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Physicochemical Characterization and Butanol Impact on Canola and Waste Cooking Oil Biodiesels: A Comparative Analysis with Binary Biodiesel Blends

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In this study, the physicochemical properties of canola and waste cooking oil biodiesels, as well as various binary biodiesel blends, were investigated according to TS EN 14214 and ASTM D 6751 standards. Critical parameters such as density, kinematic viscosity, cold filter plugging point (CFPP), calorific value, flash point, copper strip corrosion, water content, and ester yield were evaluated. The findings highlighted the notable density of C_{100} and W_{100} biodiesels, with the addition of butanol reducing density. While viscosity values adhered to standards, the addition of butanol was observed to decrease viscosity. CFPP values indicated compliance with standards only for C_{100} and $C_{75}W_{25}$. Flash points of C_{100} and W_{100} biodiesels met standards, but the addition of butanol to binary biodiesel blends lowered flash points. Copper strip corrosion values were determined to comply with standards for all fuels. Calorific values demonstrated the prominence of C_{100} and W_{100} biodiesels, with the addition of butanol observed to decrease calorific values in binary biodiesels. While water content favored canola biodiesel over waste cooking oil biodiesel, the addition of butanol to binary biodiesels increased water content. Regarding ester yield, C_{100} biodiesel exhibited the highest yield, and the addition of butanol to binary biodiesels increased ester yield. In conclusion, this study thoroughly analyzed the physicochemical properties of biodiesel and blend fuels, revealing the impact of butanol addition on these properties.

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Introduction

In recent years, the interest in alternative and renewable energy sources has surged due to the escalating global population, the increasing energy demand, the dwindling reserves of fossil fuels, and the adverse effects of these fuels on human health and the environment, primarily through the rise in carbon dioxide levels in the atmosphere from combustion (Vaka et al., 2020; Zori et al., 2022). When examining Turkey's energy situation, it is observed that the country relies on external sources for approximately 70% of its total energy and is highly dependent on oil, accounting for about 90%. This unfavorable condition underscores the imperative for incentivizing domestic resources and expediting the diversification of energy sources (Oğuz et al. 2019). Studies on alternative fuels are gaining importance in order to reduce dependence on foreign energy. Biofuels, which are renewable and environmentally friendly, have garnered attention as potential alternatives to fossil fuels for internal combustion engines (Panwar et al., 2011). Biodiesel, a type of biofuel derived from various biological sources, has been identified as a viable alternative to diesel due to its comparable performance and its capacity to mitigate greenhouse gas emissions. The use of biofuels, including biodiesel, aligns with the global shift towards sustainable and environmentally friendly energy sources (Jeswani et al., 2020; Panwar et al., 2011).

Canola biodiesel is a first-generation biodiesel feedstock that has been widely studied for its potential in biodiesel production. Canola oil, derived from the seeds of the canola plant, is known for its high seed oil content, favorable fatty acid composition, and adaptation to temperate climates, making it a preferred feedstock for biodiesel production (Javed et al., 2014). Studies have shown that canola biodiesel has superior cold flow properties due to its high unsaturated and low saturated fat content, making it a favorable alternative to other biodiesel feedstocks (Haagenson and Wiesenborn, 2011). Additionally, research has demonstrated that canola biodiesel has the potential to substitute petroleum-based automotive lubricants, as it exhibits low cloud and pour point properties, better friction and antiwear properties, and lower viscosity (Sharma et al., 2015). Furthermore, the performance and emission characteristics of canola biodiesel in diesel engines have been found to be excellent, with higher brake thermal efficiency, lower brake-specific fuel consumption, and lower opacity compared to soybean biodiesel blends (Taib et al., 2018). In conclusion, canola biodiesel is a promising biodiesel feedstock due to its favorable properties, such as high seed oil content, superior cold flow properties, and excellent performance in diesel engines. Its potential as a sustainable and renewable alternative to petroleum-based fuels makes it a subject of extensive research and development in the field of biodiesel production.

Biodiesel production from waste frying oil has gained significant attention due to its potential to reduce raw material costs and address waste disposal issues. Waste frying oil, a byproduct of the food industry, is estimated to be about half the price of virgin oil, making it an economical and sustainable feedstock for biodiesel production (González and Reinares, 2005). Various studies have highlighted the potential of waste frying oil as a feedstock for biodiesel production, emphasizing its cost-effectiveness and its ability to replace nonrenewable energy sources (Meka and Asere, 2022). Additionally, waste frying oil has been found to be a suitable resource for the efficient conversion to biodiesel, with its use being prioritized over edible oils as a biodiesel feedstock (Chhetri et al., 2008).

