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Physicochemical Properties Enhancement of Biodiesel Synthesis from Various Feedstocks of Waste/Residential Vegetable Oils and Palm Oil

Hüseyin Çamur * and Ebaa Alassi

Department of Mechanical Engineering, Faculty of Engineering, Near East University, via Mersin 10, 99138 Nicosia, Turkey; ebaa.alassi@gmail.com

* Correspondence: huseyin.camur@neu.edu.tr; Tel.: +90-(392)-2236464

Abstract: The main aim of the present study was to improve the oxidation stability and cold flow properties of biodiesel produced from waste frying/cooking oil and palm oil. In this work, waste frying/cooking methyl ester (WFME) and palm methyl ester (PME) were prepared using an alkali-catalyzed transesterification process, and the physicochemical properties of the pure biodiesel as well as of binary blends among them were investigated. The results indicated that palm biodiesel and WFME18, produced from a mixture of frying, cooking, sunflower, and corn oils, can be used as antioxidant additives, enhancing biodiesel stability. Additionally, it was found that WFME1 and WFME12 derived from waste residential canola oil can be used as cold flow improvers for enhancing the cold flow properties of palm biodiesel. Moreover, ultra-low sulfur diesel fuel winter (ULSDFW), ultra-low sulfur diesel fuel summer (ULSDFS), kerosene (KF), and benzene (BF) were utilized to enhance the cold flow properties of the samples and meet the requirements of diesel fuel standards. The investigation of the experimental results indicated that blending WFME-PM with a low proportion of petroleum-based fuel (KF and BF) could significantly improve the cold flow properties (CP and PP) as well as oxidation stability of WFME.

Keywords: cold flow properties; oxidation stability; waste frying biodiesel; palm biodiesel; petroleum-based fuels

Citation: Çamur, H.; Alassi, E. Physicochemical Properties Enhancement of Biodiesel Synthesis from Various Feedstocks of Waste/Residential Vegetable Oils and Palm Oil. *Energies* **2021**, *14*, 4928. https://doi.org/10.3390/en14164928

Academic Editor: Byong-Hun Jeon

Received: 30 June 2021
Accepted: 3 August 2021
Published: 11 August 2021

1. Introduction

Biodiesel is a mixture of monoalkyl esters of long-chain fatty acids obtained from vegetable oil that can or cannot be eaten, waste frying oils, and animal fats. It is produced by a transesterification reaction with alcohol, generally methanol or ethanol, in the presence of a catalyst that could either be heterogeneous, homogeneous, or an enzyme. Recently, for instance, Pimentel and Patzek [1] investigated the impact of ethanol and methanol on Karanja biodiesel yield. The results indicated that the obtained biodiesel yield was higher, and less time is required to complete the reaction for methanol compared to ethanol. Hassan and Smith [2] investigated the non-catalyzed supercritical methanol (SCM) process for continuous biodiesel production. Hassan et al. [3] produced biodiesel by two-step sub/supercritical water and ethanol processes for non-catalytic biodiesel production. Moreover, numerous methods have been implemented to reduce the time of reaction and increase biodiesel yield. For instance, Hassan et al. [4] studied the continuous biodiesel production process under sub- and supercritical conditions using a trace amount of potassium hydroxide (KOH) as a catalyst, and CO_2 was added as a co-solvent to reduce the reaction time and increase biodiesel yield. Biodiesel has very little sulphur and is non-toxic, and is thus recognized as an environmentally friendly product.

In general, the quality of biodiesel is associated with several physicochemical properties such as oxidation stability, viscosity, density, and cold flow. Viscosity and density are essential properties of biodiesel [5–7]. In general, biodiesel has higher viscosity and

density compared to fossil diesel, which can influence the performance of the engine and the emission characteristics, mainly at low temperature [8–11]. These properties depend on the fatty acid profile and, in consequence, on the raw materials used for biodiesel production [12]. Another important property of biodiesel is oxidation stability. It affects the stability of biodiesel during an extended storage period [13,14]. The stability of biodiesel is dependent on the number of allylic and bis-allylic sites in the unsaturated fatty acid ester chains [15,16]. Moreover, cold flow properties (CFP) are an essential performance indicator for fossil diesel fuels. Poor CFP is the main problem associated with the direct use of biodiesel in diesel engines during cold weather [17,18]. In general, the cold flow properties of diesel fuel are determined by measuring cloud point (CP) and pour point (PP). Therefore, they are utilized to define the CFP of the fuel [19,20]

As an ongoing study by the authors concerning the development of the stability and cold flow properties of biodiesel by using cold flow improvers [21,22], this study had the goal of putting forward new experimental data to evaluate the quality performance of petroleum-based fuels/ biodiesel blends containing biodiesel produced from waste frying oils and palm oil. Based on scientific studies related to palm biodiesel, the authors noticed that palm biodiesel has high oxidation stability and poor cold flow properties compared to other fuels. Additionally, various authors have mixed petroleum-based fuels such as diesel and kerosene with palm biodiesel to enhance the cold flow properties of the fuel. Based on these considerations, the current study was focused on achieving better low-temperature properties with improved oxidation stability for blends of waste frying biodiesel, palm biodiesel, and petroleum-based fuels for the first time. In this study, 39 methyl biodiesel samples were produced from residential oil and waste frying oil collected from hotels, cafes, and restaurants. In addition, palm biodiesel was purchased from the local market. Furthermore, to use biodiesel in diesel engines according to ASTM D975 and European standard EN 590 for diesel fuel, the biodiesel samples were blended with petroleum-based fuels (ULSDFW, ULSDFS, KF, and BF) in different proportions. The physical and chemical properties of the samples were also assessed to reduce their kinematic viscosity and density and improve their cold-flow properties.

2. Materials and Methods

In this section, the preparation of biodiesel-petroleum-based fuel blends and measurements of their properties according to ASTM standards are explained. Figure 1 schematically illustrates the description of the proposed study.

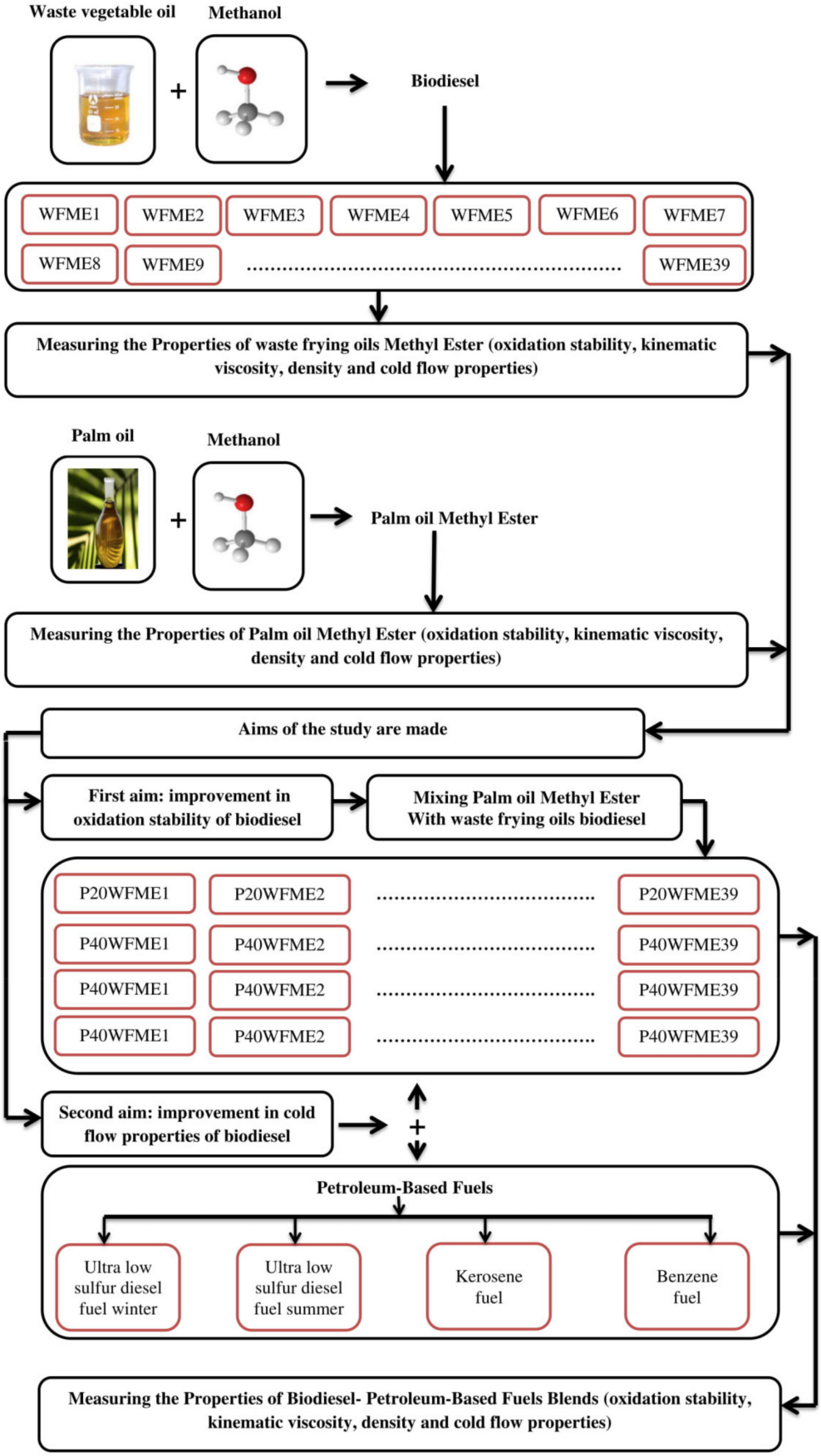

Figure 1. Schematic description of the proposed study.

2.1. Pure Biodiesel Samples

In this work, waste vegetable oils were collected after having been used for cooking purposes several times. The waste frying oils were collected from households, traditional restaurants, hotels, and cafés. These places mainly cooked dishes including animal fats (fish, meat, chicken, and meat/chicken pies) and vegetables (chips, eggplant, zucchini, cauliflower, and vegetable pies). The vegetable cooking oils most widely used in these establishments are corn oil, sunflower, canola, and rapeseed oils. Consequently, 39 biodiesel samples with significantly different compositions were obtained from different sources of waste frying/residential vegetable oils. Furthermore, biodiesel was produced from palm oil, which was purchased from the local market. Biodiesel was produced by transesterification of waste frying oil, refining with methanol (methanol/oil molar ratio 3/1) using NaOH as catalyst (1% w/w based on oil). The description of this reaction is discussed in Refs. [21,22]. The final product was pure biodiesel and was ready to use as fuel, waste frying methyl ester (WFMEY, where Y means the number of biodiesel sample), and refined palm methyl ester (PME). In the present study, each set of samples was divided into 6 samples of 900 mL each, which were placed in a 1000 ml glass bottle; the lid of each sample container was vented with a glass tube to allow for evaporation. Furthermore, the biodiesel samples were analyzed by gas chromatography (GC) to evaluate their fatty acid profiles (see Table S1 in Supplementary Materials). This analysis has been discussed in Ref. [21]. Table 1 shows the estimated monounsaturated (MUFAMEs), polyunsaturated (PUFAMEs), saturated (SFAMEs), degree of unsaturation (DU), and long-chain saturated factors (LCSF) for all biodiesel samples. The required formulas used to estimate MUFAMEs, PUFAMEs, SFAME, DU, and LCSF are expressed by Equations (1)–(5).

$$\sum MUFAMs = \sum wt\%Cxx : 1 \quad (1)$$

$$\sum PUFAMs = \sum wt\%Cxx : 2 + \sum wt\%Cxx : 3 \quad (2)$$

$$\sum SFAMs = \sum wt\%Cxx : 00 \quad (3)$$

$$DU = [monounsatuarted\ Cn : 1] + 2[polyunsaturated\ Cn : 2,\ 3] \quad (4)$$

$$LCSF = 0.1 \times [C16 : 0] + 0.5 \times [C18 : 0] + [C20 : 0] + 1.5 \times [C22 : 0] + 2 \times [C24 : 00] \quad (5)$$

Table 1. Estimated values for MUFAMEs, PUFAMEs, SFAME, DU, and LCSF.

