The change in solid fraction during solidification of the biodiesel sample was determined using Equations (49)–(51). Integration of the area between the cooling curve rate and baseline graph gives relevant information on the solidification (see Figure S3 as supplementary material). The changes in solid fraction during solidification of some selected samples are plotted in Figure 10.

![Solid fraction vs Temperature](image)

**Figure 10.** Variation of solid fraction during freezing of some selected biodiesel samples.

### 3.3. Biodiesel Correlations

As previously mentioned, the most widely used expressions in the literature for predicting the kinematic viscosity and density of biodiesel blends were examined to establish whether they can predict the blended viscosity and density of mixtures of biodiesel. Moreover, 10 correlations were proposed by the authors to estimate the properties of the fuel samples. The accuracy of the developed correlations was also tested by predicting the viscosity and density of the biodiesel-diesel blends.

#### 3.3.1. Kinematic Viscosity Correlations

The correlative ability of Equations (5)–(31) for 12 different binary mixtures that show a nonlinear behavior of the logarithm of the kinematic viscosity as a function of the volume fraction and temperature was investigated.

In this section, the viscosity correlations are divided into four different types of correlations; I—viscosity-temperature correlations (Table 8), II—viscosity-volume fraction correlations, III—viscosity-volume fraction-temperature correlations (Table 9) and IV—viscosity correlations depending on the temperature, volume fraction of biodiesels and properties of the pure biodiesel.
Table 8. Kinematic viscosity-temperature correlations for different biodiesel blends; correlation parameters and $R^2$.

| Equation Number | Blends | T [K] | Correlation Parameters | R²  |
|-----------------|--------|-------|------------------------|-----|
| 5               | WCME 1 | 279–353 | 2.117 $-453.038$ $7.04 \times 10^5$ | 0.997 |
|                 | WCME 2 | 273–353 | 3.493 $-3273.49$ $8.28 \times 10^5$ | 0.999 |
|                 | WCME 3 | 279–353 | 0.862 $-1634.08$ $5.71 \times 10^5$ | 1.000 |
|                 | WCME1-WCME3-75% | 279–353 | 0.168 $-1174.72$ $4.96 \times 10^5$ | 1.000 |
| 6               | WCME 1 | 279–353 | $-1.629$ $466.388$ $-162.536$ | 0.997 |
|                 | WCME 2 | 273–353 | $-1.483$ $434.452$ $-166.933$ | 0.999 |
|                 | WCME 3 | 279–353 | $-1.931$ $567.199$ $-146.216$ | 1.000 |
|                 | WCME1-WCME3-50% | 279–353 | $1.000$ $495.710$ $-151.675$ | 0.994 |
| 18              | WCME1-WCME2-50% | 275–353 | $13.567$ $9.017$ $1.56 \times 10^{12}$ $-4.637$ | 0.896 |
|                 | WCME1-WCME3-50% | 279–353 | $14.096$ $14.096$ $7.38 \times 10^{12}$ $-4.472$ | 0.912 |
|                 | WCME2-WCME3-75% | 273–353 | $9.061$ $8.07$ $0.000$ $8.070$ | 0.128 |
| 20              | WCME 1 | 279–353 | $2.00 \times 10^{12}$ $-4.637$ | 0.994 |
|                 | WCME 3 | 279–353 | $1.56 \times 10^{12}$ $-4.597$ | 0.906 |
|                 | WCME1-WCME3-25% | 279–353 | $14.096$ $14.096$ $7.38 \times 10^{12}$ $-4.472$ | 0.912 |
| 21              | WCME 1 | 279–353 | $9.017$ $8.781$ $-0.023$ | 0.987 |
|                 | WCME 3 | 279–353 | $8.781$ $8.781$ $-0.023$ | 0.991 |
|                 | WCME1-WCME3-75% | 279–353 | $8.671$ $8.671$ $-0.023$ | 0.992 |

Table 9. Kinematic viscosity-volume fraction-temperature correlations for different biodiesel blends; correlation parameters and $R^2$.