However, the composition of waste frying oil, including its density, viscosity, total polar material, water content, acid value, iodine value, peroxide value, and heating content, can impact the yield and quality of biodiesel. High water and free fatty acid content in waste frying oil can lead to decreased biodiesel yield and quality due to adverse reactions during transesterification (Souissi et al., 2018). Additionally, the presence of heavy oligomeric admixtures in waste frying oil may pose challenges in achieving the necessary purity of the resulting biodiesel, requiring advanced purification methods (Şanlı et al., 2011; Zubenko et al., 2021).

In recent years, the growing global interest in alternative and sustainable energy sources has spurred extensive research into the utilization of biodiesel blends as a viable solution to address the escalating demand for cleaner fuels. This investigation draws on diverse studies to explore the impact of blending different biodiesel sources on fuel properties, engine performance, and emissions. Gupta and Sharma (2023) study scrutinized binary mixtures of Jatropha curcas biodiesel and waste cooking oil (WCO), revealing that an 80% WCO and 20% J. curcas blend exhibited optimal fuel properties when compared to conventional diesel. While this blend showcased a substantial reduction in carbon monoxide and unburned hydrocarbons emissions, it also exhibited heightened carbon dioxide and nitrogen oxide (NOx) emissions, emphasizing the intricate relationship between blend composition and emissions at full load conditions. Similarly, Singh et al. (2020) investigated engine and emission parameters using a blend of 70% amla seed oil biodiesel and 30% eucalyptus oil (AB70EU30). Their findings underscored the potential for enhanced Brake Thermal Efficiency (BTE) and reduced Brake Specific Fuel Consumption (BSFC) compared to traditional diesel, accompanied by an improvement in combustion characteristics. Nita et al. (2011) examination of poppy and waste cooking biodiesel-diesel blends highlighted significant reductions in CO, HC, and PM emissions, albeit with a concurrent increase in NOx emissions. Da Ponte et al. (2015) focused on second-generation biodiesel by blending castor oil with cotton, rapeseed, and soybean oils, showcasing the promise of alternative feedstocks with reduced reliance on edible raw materials. Nayak et al. (2021) study explored a binary mixture of fish oil and waste cooking biodiesel (WCB), revealing a superior quality fuel with decreased smoke, CO, and HC emissions. Additionally, Habibullah et al. (2015) demonstrated the advantages of combining palm and coconut biodiesel, showcasing reduced NOx emissions and improved brake power. Iqbal et al. (2015) experimentation with MATLAB-optimized blends of palm, coconut, and jatropha biodiesels resulted in improved overall fuel properties and engine performance. Bhuiya et al. (2017) investigation into poppy and waste cooking biodiesel-diesel blends underscored their potential to significantly reduce CO, HC, and PM emissions. Lastly, Ramuhaheli et al. (2022) study introduced a hybrid biodiesel blend from waste cooking oil and soya bean oil, showcasing improved engine performance and reduced emissions. This comprehensive review aims to distill insights from these diverse studies to offer a nuanced understanding of the implications of biodiesel blending on fuel characteristics and engine behavior.

In this research, biodiesel was produced from canola seeds and waste frying oil. Three different binary biodiesels were created by blending these biodiesels in volumetric ratios of 1:1 and 1:3. These biodiesels are $C_{25}W_{75}$ (25% canola biodiesel-75% waste frying biodiesel), $C_{50}W_{50}$ (50% canola biodiesel-50% waste frying biodiesel) and $C_{75}W_{25}$ (75% canola biodiesel-25% waste frying biodiesel). Additionally, three different blended fuels were obtained by adding 5%, 10% and 20% butanol to binary biodiesels in equal amounts by volume. These blended fuels are $C_{47.5}W_{47.5}B_5$ (47.5% canola biodiesel-47.5% waste frying biodiesel-5% butanol), $C_{45}W_{45}B_{10}$ (45% canola biodiesel-45% waste frying biodiesel-10% butanol) and $C_{40}W_{40}B_{20}$ (40% canola biodiesel-40% waste frying biodiesel-20% butanol). The study aimed to determine the physicochemical properties of these binary biodiesels and binary biodiesel-butanol fuels and to evaluate their compliance with relevant biodiesel standards (EN 14214, ASTM D-6751).