Variable	WFME1	WFME2	WFME3	WFME4	WFME5	WFME6	WFME7
MUFAMEs	64.01	62.83	61.65	60.47	58.11	55.75	52.22
PUFAMEs	6.99	6.65	6.31	5.97	5.29	4.61	3.59
SFAMEs	7.74	9.50	11.27	13.03	16.55	20.08	25.38
DU	120.67	118.46	116.25	114.04	109.62	105.20	98.58
LCSF	1.81	2.01	2.20	2.40	2.79	3.19	3.78
Variable	**WFME8**	**WFME9**	**WFME10**	**WFME11**	**WFME12**	**WFME13**	**WFME14**
MUFAMEs	48.68	46.32	42.78	40.42	65.43	60.43	55.43
PUFAMEs	2.56	1.88	0.86	0.18	6.99	5.63	4.27
SFAMEs	30.65	34.20	39.47	42.99	7.74	14.79	21.84
DU	91.93	87.52	80.88	76.46	120.09	110.96	101.84
LCSF	4.37	4.77	5.35	5.75	1.81	2.60	3.38
Variable	**WFME15**	**WFME16**	**WFME17**	**WFME18**	**WFME19**	**WFME20**	**WFME21**
MUFAMEs	50.42	45.42	40.42	52.07	51.00	49.93	48.85
PUFAMEs	2.90	1.54	0.18	5.20	4.96	4.72	4.47
SFAMEs	28.89	35.94	42.99	22.13	22.70	23.28	23.85
DU	92.71	83.59	74.46	103.03	103.43	103.83	104.23
LCSF	4.17	4.96	5.75	4.13	4.20	4.28	4.35
Variable	**WFME22**	**WFME23**	**WFME24**	**WFME25**	**WFME26**	**WFME27**	**WFME28**
MUFAMEs	47.78	46.71	50.45	54.20	57.94	61.69	54.74
PUFAMEs	4.23	3.99	4.59	5.19	5.79	6.39	5.56
SFAMEs	24.43	25.00	21.55	18.10	14.64	11.19	19.25
DU	104.63	105.03	108.04	111.05	114.07	117.08	106.44
LCSF	4.43	4.50	3.96	3.42	2.89	2.35	3.66
Variable	**WFME29**	**WFME30**	**WFME31**	**WFME32**	**WFME33**	**WFME34**	**WFME35**
MUFAMEs	57.41	60.09	62.76	41.68	42.94	44.19	45.45
PUFAMEs	5.92	6.27	6.63	0.94	1.70	2.47	3.23
SFAMEs	16.37	13.50	10.62	39.39	35.79	32.20	28.60
DU	109.85	113.27	116.68	80.57	86.69	92.80	98.92
LCSF	3.20	2.74	2.27	5.50	5.25	5.00	4.75
Variable	**WFME36**	**WFME37**	**WFME38**	**WFME39**	**PME**		
MUFAMEs	49.74	47.41	45.08	42.75	43.50		
PUFAMEs	4.20	3.19	2.19	1.18	0.20		
SFAMEs	26.30	30.47	34.65	38.82	44.90		
DU	97.32	91.60	85.89	80.17	65.90		
LCSF	4.45	4.78	5.10	5.42	6.36		

Based on the results listed in Table 1, it was found that WFME11 had the highest saturated fatty acid content followed by WFME17. Additionally, it was seen that WFME12 and WFME17 had the highest and lowest MUFAME and PUFAME biodiesel compositions, respectively. In the literature, there are similar previous scientific studies that have been carried out to determine the fatty acid content of waste frying vegetable oil-based biodiesel and refined palm oil-based biodiesel. Evcil et al. [23] investigated the fatty acid distributions of waste frying oil, which was collected from the Engineering Faculty Cafeteria at Near East University. The fatty acid structures of the biodiesel reported in their study were very close to the values found in the current study. El-Araby et al. [24] investigated fatty acid compositions of palm oil biodiesel. Their results were very close to the value found for the PME in the current study. Dunn [25] investigated the fatty acid distributions of some vegetable refined waste oils such as soybean oil and used cooking oil. The fatty acid distributions of used cooking oil were similar to those found in this study.

2.2. Fuels

Four different petroleum-based fuels (ULSDFW, ULSDFS, KF, and BF) were purchased from a local gasoline station. The properties of the selected fuels were measured according to the European Committee for Standardization (EN) specifications in Europe and ASTM specifications in the USA, as shown in Table S2 in the Supplementary Materials. Table 2 lists the properties of these selected fuels.

Table 2. Selected physicochemical properties of tested fuels.

Property	Unit	Test Method	ULSDFW	ULSDFS	KF	BF
Density at 15 °C	kg/m^3	ASTMD 4052	832.5	825.8	798.6	740.94
Viscosity at 40 °C	mm^2/s	ASTM D 455	3.0	2.7	1.173	0.590
Viscosity at -20 °C	mm^2/s	ASTM D 455			3.899	
Flash Point	°C	ASTM D93	68	64		
	°C	IP 170			47	
Cetane Number	-	ASTM D613	55.0	54.0		
Oxidation Stability (at 110 °C)	h	EN 14112				
Oxidation Stability	mg/L	ASTM D2274		3		
Total acid number	mg KOH/g	ASTM D3242			0.004	
Water content	mg/kg	ASTM D6304	53	41		
CFPP	°C	EN 116				
		IP 309	-20	-8		
Cloud point	°C	ASTM D5771	-15	-6	-87	-75
Pour point	°C	ASTM D5950	-23	-13		-75
Sulphur content	mg/kg	EN ISO 20846				
	mg//kg	ASTM D5453	4.8	4.9		
	WT PCT	IP 336			0.07	
Ash content	WT PCT	ASTM D 482	0.0	0.0		
Octane number, Mon		ASTM D2700				85.1
Octane number, Ron		ASTM D 2699				95
Evaporated at 70 °C	VOL PCT	ASTM D86				41.4
Evaporated at 100 °C	VOL PCT	ASTM D86				58.6
Evaporated at 150 °C	VOL PCT	ASTM D86				81.9
Distillation residue	VOL PCT	ASTM D86			1.4	1

2.3. Evaluation of Kinematic Viscosity and Density

The kinematic viscosity and density of the fuel samples were measured using Ubbelohde viscometers and pycnometer with a bulb capacity of 25 ml following the ASTM D445 [26] and ASTM D854 [27] standards, respectively. In this study, each test was repeated 4 times and then the average was determined to reduce the experimental error. Details of the measurements were given in Refs. [21,28].

2.4. Evaluation of Oxidative Stability

Oxidative stability was evaluated using the Rancimat instrument following the standard EN 14112 [29]. The description of this reaction is discussed in Ref. [21].

2.5. Evaluation of Cloud Point and Pour Point

Cloud point (CP) and pour point (PP) were evaluated according to STM D2500 [30] and ASTM D97 [31], respectively. Measurements of CP and PP of the fuel samples are discussed in Ref. [21].

2.6. Preparation of Fuel Mixtures

In this study, PME-WFME blends were mixed with each other in various percentages from 20 to 80 (%v/v) in steps of 20 (%v/v), whereas biodiesel-selected fuel blends were mixed with each other in various percentages (5%, 7%, 10%, 15%, and 20%) and homogenized with the aid of a magnetic stirrer for 30 min. It should be noted that biodiesel/petroleum-based fuel blends with 7% v/v and 10% v/v were chosen based on the EN 590 and EN 16734 standards.

3. Results

3.1. Characterization of Pure Biodiesel

The physicochemical properties (KV, D, OX, CP, and PP) of the pure biodiesel samples are discussed and compared below.

3.1.1. Kinematic Viscosity

Table 3 shows the KV values of the biodiesel samples produced from waste/residential frying oil and palm oil at 40 °C. It is observed that the KV values at 40 °C ranged between 4.33 and 5.71 mm^2/s. The minimum and maximum values of KV were obtained from WFME18 and WFME5, respectively. Based on the results in Table 3, it was found that the KV values of all samples were within the range of the ASTM D445 (1.9-6.0 mm^2/s). The EN ISO 3104 standard requires viscosity values at 40 °C of 3.5-5.0 mm^2/s, and all biodiesel samples met this requirement except for WFME3, WFME4, WFME5, WFME6, WFME7, and WFME8, which had KV values above the maximum limit of the kinematic viscosity specification in EN ISO 3104. In the case of the ASTM D975 standard (1.9–4.10 mm^2/s), the KV values of all biodiesel samples were above the maximum kinematic viscosity specification. Furthermore, in the case of European regulation EN 590, the KV values of WFME18, WFME19, WFME20, WFME28, WFME29, WFME36, WFME37, and WFME38 were below the maximum value for KV specified in the standard, while the other samples were above the maximum value of 4.0 mm^2/s.

Table 3. Kinematic viscosity of all biodiesel samples at 40 °C.

Sample	KV [mm^2/s]	Sample	KV [mm^2/s]	Sample	KV [mm^2/s]
WFME1	4.55	WFME15	4.64	WFME29	4.42
WFME2	4.78	WFME16	4.63	WFME30	4.59
WFME3	5.01	WFME17	4.62	WFME31	4.57
WFME4	5.24	WFME18	4.33	WFME32	4.65
WFME5	5.71	WFME19	4.36	WFME33	4.63
WFME6	5.57	WFME20	4.49	WFME34	4.61
WFME7	5.38	WFME21	4.51	WFME35	4.60
WFME8	5.11	WFME22	4.57	WFME36	4.37
WFME9	4.94	WFME23	4.59	WFME37	4.43
WFME10	4.75	WFME24	4.63	WFME38	4.48
WFME11	4.62	WFME25	4.60	WFME39	4.55
WFME12	4.55	WFME26	4.58	PME	4.68
WFME13	4.58	WFME27	4.56		
WFME14	4.66	WFME28	4.37		

Moreover, the results for KV in the present study were compared with those in the literature. It was found that the KV values of all samples of biodiesel derived from waste frying oil were close to the KV values obtained from the literature [22,23,32,33]. Additionally, the KV value for PME (4.68 mm2/s) was similar to Ref. [34], at 4.65 mm^2/s. Further, it was found that the KV value for PME was within the range of values obtained in the literature (4.6889 mm^2/s [35], 4.52 mm^2/s [36], 4.5 mm^2/s [37], 5.07 mm^2/s [38], 4.42 mm^2/s [39], 4.1 mm^2/s [40], 4.5 mm^2/s [41], 4.66-4.87 mm^2/s [42], 4.5 mm2/s [43], 4.71 mm^2/s [44], and 4.33 mm^2/s).

It was noticed that the WFME3, WFME4, WFME5, WFME6, WFME7, and WFME8 had the highest value for KV compared to other types of WFMEs and PME. This behavior occurs because kinematic viscosity increases with chain length and unsaturation level, as shown in Table S1 in the Supplementary Materials. Along with these major fatty acids, there were other varieties of fatty acids in minor amounts that had some influence on fuel properties. However, no single fatty acid was responsible for any particular fuel property. Generally, according to Ref. [45], biodiesel properties obtained from waste vegetable oils are dependent on the frying conditions and fatty acid composition of methyl ester. Additionally, the viscosity of methyl ester increases with increasing chain length and decreases with increasing level of unsaturation [46]. Furthermore, the viscosity of biodiesel depends on the structural composition of oil used in biodiesel production. Viscosity increases with the number of CH2 moieties in the fatty ester chain [47]. This implies that the presence of multiple bonds imparts lower viscosity to biodiesel.