| Equation Number | Blends | T [K] | Correlation Parameters | R²  |
|-----------------|--------|-------|------------------------|-----|
| 12              | WCME1-WCME2 | 273–353 | $-5.961$ $0.319$ $2332.425$ $-103.074$ | 0.995 |
|                 | WCME1-WCME3 | 279–353 | $-5.119$ $-0.143$ $2093.396$ $47.694$ | 0.994 |
|                 | WCME2-WCME3 | 273–353 | $-5.814$ $-0.127$ $2282.583$ $14,625.786$ | 0.992 |
| 23              | WCME1-WCME2 | 273–353 | $15.567$ $-3.514$ $0.000$ $-3.514$ | 0.286 |
|                 | WCME1-WCME3 | 279–353 | $14.096$ $-4.056$ $-0.012$ $-4.056$ | 0.212 |
|                 | WCME2-WCME3 | 273–353 | $-9.061$ $8.07$ $0.000$ $8.070$ | 0.128 |
| 28              | WCME1-WCME2 | 273–353 | $-6.019$ $0.484$ $2348.965$ $0.000$ | 0.995 |
|                 | WCME1-WCME3 | 279–353 | $-5.295$ $0.000$ $2120.892$ $1.425$ | 0.994 |
|                 | WCME2-WCME3 | 273–353 | $-5.975$ $0.110$ $2328.612$ $0.000$ | 0.991 |
| 29              | WCME1-WCME2 | 273–353 | $-5.930$ $0.111$ $2323.783$ $-12,510.043$ | 0.995 |
|                 | WCME1-WCME3 | 279–353 | $-5.173$ $-0.093$ $2085.63$ $9545.73$ | 0.994 |
|                 | WCME2-WCME3 | 273–353 | $-5.814$ $-0.127$ $2282.583$ $14,625.786$ | 0.992 |
| 30              | WCME1-WCME2 | 273–353 | $-5.820$ $2292.567$ $-12.322$ | 0.995 |
|                 | WCME1-WCME3 | 279–353 | $-5.272$ $2114.226$ $6.337$ | 0.994 |
|                 | WCME2-WCME3 | 273–353 | $-5.978$ $2328.337$ $15.611$ | 0.992 |
| 31              | WCME1-WCME2 | 273–353 | $-5.806$ $2297.325$ $-3521.23$ | 0.995 |
|                 | WCME1-WCME3 | 279–353 | $-5.261$ $2111.2$ $1860.93$ | 0.994 |
|                 | WCME2-WCME3 | 273–353 | $-5.948$ $2320.881$ $4466.652$ | 0.992 |
| 32              | WCME1-WCME2 | 273–353 | $-8.426$ $2980.013$ $-0.136$ | 0.998 |
|                 | WCME1-WCME3 | 279–353 | $-7.16$ $2619.55$ $-0.497$ | 0.996 |
|                 | WCME2-WCME3 | 273–353 | $-8.682$ $3059.044$ $-0.031$ | 0.996 |

The correlation parameters and R-squared values for estimating the kinematic viscosity of some selected biodiesel blends as a function of temperature are shown in Table 8. For a perfect fit, for example, $R^2$ becomes 1, which means that the equation explains 100% of the variability of the measured data. As shown in Table 8, the R-squared values ranged from 0.896 to 1.000 for all blends. These values...
indicate that the correlations properly fit the experimental data and represent the viscosity-temperature relationship almost exactly.

Moreover, among the correlations used for predicting the viscosity of biodiesel blends as a function of temperature and volume fraction (Table 9), Equation (31), developed by Kassem and Çamur [30], showed an accurate fit for WCME1-WCME2, WCME1-WCME3 and WCME2-WCME3 blends. Nevertheless, Equation (23) shows that the fit is not good enough for biodiesel blends because the smaller R-squared value (see Table 9) indicates that there is a large deviation between the experimental and predicted data for the biodiesel blends.

Furthermore, in order to improve the accuracy of the empirical equations (Equations (7)–(10), Equation (13), Equations (15)–(17) and Equations (24)–(27)) mentioned above, the measured and calculated values of kinematic viscosity were compared and R-squared was calculated. The results indicate that the R-squared values varied between 0.162 and 0.312 (the authors did not tabulate the results in the paper to save space), which are generally considered to be too weak to describe the actual kinematic viscosity of the blends. Therefore, it can be concluded that it is not possible to use these equations as predictor equations to estimate the viscosity of biodiesel blends.

### 3.3.2. Density Correlations

The equations from Section 2.6 were tested to predict the densities of the mixtures of biodiesel and R-squared were calculated. Table 10 presents the correlation parameters of eight equations (Equation (32), Equations (34)–(40)) and R-squared values for all biodiesel blends. It was found that the R-squared values of WCME1-WCME2 and WCME1-WCME3 blends were in the range of 0.659 and 0.731, which shows that the obtained values were not satisfactory and estimated values. Moreover, as seen in Table 10, the R-squared values of the WCME2-WCME3 blends range between 0.683–0.940 and 0.899–0.940, respectively, which indicates that there is an excellent agreement between the measured and predicted data for the biodiesel blends.