Material and Methods

The waste frying oil used in the research was obtained by using sunflower oil as domestic frying oil. Canola oil was obtained from the market. In addition, 99.5% n-butanol with a density of 0.80 g cm^{-3} at 20 ºC was used to be mixed into the binary biodiesels at certain ratios. Canola oil and waste frying oil were firstly filtered through filter paper and the following steps were carried out after filtration.

Biodiesel Production Stages

Filtration Process for Oils: Canola oil and waste frying oil undergo separate filtration processes to remove solid particles and other impurities from the raw oils.

Transesterification Method: The transesterification method is employed for the production of canola biodiesel (C_{100}) and waste frying biodiesel (W_{100}). Methyl alcohol

(CH_3OH) is used to separate glycerol components in the oils. Sodium hydroxide (NaOH) is used as a catalyst to break down triglycerides.

Preparation of Reaction Mixture: Approximately 20% of the filtered raw oil is mixed with methyl alcohol, and 3.5 g of sodium hydroxide is used for every liter. The mixture is stirred until methyl alcohol and sodium hydroxide are dissolved, resulting in methoxide.

Execution of the Reaction: Methoxide is combined with raw oil, and the mixture is stirred at the reaction temperature (55 ºC) controlled by a thermostat. The reaction temperature is maintained for one hour while stirring continues. An eight-hour waiting period allows the separation of methyl ester and glycerol in the oil, with glycerin being separated from methyl ester.

Reduction of Water Content and Drying: The separated methyl ester is heated to 75ºC, eliminating the remaining methyl alcohol. A washing process is conducted to remove unreacted alcohol, fatty acids, catalyst residues, and residual glycerol. The washing process employs approximately 20% pure water, and the mixture is left to settle for the precipitation of water and non-reacted substances. Wastewater is then separated from the raw biodiesel. The biodiesel is subjected to a drying process by heating it to 100ºC using a magnetic stirrer, eliminating the remaining water.

These stages encompass the processing of raw materials, execution of reactions, and purification of the final product in biodiesel production (Eryilmaz et al., 2022; Şahin and Mengeş, 2022).

The production steps described in the transesterification process were applied for both oils and C_{100} and W_{100} biodiesels were produced.

The % blend ratios of the fuels and blends prepared for the determination of fuel properties and the general appearance of these fuels are shown in Table 1 and Figure 1, respectively. For ease of use, canola biodiesel is symbolized as "C", waste cooking oil biodiesel as "W" and n-Butanol as "B". The numbers added as indices under the symbols represent the blend ratios of the fuels.

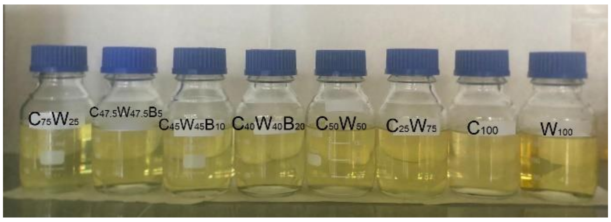

Figure 1. Prepared biodiesels and fuel blends

In the study, the fuel analysis laboratory and the measurement devices in the laboratory, which was established within the scope of the DPT 2004/7 project (Öğüt et al., 2004), were used to determine the physicochemical properties (kinematic viscosity, density, calorific value, water content, flash point, cold filter plugging point, copper strip corrosion, ester yields) of all fuels and blends.

The analyses of the fuels and blends were carried out according to the working methods of the devices used in the measurement (Table 2) and the results of the analyses were based on the EN 14214 European Union and ASTM D 6751 American standards applied for biodiesel today and their compliance with these standards was determined (Table 3).