Moreover, the kinematic viscosities of biodiesel were measured at various temperatures and 1 atm. As an example, Figure 2 shows the KV as a function of the temperature for selected samples. The results indicate that the KV of all samples of biodiesel decreased with the increase of temperature. This behavior was obtained for all fuel samples. According to Kafuku and Mbarawa [48], the crystallization of methyl esters, especially the saturated methyl esters, causes the increase of viscosity at low temperatures.

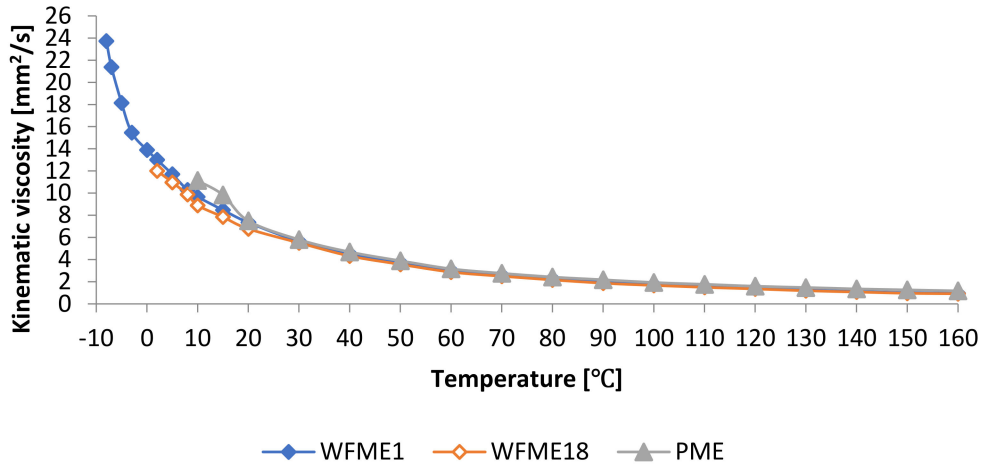

Figure 2. Kinematic viscosity–temperature relationship for selected biodiesel samples.

3.1.2. Density

The most important property of any fuel is density (D), which is defined as mass per unit volume. A fuel with a high mass/unit volume ratio (D) affects the functionality of an engine and the injector mechanism. According to EN 14214:2012, the recommended values for density vary from 860 to 900 kg/m^3 at 15 °C. Figure 3 (an example) shows the effect of temperature on the densities of some studied samples of biodiesel. As shown in this figure, densities are linearly related to temperature and decrease with increases in temperature, as expected.

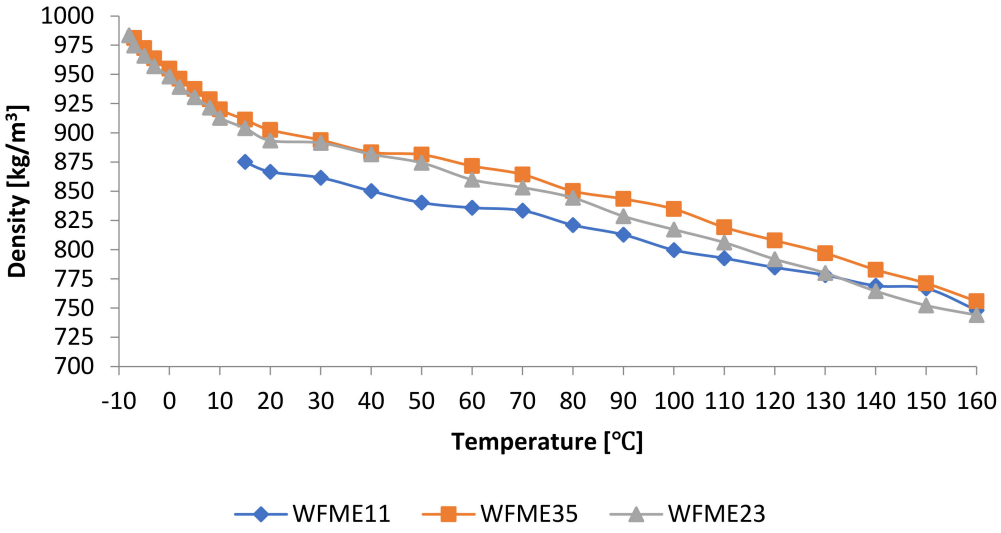

Figure 3. Density – Temperature relationship for selected biodiesel samples.

Table 4 shows the densities of all studied biodiesel samples measured at 15 °C. It was found that the obtained density data for all biodiesel samples were above the minimum recommended value of ASTM D854 (860 kg/m^3 at 15 °C). Data from Table 4 show that density values for WFME1–WFME11 and PME followed the standard demanded for diesel fuel (EN14214 standard recommended values: 860–900 kg/m^3 at 15 °C). Furthermore, it is seen that the density values for other samples (WFME12–WFME39) were above the maximum specified limit in EN14214. Comparison with other results in the literature indicated that PME possessed a slightly lower density (875 kg/m^3) than that of [41] at 878kg/m^3 and had a density comparable to that of [49] at 875 kg/m^3. Moreover, PME

possessed a higher density than that of [50] at 860 kg/m^3, but was lower than that of [36,43] at 898 kg/m^3 and 877–882 kg/m^3, respectively.

Table 4. Density of all biodiesel samples at 15 °C.

Sample	D [kg/m^3]	Sample	D [kg/m^3]	Sample	D [kg/m^3]
WFME1	895.4	WFME15	920.6	WFME29	933
WFME2	895.2	WFME16	910.4	WFME30	941.8
WFME3	895	WFME17	902,9	WFME31	932
WFME4	894.8	WFME18	933.5	WFME32	920.5
WFME5	894.3	WFME19	927.5	WFME33	915.8
WFME6	891.6	WFME20	921.7	WFME34	913.9
WFME7	887.6	WFME21	915.8	WFME35	911.2
WFME8	891.9	WFME22	909.9	WFME36	931.9
WFME9	894.8	WFME23	903.9	WFME37	924.7
WFME10	883.1	WFME24	934.5	WFME38	917.4
WFME11	875.2	WFME25	925.4	WFME39	910.1
WFME12	923.7	WFME26	923.1	PME	875.0
WFME13	907.6	WFME27	919.1		
WFME14	930.7	WFME28	936.1		

3.1.3. Oxidation Stability

The oxidation stability results obtained for all biodiesel samples are listed in Table 5. The highest and lowest stability values were obtained for PME and WFME15, with a value of 18.35 h and 6.14 h, respectively. This behavior can be explained on the basis of the content of the methyl esters linoleic (C18:2) and linolenic fatty acids (C18:3) because they are more susceptible to oxidation due to the presence of double allylic moieties in their chains [40]. Based on Table S1 in the Supplementary Materials, the content of C18:2 (11.00 wt%) and C18:3 (0.2 wt%) in PME was low, whereas WFME15 had a high content of C18:2 (18.4 wt%) and C18:3 (2.90 wt%). In comparison to the oxidative stability requirements contained in ASTM D6751 and EN 14214, all studied biodiesel samples were above the minimum specified limit in ASTM D6751 (> 3.0 h) and EN 14214 (> 6.0 h).In the case of EN 14214:2014 ($\geq$8 h), the results indicated that the stability values for WFME4–WFME10, WFME18–WFME24, WFME28–WFME39, and PME were above the minimum limit specified by the standard.

Table 5. Oxidation stability (OX) values for all biodiesel samples.

Sample	OX [h]	Sample	OX [h]	Sample	OX [h]
WFME1	7.25	WFME15	6.14	WFME29	11.40
WFME2	7.61	WFME16	7.78	WFME30	10.10
WFME3	7.99	WFME17	7.85	WFME31	8.80
WFME4	8.39	WFME18	14.00	WFME32	7.93
WFME5	8.81	WFME19	12.85	WFME33	8.02
WFME6	9.25	WFME20	11.71	WFME34	8.10
WFME7	8.79	WFME21	8.91	WFME35	8.19
WFME8	8.61	WFME22	9.42	WFME36	12.77
WFME9	8.44	WFME23	8.27	WFME37	11.54
WFME10	8.27	WFME24	8.12	WFME38	10.31
WFME11	7.56	WFME25	7.96	WFME39	9.08
WFME12	7.50	WFME26	7.81	PME	18.35
WFME13	7.57	WFME27	7.65		
WFME14	7.64	WFME28	12.70		

Moreover, the stability of all studied pure biodiesel samples in the present study was compared with those in the literature. PME possessed lower oxidation stability (18.35 h) than that of [41] with 23.56 h, but higher than that of [38,40] with 4.0 h and 8.85 h, respectively. In addition, the stability of PME was found to be within the range of 7.94–18.9 h [43]. Furthermore, the stability of WFME15 was comparable to that of [51,52] at 6.1 h. In general,

for biodiesel produced from waste frying oil, the stability of all samples ranged from 6.14 h (WFME15) to 14.00 h (WFME18). This range of values was considerably higher than the stability (0.91 h) found by Uzun et al. [53], who produced waste frying oil biodiesel with a higher unsaturated fatty acid content (89.68 wt.%) in comparison to the samples produced in this study (75.1 wt.%), and the stability (2.88 h) found by Bharti and Singh [54].

3.1.4. Cold Flow Properties

Tables 6 and 7 show the obtained CP and PP values for all biodiesel samples, respectively. It was found that the CP and PP values were within the range of −2.0–17.4 °C and −10.0–15.0 °C, respectively. The considerably higher CP and PP values obtained for WFME11 and PME were attributed to the presence of a higher percentage of SFAME (42.99 wt% for WFME11 and 44.90 wt% for PME), as it is known that the melting point increases with decreasing double bond content.

Table 6. Cloud point (CP) values for all biodiesel samples.

Sample	CP [°C]	Sample	CP [°C]	Sample	CP [°C]
WFME1	−1.00	WFME15	7.60	WFME29	3.40
WFME2	0.70	WFME16	10.00	WFME30	1.60
WFME3	2.50	WFME17	7.00	WFME31	−0.20
WFME4	4.40	WFME18	5.36	WFME32	7.76
WFME5	5.60	WFME19	3.72	WFME33	5.52
WFME6	9.00	WFME20	2.08	WFME34	3.28
WFME7	11.30	WFME21	0.44	WFME35	1.04
WFME8	12.70	WFME22	−1.20	WFME36	7.60
WFME9	13.70	WFME23	−1.36	WFME37	8.20
WFME10	15.90	WFME24	−1.52	WFME38	8.80
WFME11	17.40	WFME25	−1.68	WFME39	9.40
WFME12	−2.00	WFME26	−1.84	PME	15.00
WFME13	0.40	WFME27	5.20		
WFME14	2.80	WFME28	7.60		

Table 7. Pour point (PP) values for all biodiesel samples.

Sample	PP [°C]	Sample	PP [°C]	Sample	PP [°C]
WFME1	−9.00	WFME15	−0.70	WFME29	−3.04
WFME2	−6.80	WFME16	2.40	WFME30	−5.36
WFME3	−4.60	WFME17	5.50	WFME31	−7.68
WFME4	−2.40	WFME18	1.60	WFME32	3.20
WFME5	−1.00	WFME19	0.08	WFME33	0.90
WFME6	1.70	WFME20	−1.44	WFME34	−1.40
WFME7	3.40	WFME21	−2.96	WFME35	−3.70
WFME8	5.90	WFME22	−4.48	WFME36	2.38
WFME9	7.50	WFME23	−6.00	WFME37	3.16
WFME10	9.60	WFME24	−6.80	WFME38	3.94
WFME11	11.10	WFME25	−7.60	WFME39	4.72
WFME12	−10.00	WFME26	−8.40	PME	15.00
WFME13	−6.90	WFME27	−9.20		
WFME14	−3.80	WFME28	−0.72		

The comparison of results obtained in the current study with those in the literature showed that WFME1–WFME39 possessed comparable CPs and PPs of -2–17.4°C and −10–11 °C, respectively, compared to the values obtained in Refs. [21–23,28,32,53,54]. The CP value for PME was lower than that found by May et al. [44] and Benjumea et al. [55] (CP = 16 °C), while it was similar the values reported in Refs. [38,41]. The PP result for PME was similar to that reported in Refs. [41,44].