#### Table 10. Fraction-temperature correlations for biodiesel blends; correlation parameters and $R^2$.

| Blend          | Equation Number | Property          | T [K]            | Correlation Parameters | $R^2$ |
|---------------|-----------------|-------------------|------------------|------------------------|-------|
| WCME1-WCME2   | 32              | $\rho(T)$         | 273–353          | A: 1209.997, B: -0.973, C: -0.966, D: 0.0069, E: 1.210499, R^2: 0.883 |       |
|               | 34              | $\rho(VF, T)$     |                  | A: -5.579, B: -0.966, C: 1.210499, R^2: 0.889 |       |
|               | 35              | $\rho(T)$         |                  | A: 2358.365, B: -8.396, C: 0.012, R^2: 0.935 |       |
|               | 36              | $\rho(T)$         |                  | A: 4315.195, B: -0.009, C: 344.642, R^2: 0.940 |       |
|               | 37              | $\rho(T)$         |                  | A: -362.047, B: 0.227, C: 2424.497, R^2: 0.894 |       |
|               | 38              | $\rho(VF, T)$     |                  | A: 2323.184, B: -8.120, C: -33.019, R^2: 0.938 |       |
|               | 39              | $\rho(VF, T)$     |                  | A: 1267.567, B: -0.001, C: -5.453, R^2: 0.893 |       |
|               | 40              | $\rho(VF, T)$     |                  | A: 1235.044, B: -1.048, C: -60.257, R^2: 0.892 |       |
| WCME1-WCME3   | 32              | $\rho(T)$         | 279–333          | A: 1182.479, B: -0.915, C: 1178.238, R^2: 0.899 |       |
|               | 34              | $\rho(VF, T)$     |                  | A: 8.482, B: -0.915, C: 1178.238, R^2: 0.915 |       |
|               | 35              | $\rho(T)$         |                  | A: 1963.578, B: -5.928, C: 0.008, R^2: 0.925 |       |
|               | 36              | $\rho(T)$         |                  | A: 946.808, B: 0.000, C: 1663.791, R^2: 0.933 |       |
|               | 37              | $\rho(T)$         |                  | A: -310.273, B: 0.237, C: 2106.391, R^2: 0.906 |       |
|               | 38              | $\rho(VF, T)$     |                  | A: 1970.518, B: -5.965, C: -13.881, R^2: 0.940 |       |
|               | 39              | $\rho(VF, T)$     |                  | A: 1230.664, B: -0.001, C: 8.486, R^2: 0.917 |       |
|               | 40              | $\rho(VF, T)$     |                  | A: 1189.419, B: -0.951, C: -13.881, R^2: 0.915 |       |
| WCME2-WCME3   | 32              | $\rho(T)$         | 273–353          | A: 1200.361, B: -1.019, C: 1184.861, R^2: 0.659 |       |
|               | 34              | $\rho(VF, T)$     |                  | A: 17.024, B: -0.998, C: 1184.861, R^2: 0.693 |       |
|               | 35              | $\rho(T)$         |                  | A: 2543.716, B: -9.700, C: 0.014, R^2: 0.707 |       |
|               | 36              | $\rho(T)$         |                  | A: 3385.867, B: -0.007, C: 123.966, R^2: 0.708 |       |
|               | 37              | $\rho(T)$         |                  | A: 397.291, B: 0.223, C: 2310.942, R^2: 0.668 |       |
|               | 38              | $\rho(VF, T)$     |                  | A: 2450.237, B: -12.219, C: 3.081, R^2: 0.731 |       |
|               | 39              | $\rho(VF, T)$     |                  | A: 1249.488, B: -0.001, C: 16.912, R^2: 0.697 |       |
|               | 40              | $\rho(VF, T)$     |                  | A: 1194.360, B: -1.762, C: 0.734, R^2: 0.690 |       |
3.3.3. Kinematic Viscosity—Density Correlations

Each of the three equations (Equations (41), (43) and (44)) in Table 11 predicting the viscosity or density of the biodiesel blends were tested and compared to the experimental values as measured in the laboratory. The statistical values $R^2$ and correlation parameters were calculated for each of the blends and are presented in Table 11. As a result, Equation (41) developed by Kassem and Çamur [30] showed an accurate fit for the WCME1-WCME2, WCME1-WCME3 and WCME2-WCME3 blends. For example, for WCME1-WCME2, the $R$-squared was 0.981.