Table 1. Prepared fuels and % blend ratios

Fuels	C (%)	W (%)	B (%)
C_{100}	100	-	-
W_{100}	-	100	-
$C_{25}W_{75}$	25	75	-
$C_{50}W_{50}$	50	50	-
$C_{75}W_{25}$	75	25	-
$C_{47.5}W_{47.5}B_5$	47.5	47.5	5
$C_{45}W_{45}B_{10}$	45	45	10
$C_{40}W_{40}B_{20}$	40	40	20

Table 2. Specifications and measurement standards of test equipment

Fuel Property	Devices	Measurement Range	Unit	Measurement Accuracy	Standard
Density	Kem Kyoto DA-130N	0.0000 - 2.0000	g cm^{-3}	±0.0001	EN ISO 3675 EN ISO 12185
Kinematic viscosity	Koehler K23377	Ambient temperature–150	ºC	±0.01	EN ISO 3104
Flash point	Koehler K16270	Ambient temperature - 370	ºC	±0.01	EN ISO 2719 EN ISO 3679
Water content	Kem Kyoto MKC-501	10µg-100mg	µg	±0.01	EN ISO 12937
Calorimeter	IKA C 200	0-40.000	J	±0.0001	DIN 51900
Cold filter plugging point	Tanaka AFP-102	With a coolant down to -60°C	°C	±0.01	ASTMD6379
Copper strip corrosion	Koehler K 25330	0-190	°C	±0.01	EN ISO 2160

Table 3. Biodiesel Standards

Property	Unit	European Standards		American Standards	
		EN 14214	Test Method	ASTM D 6751	Test Method
Density (at 15°C)	(g cm^{-3})	0.86-0.90	EN ISO 3675 EN ISO 12185	-	-
Kinematic Viscosity (at 40°C)	(mm^2 s^{-1})	3.5-5.0	EN ISO 3675 EN ISO 12185	1.9-6.0	D 445
Flash Point	(°C)	≥120	EN ISO 3104 ISO 3105	≥130	D93
Calorific Value	Mj/kg	≥38		37.27	D4809
Water Content	(mg kg^{-1})	≤500	ISO 3987	≤500	D 2709
Copper Strip Corrosion (3h/50°C)	Degree of corrosion	Class 1	EN ISO10370	≤3	D 130
Cold Filter Plugging Point	(°C)	<-10	EN 14107	-	-