3.2. Effect of Blends of Palm Oil Biodiesel on Waste/Residential Frying Oil Biodiesel

In this study, PME was added to all samples of biodiesel produced from waste frying oil in various proportions (20%, 40%, 60%, and 80%) to improve the oxidation stability mainly of the WFME samples that had stability of less than 8 h (EN 14214:2014). Figure 4 shows the properties (kinematic viscosity, density, oxidation stability, CP, and PP) of the PME-WFME blends. Table 8 presents the sample number and name of each biodiesel used in this study.

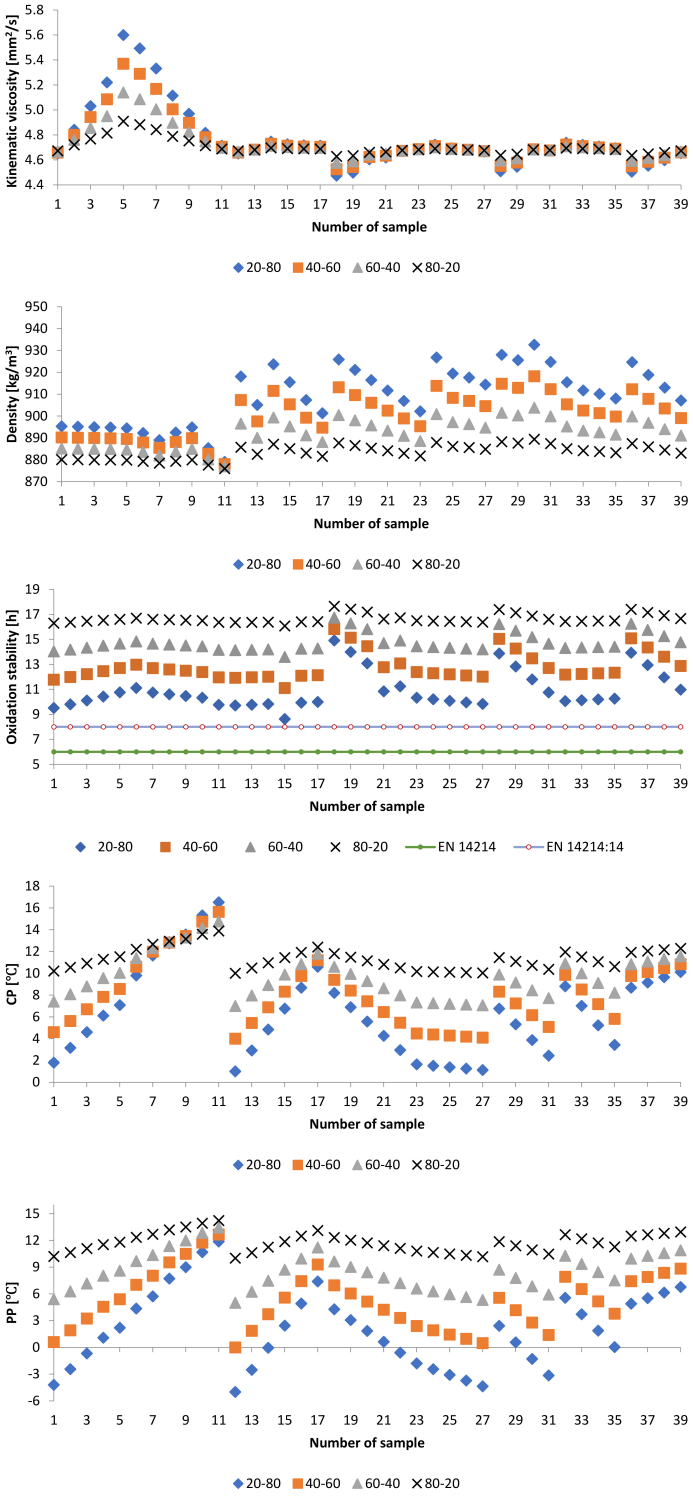

Figure 4. Properties of the mixture (PME-WFME) with various ratios (20:80 means 20% v/v of palm oil biodiesel and 80% .

v/v of waste frying oil biodiesel; 40:60 means 40% v/v of palm oil biodiesel and 60% v/v of waste frying oil biodiesel; 60:40 means 60% v/v of palm oil biodiesel and 40% v/v of waste frying oil biodiesel; and 80:20 means 80% v/v of palm oil biodiesel and 20% v/v of waste frying oil biodiesel)

Table 8. PM-WFME samples used in this study.

Sample No.	Sample Name	Sample No.	Sample Name	Sample No.	Sample Name
1	PMWFME1	14	PMWFME14	27	PMWFME27
2	PMWFME2	15	PMWFME15	28	PMWFME28
3	PMWFME3	16	PMWFME16	29	PMWFME29
4	PMWFME4	17	PMWFME17	30	PMWFME30
5	PMWFME5	18	PMWFME18	31	PMWFME31
6	PMWFME6	19	PMWFME19	32	PMWFME32
7	PMWFME7	20	PMWFME20	33	PMWFME33
8	PMWFME8	21	PMWFME21	34	PMWFME34
9	PMWFME9	22	PMWFME22	35	PMWFME35
10	PMWFME10	23	PMWFME23	36	PMWFME36
11	PMWFME11	24	PMWFME24	37	PMWFME37
12	PMWFME12	25	PMWFME25	38	PMWFME38
13	PMWFME13	26	PMWFME26	39	PMWFME39

3.2.1. Kinematic Viscosity at 40 °C and Density at 15 °C of PME-WFME Blends

The KV values of the WFME samples without adding PME were varied, from 4.42 mm^2/s (WFME18) to 5.83 mm^2/s (WFME5) (see Table 3). After adding PME in various concentrations, the KV results ranged between 4.47 mm^2/s (P20WFME18) and 5.60 mm^2/s (P20WFME5), i.e., the kinematic viscosity of WFME5 decreased by 0.230 mm^2/s, whereas that of WFME18 increased by 0.015 mm^2/s. It should be noted that binary blending was performed as PXWFMEY, where X means the weight percentage of biodiesel in the blends of WFMEY, and Y represents the number of the sample. For example, P40WFME17 corresponded to a 40% concentration of PME added to biodiesel (WFME) sample number 17. It was also found that the kinematic viscosity of WFME samples could be decreased by 15.77% or increased by 4.65% when the concentration percentage of PME was 80%. It can be concluded that it was necessary to add a high proportion of PME to the WFME samples with high viscosity greater than 5 mm^2/s in order to conveniently reduce their kinematic viscosity, as shown in Figure 4. In this case, the kinematic viscosity results for the mixture corresponded with the recommended EN 14214 values: 3.5–5.0 mm^2/s at 40°C, i.e., the KV results for P80WFME samples were below the maximum limit of the kinematic viscosity specification in EN 14214 (5.0 mm^2/s). This observation could be associated with a hydroxy (OH) group present in the methyl ricinoleate molecule, according to Mejia et al. [34].

Generally, in ASTM D6751, there is no specification for biodiesel density, but in EN 14214, the acceptable range for biodiesel density should be between 850–900 kg/m^3. Therefore, it was observed that the density decreased by approximately 6% at 80 vol% PME in the PME-WFME blends. For example, the initial density of WFME1 was 900.41 kg/m^3; after adding 80 vol% PME, the density of the mixture P80WFME1 was 880.8 kg/m^3, as shown in Figure 4. This observation could be associated with the significant proportion of polyunsaturated fatty acids and unsaturated compounds such as linoleic acid (C18:2) and oleic acid (C18:1) in the fuel [56,57].

3.2.2. Oxidation Stability of PME-WFME Blends

One of the main problems with storing biodiesel is that the fuel reacts with oxygen in the air and this causes the degradation of the fuel. According to Giakoumis [58], Hoekman et al. [59], and Kumar [60], degradation is caused by the double bonds in fatty acid structure that render the fuel more likely to be affected by the presence of air. In addition, acid number (acidity), peroxide value (rancidity), air, metals, heat, light or pressure,

and polymer amount are all factors that influence the degradation stability of biodiesel, according to Knothe [61]. EN 14214:14 and ASTM D 6751 have set the minimum value of oxidative stability for biodiesel to 8 h and 3 h, respectively, using the Rancimat method. Based on our results (Figure 4), the oxidation stability of all samples was above the minimum specified limit in ASTM D6751 (> 3.0 h). As mentioned previously, 26 WFME samples met the oxidation stability of the recommended specification by EN 14214:2014. In general, adding oxidant inhibitors improves the oxidation stability of a fuel, therefore enabling it to be stored for a longer period without degrading [62]. Adding oxidant inhibitors to biodiesel increases the oxidation stability, and the higher the concentration of inhibitors, the higher the oxidation stability due to the presence of the OH group. As reported in Ref. [21], the oxidation stability of the mixture of WFME-WCME (waste frying methyl ester – waste canola methyl ester) did not meet the recommended oxidation stability specification in EN 14214:2014 ($\geq$8h). Recently, Kassem et al. [22] found that frying sunflower and rapeseed oil methyl ester (FSRME) fell below the 8 h minimum at 2 months at all storage temperatures (5, 25, and 40 °C). Consequently, PME was used as an additive to enhance the stability of biodiesel derived from waste frying oils. With 20% PME added to the WFME samples, the oxidation stability of all samples was above the recommended specification given by EN 14214:14, as shown in Figure 4. Therefore, adding a high proportion of PME to WFME samples helped to increase the oxidation stability of biodiesel. As mentioned in the previous section, WFME15 had the minimum oxidation stability compared to the other WFME samples. Upon addition of antioxidant (20% PME), the oxidation stability of WFME15 increased from 6.14 h to 8.62 h and accordingly met the recommended oxidation stability specification in EN 14214:14 ($\geq$8h). These results indicated that the samples could be stored for longer periods under various storage conditions. This expectation should be investigated in the future, and the results compared with previous studies [21,22].

3.2.3. Cold Flow Properties of PME-WFME Blends

As in this study, many past studies have demonstrated that palm biodiesel has poor cold flow properties due to the predominant properties of saturated fatty acid in palm biodiesel [63,64]. Additionally, due to gum formation and crystallization of fuel particles, which are caused by the inferior cold flow properties of biodiesel, mixing palm biodiesel with other fuels such as diesel, kerosene, and ethanol helps to improve the cold flow properties of the mixture [64].

As mentioned previously, the CP and PP values were within the range of −2.0–17.4 °C and −10.0–15.0 °C, respectively, for the pure biodiesel samples including PME. Several authors have researched the cold flow properties of different types of biodiesels [21,64,65]. Therefore, the effect on cold flow properties of adding WFME to PME in various proportions is discussed in this section.

In this section, PME was blended with WFME samples to investigate its cold flow properties. Figure 5 shows the effect of fatty acids on the cold flow properties of biodiesel. It was found that the cold flow temperatures of PME-WFME blends varied from 1.0 °C to 16.52 °C for CP and from -5.0 °C to 11.88 °C for PP. It was observed that CP and PP of 13 °C and 15 °C, respectively, were lowered to 7.0 °C and 5.0 °C for the P60WFME12 blend with 60% addition of PME on a volume basis to WFME12 (see Figure 4), which indicated that WFME12 improved CP and PP by 46.15% and 66.67%, respectively, as shown in Figure 5. Further, it was found that WFME1, WFME12, and WFME27 were the most effective in improving the cold flow properties (CP and PP) of PME, as shown in Figure 5. Specifically, WFME1, WFME12, and WFME27 improved CP by 21.54–86.15%, 23.08–92.31%, and 22.83–91.32%, respectively, and PP by 32.00–128.00%, 33.33–133.31%, and 32.77–129.07% respectively, depending on the concentration of WFME in the mixture (PME-WFME), as shown in Figure 5.