| Blend         | Equation Number | Property | T [K]   | Correlation Parameters | $R^2$ |
|---------------|-----------------|----------|---------|------------------------|-------|
|               |                 |          |         | A          | B       | C       | D       |       |
| WCME1-WCME2   | 41              | $v(\rho)$| 273–353 | -5642.06   | 548.362 | -0.062  | 89.559  | 0.981  |
|               | 43              | $\rho(v)$| 273–353 | 6.508      | 1.053   | 864.312 | -       | 0.969  |
|               | 44              | $\rho(v)$| 273–353 | 998.409    | -195.979| -        | -       | 0.892  |
| WCME1-WCME3   | 41              | $v(\rho)$| 279–353 | -4232.01   | 538.81  | -0.043  | 63.521  | 0.958  |
|               | 43              | $\rho(v)$| 279–353 | 22.646     | 0.66    | 829.195 | -       | 0.942  |
|               | 44              | $\rho(v)$| 279–353 | 986.448    | -189.335| -        | -       | 0.903  |
| WCME2-WCME3   | 41              | $v(\rho)$| 273–353 | -8805.52   | 362.959 | -0.047  | 77.419  | 0.730  |
|               | 43              | $\rho(v)$| 273–353 | 10.036     | 0.908   | 832.744 | -       | 0.719  |
|               | 44              | $\rho(v)$| 273–353 | 977.206    | -202.188| -        | -       | 0.669  |

3.3.4. Empirical Modeling of Kinematic Viscosity and Density Developed by Authors

In this section, 10 empirical equations have been investigated by the authors to predict the kinematic viscosities and densities of biodiesel blends in a wide temperature range, as shown in Table 12. These equations depend on the temperature, volume fraction of pure components (biodiesels), and properties of pure components (biodiesels). The correlation parameters and $R$-squared values for each of the developed equations are listed in Table S1 as supplementary materials. The $R$-squared is a quantitative measure of goodness of fit of the correlation to the measured data. The minimum $R$-squared was obtained from Equation (58) for WCME2-WCME3 as 0.836. However, the $R$-squared values varied between 0.971 and 0.999 for all fuel samples, which shows that the developed correlations accurately fit the measured data.

To prove the authenticity of the developed correlations, Equations (52)–(61) were tested to estimate the kinematic viscosity and density values of other fuel samples including biodiesel-diesel blends given in the literature [52,66–69]. The correlation parameters and $R$-squared values for all developed correlations are presented in Table S2 as supplementary materials. Ramírez-Verduzco et al. [52] proposed four correlations to estimate the density and kinematic viscosities of biodiesel blends with ultra-low sulfur diesel. They found that the $R$-squared values were 0.999 for all correlations. Based on the $R$-squared values in Table S2 as supplementary materials, it is found that Equations (52) and (53) show the best fit for variations of kinematic viscosities with volume fraction and testing temperature simultaneously. In addition, Equations (59)–(61) show the best fit for variations of densities with volume fraction and testing temperature simultaneously. It can be concluded that the new Equations (52), (53), (59)–(61) correlate the kinematic viscosity and densities better than the equations proposed by Ramírez-Verduzco et al. [52]. Moreover, it was observed that the minimum $R$-squared values obtained from Equations (57)–(59) are not satisfactory, while the other equations appear to be the best fits for estimating the density of biodiesel-diesel blends [66].
Table 12. Mathematical equations developed by the authors.

| Equation Number | Property                                      | Equations                                                                                                                                 |
|-----------------|-----------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------|
| 52              | $\nu(T, VF)$                                   | $\nu = \exp\left(A + B T + C VF_1 + D VF_2 + E T^2 + F VF_1^2 + G VF_2^2 + H T VF_1 + L T VF_2 + K VF_1 VF_2\right)$   |
| 53              | $\nu(T, VF, \rho)$                             | $\nu = \exp\left(A + B T + C VF_1 + D VF_2 + E \rho + F T^2 + G VF_1^2 + H VF_2^2 + L T VF_1 + K T VF_2 + M T \rho + N VF_1 VF_2 + O VF_1 \rho + P VF_2 \rho\right)$ |
| 54              | $\nu(T, VF, \rho)$                             | $\nu = A + B T + C VF_1 + D \rho + E T^2 + F VF_1^2$                                                                                     |
| 55              | $\nu(T, VF, v_1, v_2)$                         | $\nu = \exp\left(A + B T + C VF_1 + D VF_2 + E v_1 + F v_2 + G VF_1^2 + H VF_2^2 + L v_1^2 + K v_2^2\right)$                          |
| 56              | $\nu(VF, v_1, v_2)$                            | $\nu = A + B VF_1 + C v_1 + D v_2 + E VF_1^2$                                                                                           |
| 57              | $\nu(T, VF, v_1, v_2)$                         | $\nu = \exp(A + B T + C VF_1 + D VF_2 + E v_1 + F v_2)$                                                                                |
| 58              | $\nu(VF, v_1, v_2, \rho_1, \rho_2)$           | $\nu = A + B VF_1 + C VF_2 + D v_1 + E v_2 + F \rho_1 + G \rho_2$                                                                      |
| 59              | $\rho(T, VF)$                                  | $\rho = \exp\left(A + B T + C VF_1 + D VF_2 + E T^2 + F VF_1^2 + G VF_2^2 + H T VF_1 + L T VF_2 + K VF_1 VF_2\right)$             |
| 60              | $\rho(T, VF, \rho_1, \rho_2)$                 | $\rho = \exp(A + B T + C VF_1 + D VF_2 + E \rho_1 + F \rho_2)$                                                                        |
| 61              | $\rho(VF, \rho_1, \rho_2)$                    | $\rho = A + B VF_1 + C VF_2 + D \rho_1 + E VF_1^2 + F VF_2^2 + G VF_1 \rho_1$                                                           |