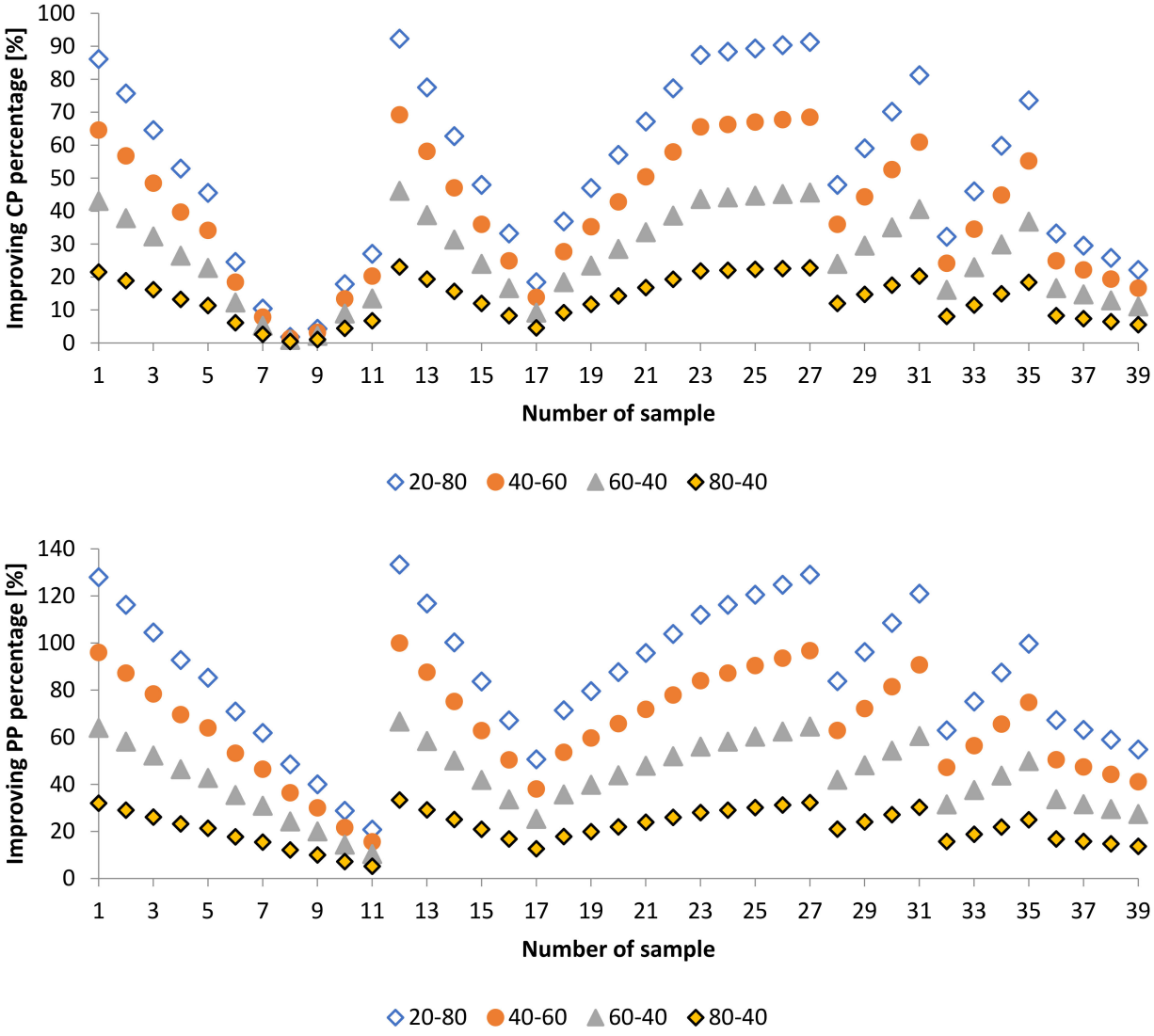

Figure 5. Improving cold flow percentage of the mixture (PME-WFME) with various ratios (20:80 means 20% v/v of palm oil biodiesel and 80% v/v of waste frying oil biodiesel; 40:60 means 40% v/v of palm oil biodiesel and 60% v/v of waste frying oil biodiesel; 60:40 means 60% v/v of palm oil biodiesel and 40% v/v of waste frying oil biodiesel; and 80:20 means 80% v/v of palm oil biodiesel and 20% v/v of waste frying oil biodiesel).

Generally, various authors have used petroleum diesel, kerosene, and ethanol to improve the low-temperature properties of palm biodiesel. For instance, Verma et al. [64] blended palm biodiesel with petroleum diesel, kerosene, and ethanol to improve the cold flow properties of biodiesel. They found that petroleum diesel, kerosene, and ethanol improved CP by 57.61%, 78.57%, and 60.48%, respectively, and improved PP by 78.57%, 85.78%, and 63.96% respectively for the B20 blend (20% biodiesel and 80% petroleum diesel, kerosene, or ethanol). Based on our results and those of previous studies [63,64], it can be concluded that WFME with low CP and PP can be used as cold flow improvers to improve the cold flow properties of PME.

3.3. Effect of Adding Petroleum-Based Fuels to Pure Biodiesel Samples on the Properties of the Mixture

Biodiesel is considered an alternative energy source to replace petroleum-based fuels in the transportation sector. Moreover, due to flow problems especially in cold weather conditions, viscosity, density, pour point, and cloud point are considered as the main properties that affect biodiesel flow. Furthermore, to meet the kinematic viscosity and

density specifications in ASTM D975 (recommended values: 1.9–4.10 mm^2/s at 40 °C and 860–900 kg/m^3 at 15 °C) and EN 90 (recommended values: 2.0-4.5 mm^2/s at 40 °C), it is hence necessary to blend biodiesel with petroleum-based fuel. Thus, ULSDFW, ULSDFS, KF, and BF were utilized to improve the properties of biodiesel and solve problems that occur due to cold flow.

Figures 6–9 present the experimental results of kinematic viscosity at 40 °C and density at 15 °C for WFME and PME biodiesel samples blended with the petroleum-based fuels (ULSDFW, ULSDFS, KF, and BF). Table 9 presents the sample number and name of each biodiesel used in this study. The initial values of kinematic viscosity and density for all biodiesel samples varied from 5.83 mm^2/s to 4.42 mm^2/s and from 875.00 kg/m^3 to 947.02 kg/m^3, respectively. Based on the results, it was found that few biodiesel samples met the recommended kinematic viscosity and density specifications in the ASTM D975 and EN 90 standards, as shown in Figures 6 and 7. Moreover, the results indicated that the kinematic viscosities of biodiesel petroleum-based fuels (KF and BF) with 10%, 15%, and 20 vol% KF and BF were in line with the specifications in diesel fuel standards (ASTM D975 and EN 90), as shown in Figure 6. Similar results were obtained by Kassem et al. [22], who found that BG85 (15% gasoline and 85% biodiesel) and BG80 (20% gasoline and 80% biodiesel) met the mixed pure biodiesel with gasoline diesel fuel quality standards. Additionally, the kinematic viscosity of all studied blends of biodiesel with petroleum-based fuels was reduced with increases in petroleum-based fuel content. Furthermore, it was observed that some blends were above the maximum recommended kinematic viscosity specifications in the ASTM D975 and EN 90 standards, as shown in Figures 6–9.

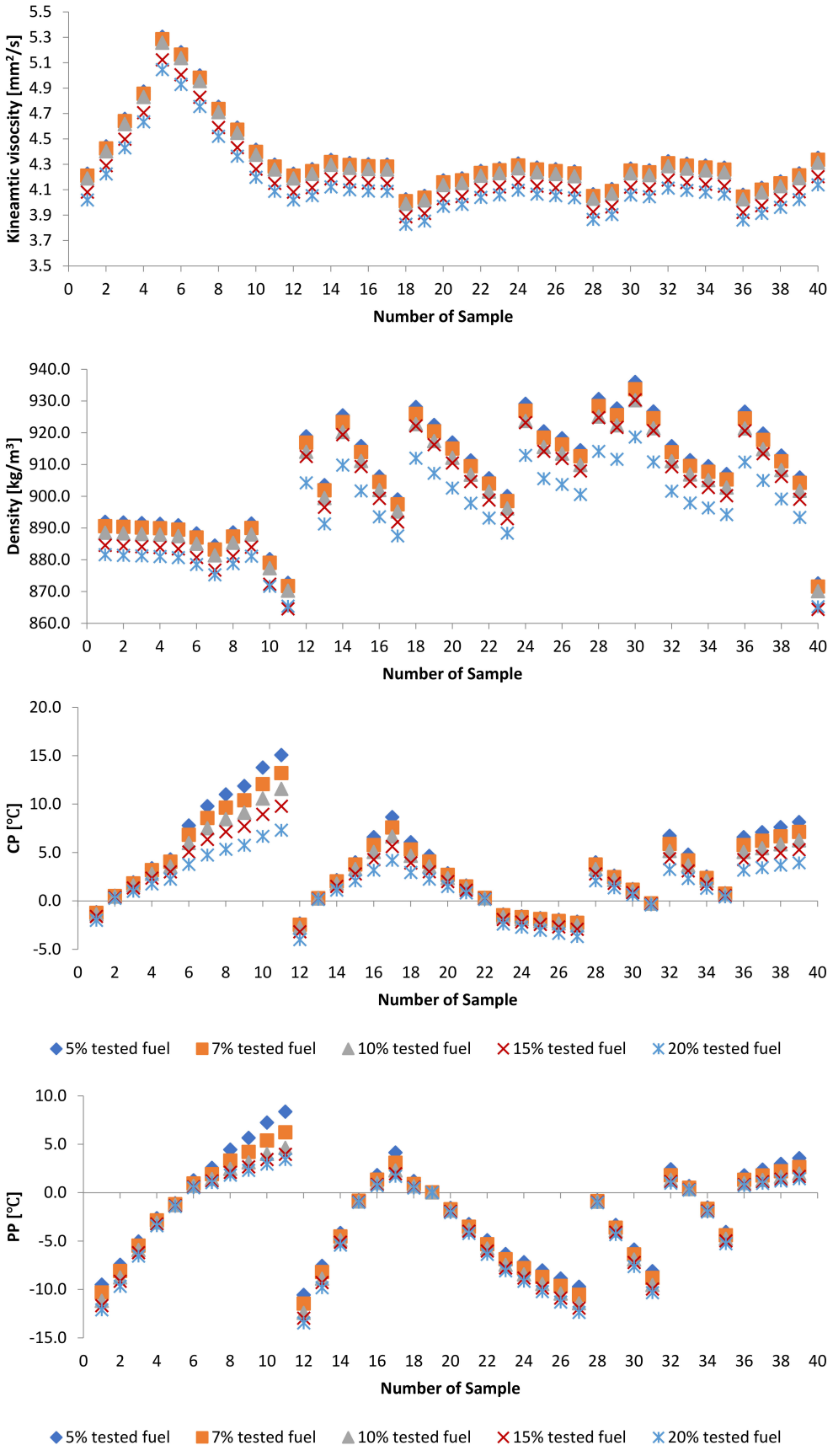

Figure 6. Properties of biodiesel-ULSDFW blends with various volume ratios.