$\nu$ Kinematic viscosity of blend in mm$^2$/s  
$v_1$ Kinematic viscosity of pure component 1 in mm$^2$/s  
$v_2$ Kinematic viscosity of pure component 2 in mm$^2$/s  
$T$ Test temperature in K  
$VF_1$ Volume fractions of pure component 1  
$VF_2$ Volume fractions of pure component 2  
$\rho$ Density of blend in kg/m$^3$  
$\rho_1$ Density of pure component 1 in kg/m$^3$  
$\rho_2$ Density of pure component 2 in kg/m$^3$  
$A, B, C, \ldots, P$ Constant
4. Conclusions

The usage of waste cooking vegetable oils (WCVOs) as an alternative fuel in diesel engines has drawn significant attention. The WCVOs are commonly used to make biodiesel fuels composed completely from these oils or as blends with petroleum diesel. In the present study, three different biodiesels were produced from various waste cooking oils using the transesterification method. In addition, three different mixtures of waste cooking biodiesels (WCME1, WCME2 and WCME3) with significantly different compositions, various blending ratios, and ages were prepared. In this work, WCME1 was obtained from a mixture of waste frying and canola cooking oil biodiesel, WCME2 was prepared from mixture of waste frying, canola and corn cooking oil biodiesel and WCME3 was found from the mixture of waste canola and corn cooking oil biodiesel. The objective of this study was to investigate the effects of storage period on the properties of the WCME1 and WCME2 including kinematic viscosity, density, CP, PP, acid value and oxidation stability during the storage periods. The results demonstrated that storage for eight and 30 months resulted in higher kinematic viscosity, density and acid values and a lower induction period for WCME1 and WCME2, respectively. In addition, it was found that the kinematic viscosity of WCME2 with a value of 4.534 mm$^2$/s remained within the ranges listed in ASTM D455 (1.9–6.0 mm$^2$/s) and EN–14214 (3.5–5.0 mm$^2$/s) at 30 months. Moreover, this paper investigated the influence of blending WCME1, WCME2 and WCME3 at various volume ratios of 25%, 50% and 75% and temperatures on the kinematic viscosity, density and cold flow properties. In addition, the results demonstrated that WCME2 could be used to improve the cold flow properties of WCME1 and WCME3. Furthermore, the Computer–Aided Cooling Curve Analysis (CACCA) technique was assessed to improve the crystallization/melting profiles of pure methyl biodiesel as well as of their blends. Furthermore, 10 proposed correlations were used to predict the experimental data of the current study and studies in the literature. The results indicated that the developed equations can be used as universal formulas to predict the kinematic viscosity and density for different mixtures of two biodiesels.

Supplementary Materials: The following are available online at http://www.mdpi.com/1996-1073/12/19/3729/s1, Figure S1: $dT/dt$ versus $(T−T_0)$ curve of pure biodiesel blends, Figure S2: $dT/dt$ versus $(T−T_0)$ curve of some selected biodiesel blends, Figure S3: Cooling curve analysis and Newtonian zero curve for some selected samples, Table S1: Correlation parameters and $R^2$ values for Equations (53)–(62), Table S2: The accuracy of the new equations developed by the authors.

Author Contributions: R.H.S.S. measured the properties of biodiesel. Y.K. and H.Ç. analyzed the experimental data and wrote the paper.

Funding: This research received no external funding.

Acknowledgments: The authors would like to thank the Faculty of Engineering especially the Mechanical Engineering Department at Near East University for their support and encouragement.

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

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