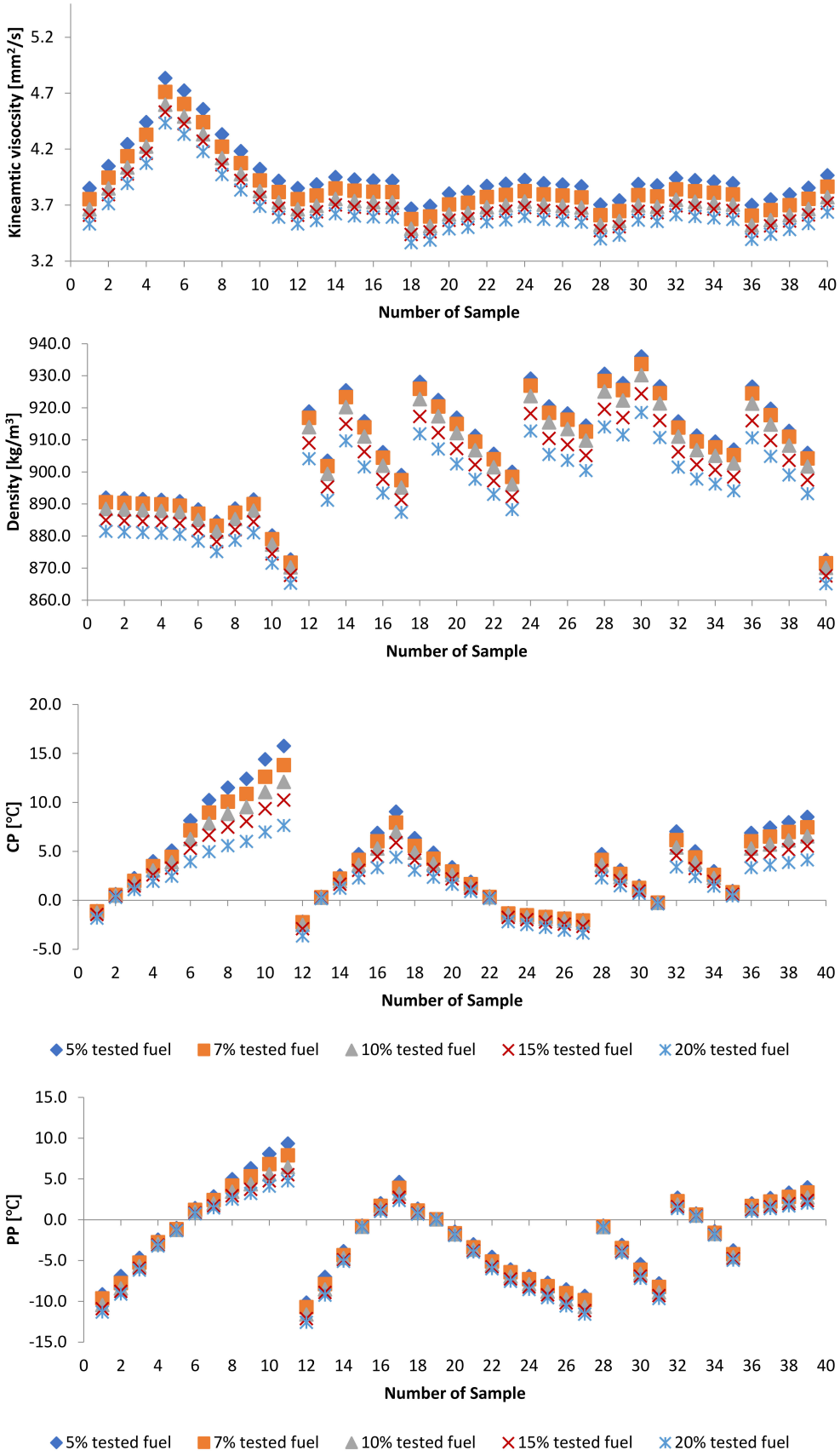

Figure 7. Properties of biodiesel-ULSDFS blends with various volume ratios.

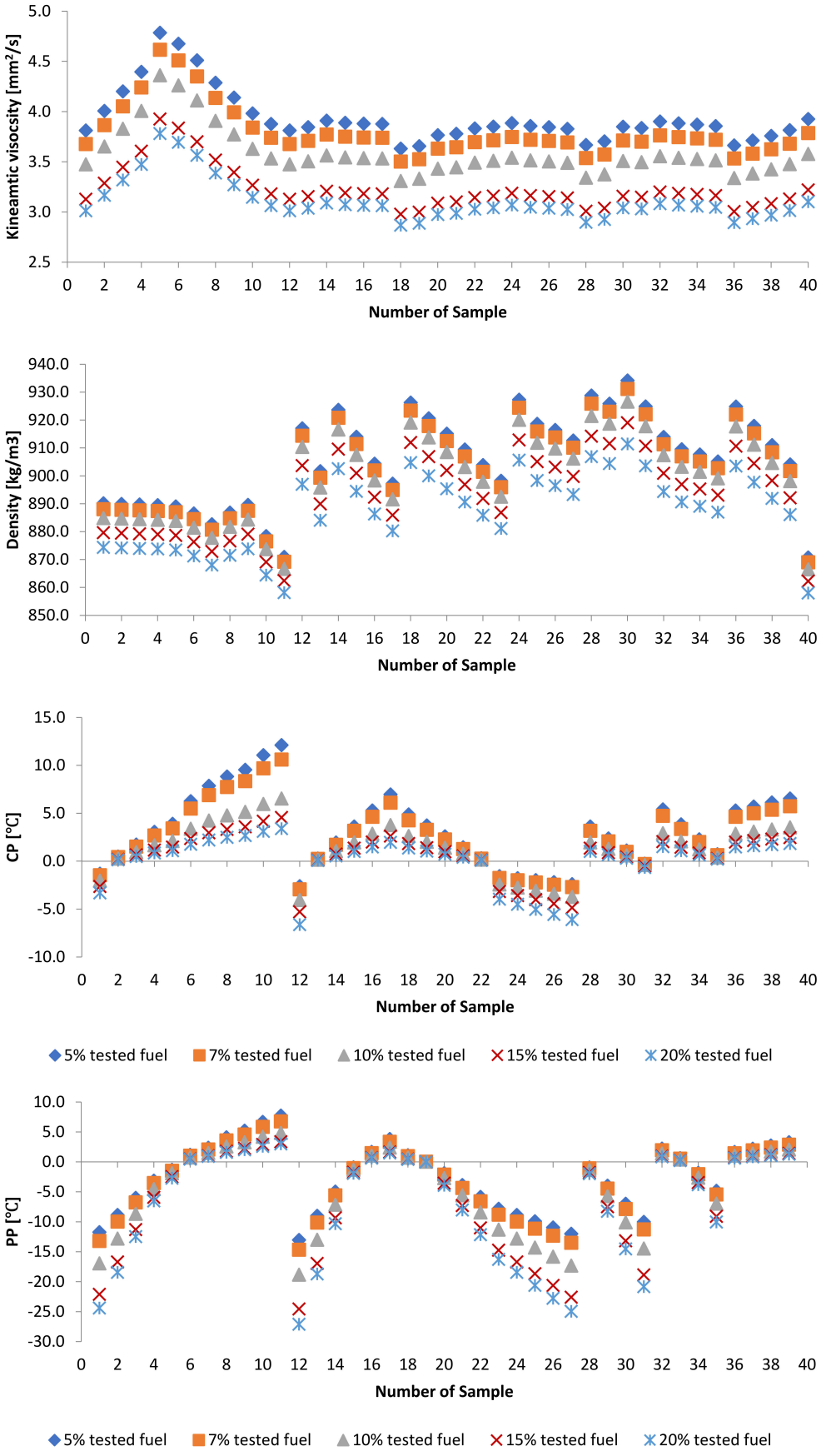

Figure 8. Properties of biodiesel-KF blends with various volume ratios.

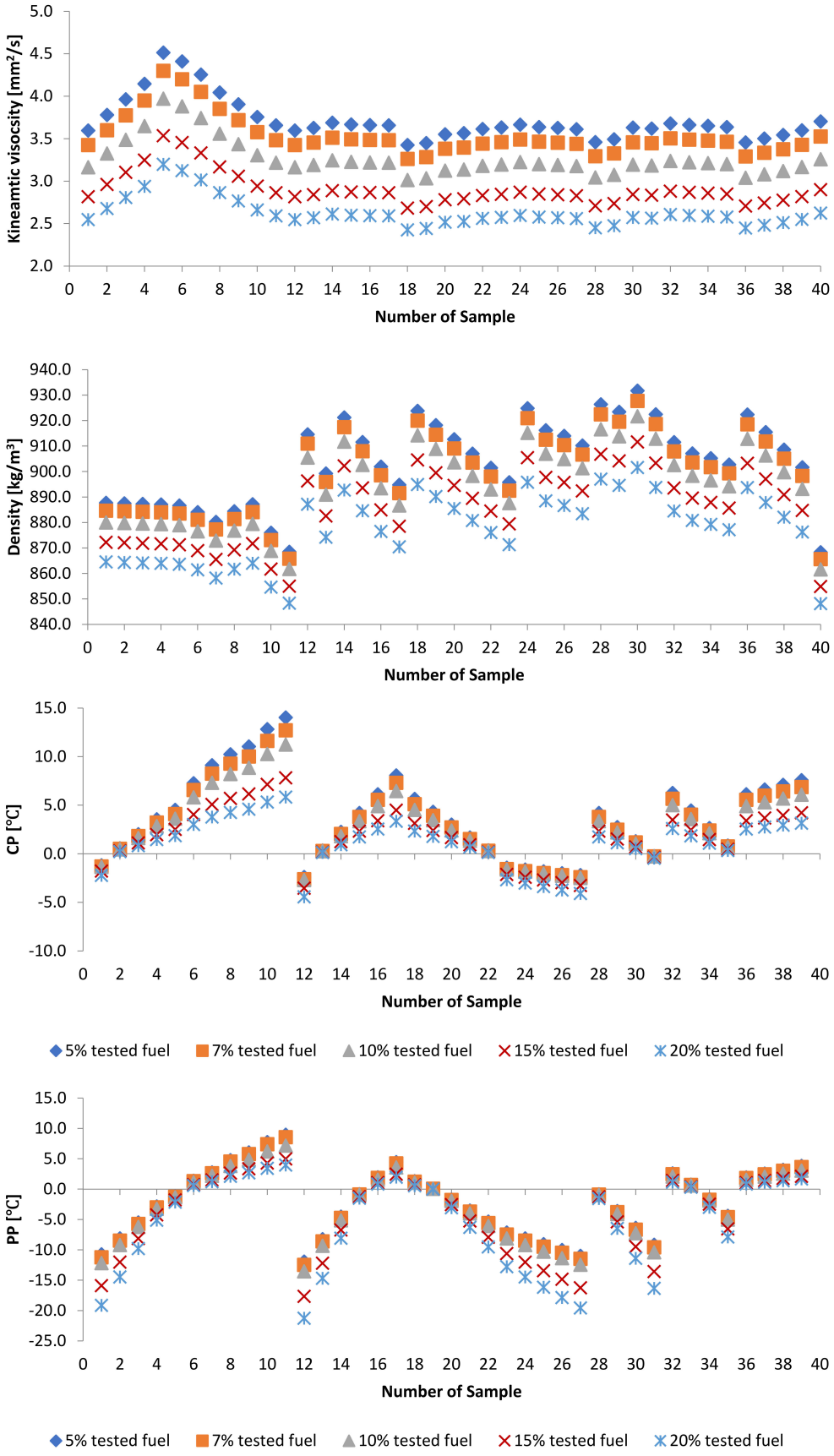

Figure 9. Properties of biodiesel-BF blends with various volume ratios.

Table 9. Pure biodiesel samples used in this study.

Sample No.	Sample Name	Sample No.	Sample Name	Sample No.	Sample Name	Sample No.	Sample Name	Sample No.	Sample Name
1	WFME1	9	WFME9	17	WFME17	25	WFME25	33	WFME33
2	WFME2	10	WFME10	18	WFME18	26	WFME26	34	WFME34
3	WFME3	11	WFME11	19	WFME19	27	WFME27	35	WFME35
4	WFME4	12	WFME12	20	WFME20	28	WFME28	36	WFME36
5	WFME5	13	WFME13	21	WFME21	29	WFME29	37	WFME37
6	WFME6	14	WFME14	22	WFME22	30	WFME30	38	WFME38
7	WFME7	15	WFME15	23	WFME23	31	WFME31	39	WFME39
8	WFME8	16	WFME16	24	WFME24	32	WFME32	40	PME

Moreover, the densities of WFME11 and PME + BF (20 vol%) samples were found to be below the minimum specification limits (860 kg/m^3), as shown in Figure 7. At 15 °C, the density of WFME11 was reduced from 875.19 to 848.34 kg/m^3 and from 875.19 to 858.15 kg/m^3, respectively, after being blended with 20% benzene and 20% kerosene, as shown in Figure 7. Additionally, a decrease in density from 875.00 to 857.89 kg/m^3 was observed when PME was blended with 10% benzene, whereas a decrease in density from 875.00 to 843.19 kg/m^3 was observed when 20% benzene was blended with PME, as shown in Figures 6–9.

Moreover, Figures 6–9 illustrate the variations of CP and PP of the biodiesel samples and their blends. It was observed that blending at the selected ratio with petroleum-based fuels (ULSDFW, ULSDFS, KF, and BF) improved the cold flow properties of biodiesel samples. For example, a reduction in CP from 17.4 °C to 7.6 °C, from 17.4 °C to 7.3 °C, from 17.40 °C to 3.4 °C, and from 17.40 °C to 5.8 °C was recorded when WFME11 was blended with 20% ULSDFS, ULSDFW, KF, and BF, respectively. Similarly, a reduction in PP from 11.1 °C to 4.8 °C, from 11.1 °C to 3.4 °C, from 11.1 °C to 3.02 °C and from 11.1 °C to 4.0 °C was observed when WFME11 was blended with 20% ULSDFS, ULSDFW, KF, and BF, respectively. For PME, a reduction in CP from 13.0 °C to 5.7 °C, from 13.0 °C to 5.5 °C, from 13.0 °C to 2.5 °C, and from 17.40 °C to 4.4 °C was observed when blended with 20% ULSDFS, ULSDFW, KF, and BF, respectively. Similarly, a reduction in PP from 15.0 °C to 6.4 °C, from 15.0 °C to 4.6 °C, from 15.0 °C to 4.09 °C, and from 15.0 °C to 5.4 °C was achieved when PME was blended with 20% ULSDFS, ULSDFW, KF, and BF, respectively. It can be concluded that 20 vol% kerosene samples showed the best results as the reported CP and PP for all fuel samples were due to the low CP and PP for kerosene. Similar results were obtained by Verma et al. [64], Lv et al. [65], Nainwal et al. [66], and Dwivedi and Sharma [67]. Thus, the results obtained in the current study were consistent with those reported in the literature.

3.4. WFME–PME-Petroleum-Based Fuels

Because of the high yield of palm oil compared to other vegetable oils, it has earned attention as a source for the production of biodiesel [68,69]. In general, cold flow properties and stability of fuel are the main problems associated with utilizing biodiesels such as palm biodiesel as an alternative to diesel. Several researchers have sought solutions to these problems. For instance, Sarin et al. [70] studied the physicochemical properties of Jatropha-palm biodiesel and their blends to improve the cold flow properties and oxidation stability of the samples. They found that the optimum mixture of Jatropha biodiesel with palm biodiesel could enhance the cold flow properties and stability of the fuel. Verma et al. [64] utilized cold flow improvers (petroleum diesel, kerosene, and ethanol) to improve the cold flow properties of biodiesel derived from palm oi1. They found that cold flow improvers improved the CP and PP of palm biodiesel by an average of 65.55% and 76.10%, respectively, when blended with 20% of petroleum diesel, kerosene, and ethanol.

Based on the results of the previous section (Section 3.1), the investigation of the experimental results showed that WFME1, WFME12, WEME23, WFME24, WFME25, WFME26, and WFME27 had the lowest values of CP and PP compared to other biodiesel samples derived from waste frying oil. Thus, this section discusses an effort to improve the properties (KV,

D, CP, and PP) of selected WFME-PME samples by mixing them with various proportions of petroleum-based fuels (ULSDFW, ULSDFS, KF, and BF). Then, the results are compared with previous experimental results in the current study to achieve better low-temperature properties with improved oxidation stability. Figures 10–13 show the KV, D, CP, and PP of WFME-PME-petroleum-based fuel blends, respectively. The results showed that the viscosity and density values of the selected samples were in accordance with standards demanded for diesel fuel (ASTM D975 and EN 90), as shown in Figures 10–13, respectively. Table 10 presents the sample number and name of each fuel used in this study.

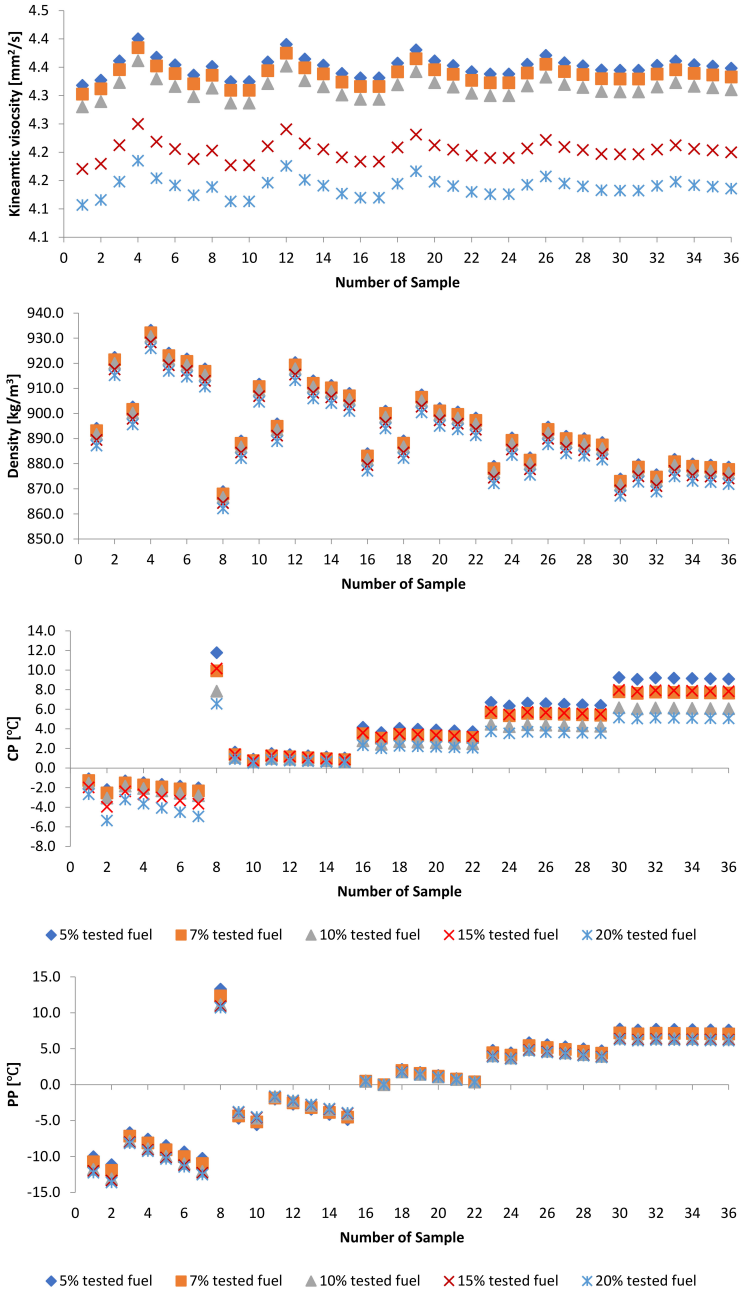

Figure 10. Properties of WFME-PME-ULSDFW with various volume ratios.

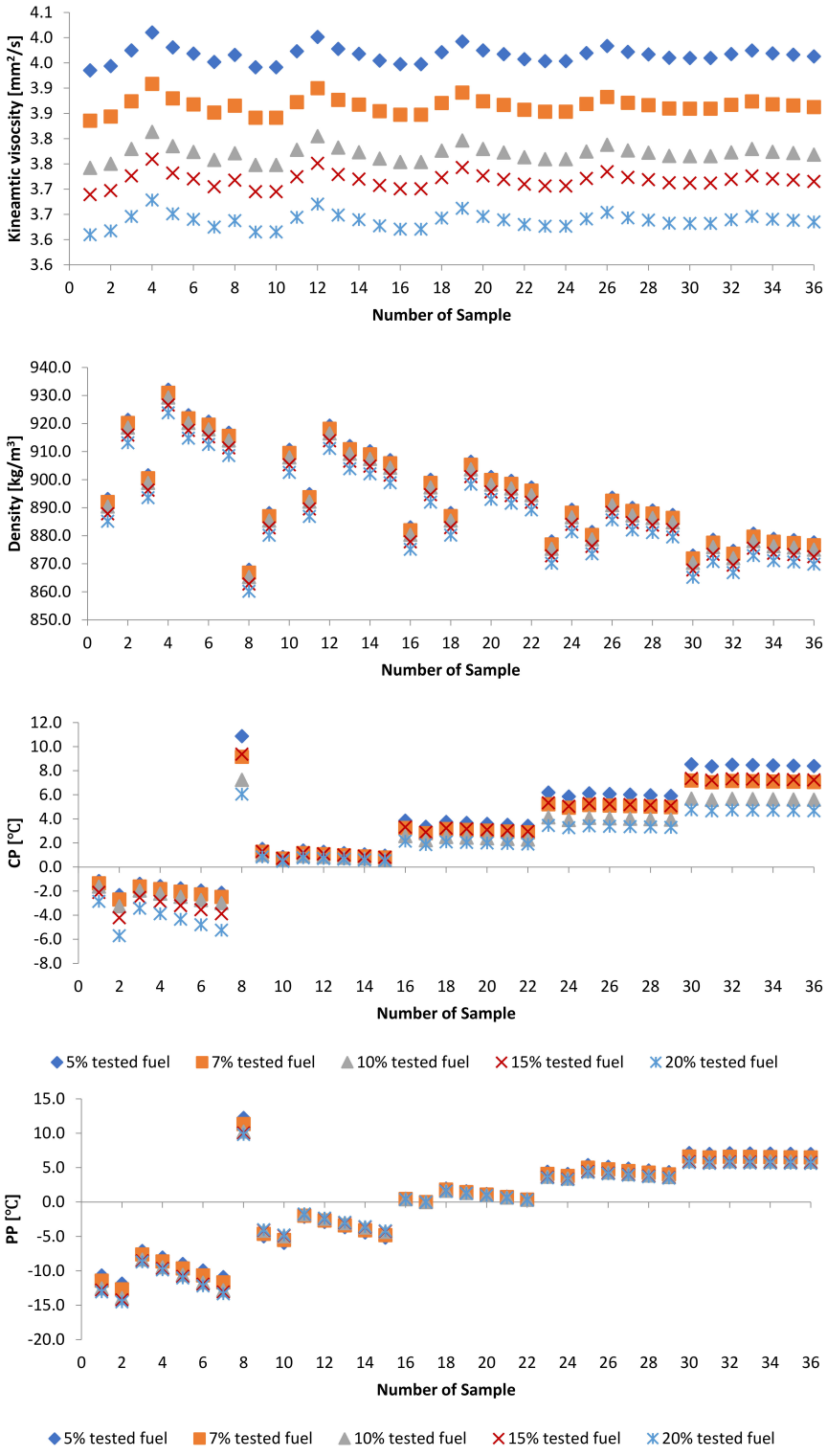

Figure 11. Properties of WFME-PME-ULSDFS with various volume ratios.

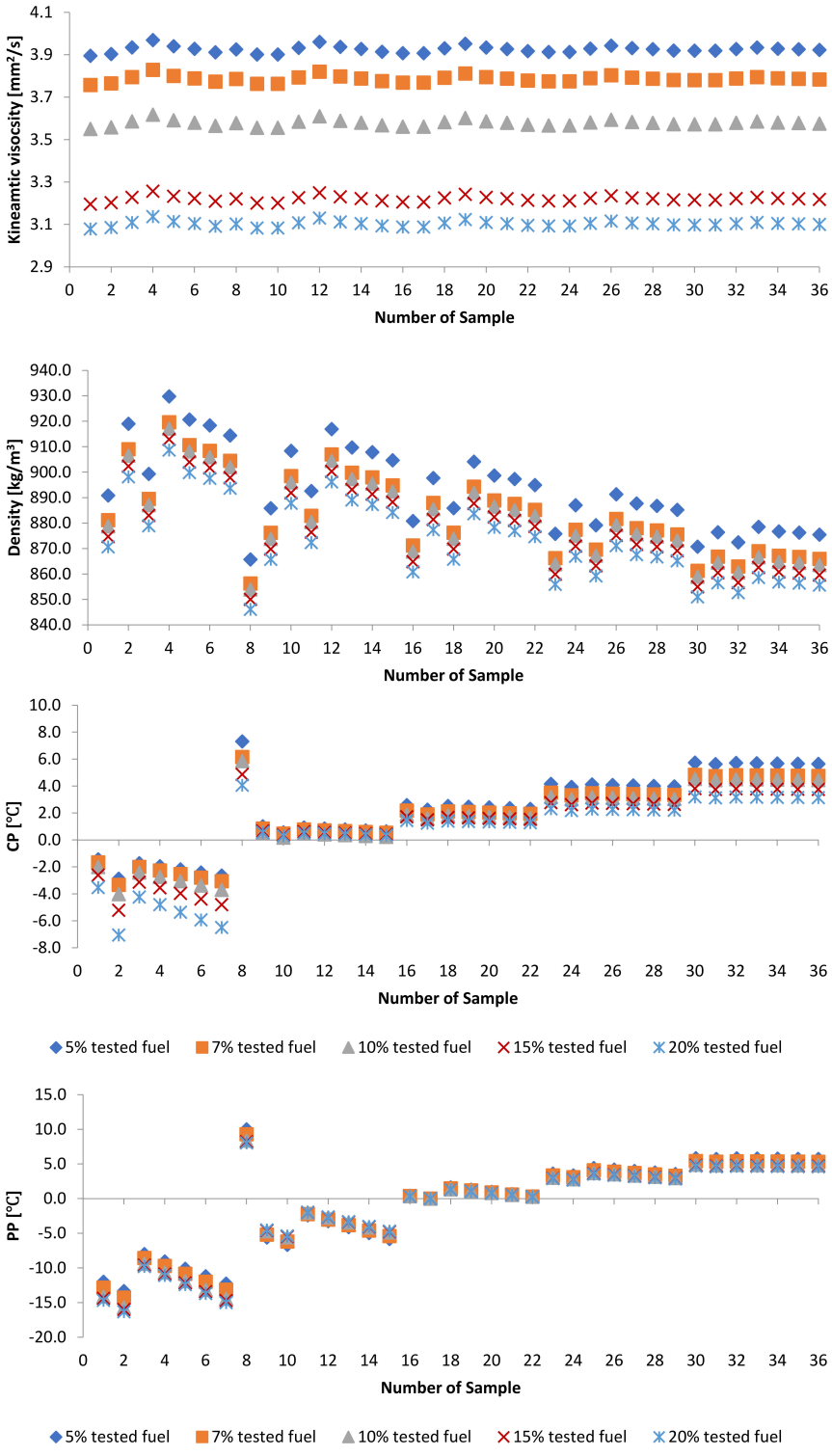

Figure 12. Properties of WFME-PME-KF with various volume ratios.

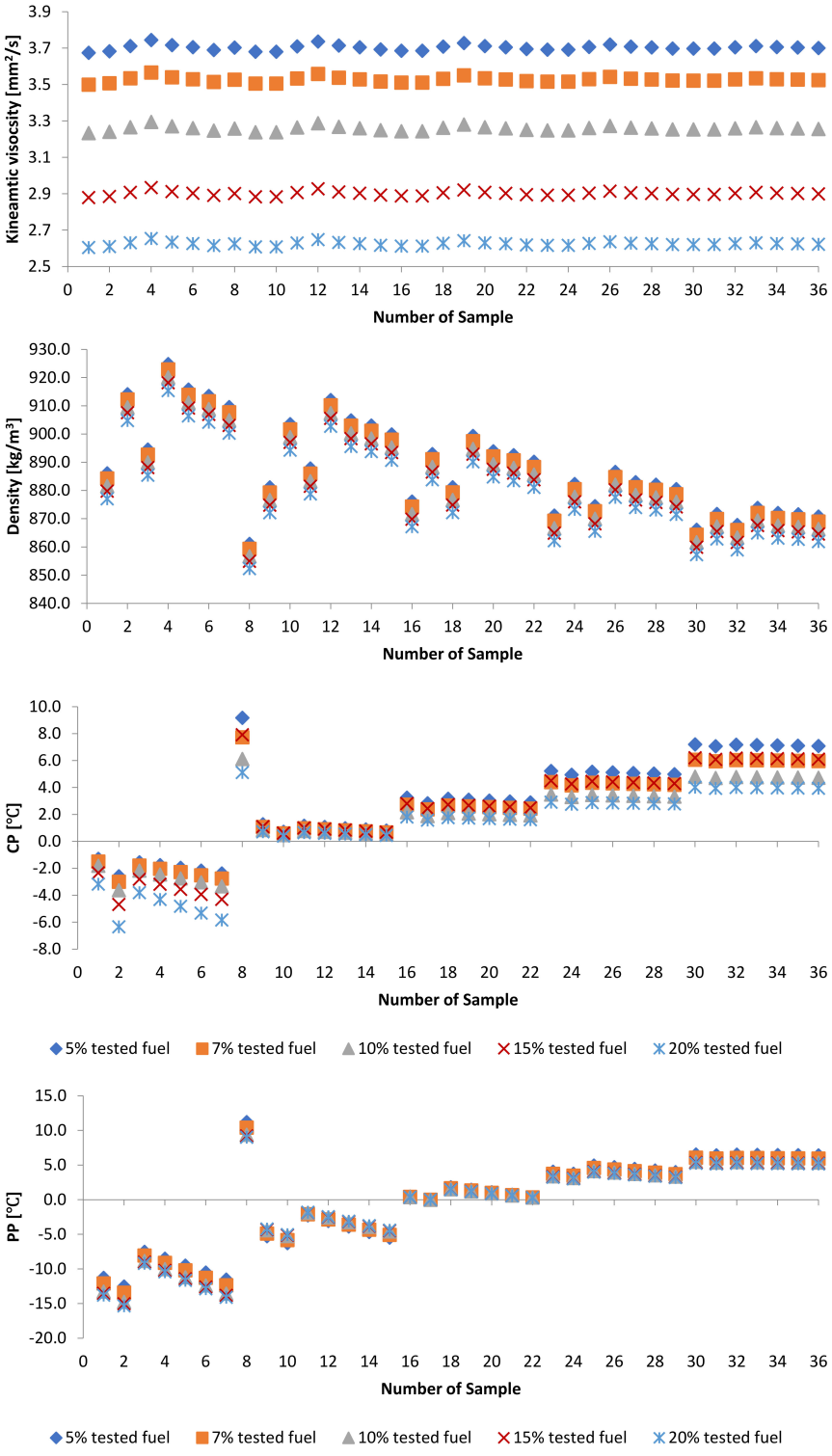

Figure 13. Properties of WFME-PME-KF with various volume ratios.

Table 10. List of samples used in this section.

Sample No.	Sample Name	Sample No.	Sample Name	Sample No.	Sample Name
1	WFME1	13	P20WFME25	25	P60WFME23
2	WFME12	14	P20WFME26	26	P60WFME24
3	WFME23	15	P20WFME27	27	P60WFME25
4	WFME24	16	P40WFME1	28	P60WFME26
5	WFME25	17	P40WFME12	29	P60WFME27
6	WFME26	18	P40WFME23	30	P80WFME1
7	WFME27	19	P40WFME24	31	P80WFME12
8	PME	20	P40WFME25	32	P80WFME23
9	P20WFME1	21	P40WFME26	33	P80WFME24
10	P20WFME12	22	P40WFME27	34	P80WFME25
11	P20WFME23	23	P60WFME1	35	P80WFME26
12	P20WFME24	24	P60WFME12	36	P80WFME27

Based on previous scientific studies, poor cold flow properties are the main problem associated with biodiesel derived from palm oil. The investigation of the experimental results (Figures 10–13) indicated that blending WFME-PM with low proportions of petroleum-based fuels could significantly improve the cold flow properties (CP and PP) as well as oxidation stability of WFME.

4. Conclusions

The objectives of the current study were to improve the low-temperature flow properties of biodiesel produced from palm oil and enhance the oxidation stability of biodiesel derived from waste frying/residential vegetable oils. The outcome of this work showed that adding palm biodiesel and WFME18, which are produced from cooking sunflower oil and corn oil, helped improve the stability of biodiesel, hence reducing degradation of the fuel. Additionally, the results indicated that the biodiesel produced from waste frying/residential canola oil could be utilized as a cold flow improver to enhance the low-temperature flow properties of PME and WFME samples. It can be concluded that an optimum mixture of PME with WFME will yield a significant improvement in the cold flow properties and oxidation stability.

Moreover, ULSDFW, ULSDFS, KF, and BF were used as cold flow improvers to improve the cold flow properties of biodiesel. Furthermore, they were utilized to reduce the kinematic viscosity and density of biodiesel samples for use as biodiesel in the transport sector that must fulfill standard requirements (ASTM D975 and EN 590). It was concluded that mixing WFME-PME with materials derived from crude oil (petroleum) reduced the kinematic viscosity and the density to acceptable levels specified in the Standard Specification for Diesel Fuel Oils (ASTM D975) and EN 590. Comparisons were made with commercially available petroleum-based fuels; the results showed that blending WFME-PME with a low proportion of petroleum-based fuels could significantly improve the cold flow properties (CP and PP) compared to PME-petroleum-based fuels.

Supplementary Materials: The following are available online at https://www.mdpi.com/article/10.3390/en14164928/s1, Table S1: Fatty acid compositions of fresh biodiesel samples, Table S2: Standard specifications for physicochemical properties of tested fuels.

Author Contributions: E.A. measured the physicochemical properties of the fuel. H.Ç. analyzed the data and wrote the paper. All authors have read and agreed to the published version of the manuscript.

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. The authors wish to express their sincere thanks to Youssef Kassem for his critical comments.

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

Symbols and Nomenclature

ASTM	American Standard Test Method
BF	Benzene
CFP	Cold flow properties
CP	Cloud point
D	Density
DU	Degree of unsaturation
GC	Gas chromatography
KF	Kerosene
KV	Kinematic viscosity
LCSF	Long-chain saturated factor
MUFAMEs	Monounsaturated
NaOH	Sodium hydroxide
OX	Oxidation stability
PME	Palm methyl ester
PP	Pour point
PUFAMEs	Polyunsaturated
SFAMEs	Saturated
ULSDFS	Ultra-low sulfur diesel fuel summer
ULSDFW	Ultra-low sulfur diesel fuel winter
WFME	Waste frying methyl ester

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