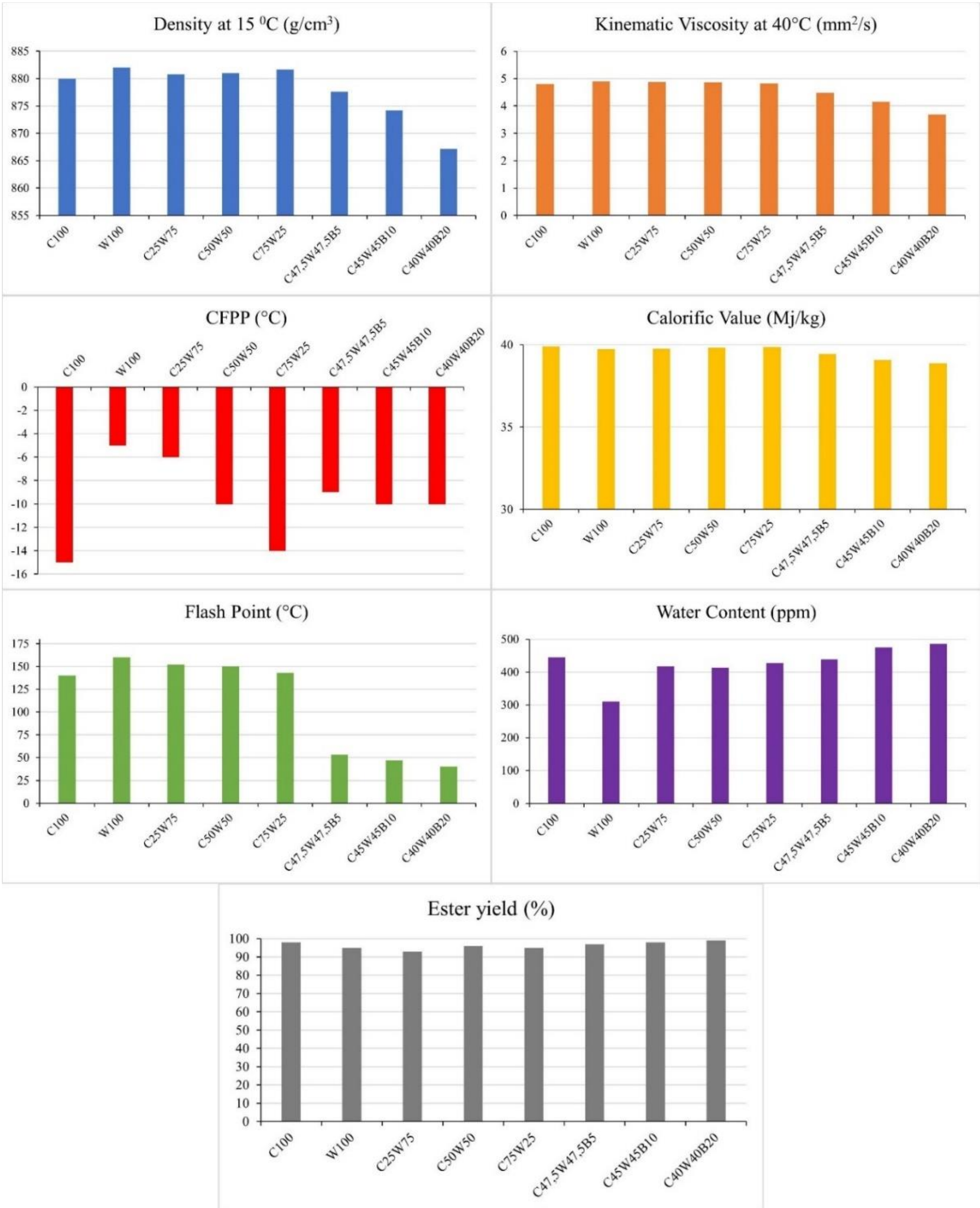

Figure 2. Fuel properties of canola biodiesel, waste cooking oil biodiesel, binary biodiesel and binary biodiesel + butanol blends

Results and Discussion

The values of some physicochemical properties for canola biodiesel, waste cooking oil biodiesel, and blended fuels are presented in Figure 2. The results have been compared with TS EN 14214 and ASTM D 6751 standards. The density values for C_{100} and W_{100} biodiesels are determined as 880 g cm^{-3} and 882 g cm^{-3}, respectively. In binary biodiesel blends, namely $C_{25}W_{75}$, $C_{50}W_{50}$, and $C_{75}W_{25}$, the density values are very close to each other. The addition of butanol to binary biodiesel blends has decreased the density values of the blended fuels. Previous studies have also indicated that the addition of alcohol reduces the density values of binary biodiesel blends (Ramuhaheli et al., 2022). It is observed that all biodiesel and blended fuels conform to the standards.

The kinematic viscosity values for all biodiesels, binary biodiesel blends, and ternary fuel blends with added butanol in this study have complied with the standards of EN 14214 and ASTM D 6751. Among the fuels, the highest viscosity value was obtained in C_{100} and W_{100} biodiesels. The viscosity values for these biodiesels were measured as 4.8 mm^2 s^{-1} and 4.9 mm^2 s^{-1}, respectively. Previous research studies have consistently found high viscosity values for canola and waste cooking oil biodiesels (Babu et al., 2020; Öztürk, 2015; Roy et al., 2013). The volumetric increase of butanol in binary biodiesel blends has resulted in a reduction in viscosity values. These reductions have varied between 6% and 23%, correlating with the increase in butanol. Similar results have been reported in the studies of Huang et al., (2020); Ramuhaheli et al. (2022).

The Cold Filter Plugging Point (CFPP) determines the temperature at which wax crystals begin to form in the fuel, potentially causing engine malfunctions by blocking fuel lines and filters (Sirviö et al., 2019). It has been observed that only the fuels C_{100} and $C_{75}W_{25}$ comply with the EN14214 standards regarding CFPP values. In this study, the CFPP values for C_{100} and W_{100} biodiesels were found to be -15 °C and -5 °C, respectively. The increase in the volumetric ratio of canola in binary biodiesel blends has reduced the CFPP value. The addition of butanol to binary biodiesel fuels has shown very little change in CFPP values.

The calorific value is a crucial feature in fuel selection (Atabani and da Silva César, 2014) and is one of the most significant characteristics to characterize a fuel (Ong et al., 2013; Silitonga et al., 2013). It has been determined that all biodiesel, binary biodiesel blends, and ternary fuel blends comply with EN14214 standards. The calorific values for C_{100} and W_{100} biodiesels are measured as 39.9 Mj kg^{-1} and 39.73 Mj kg^{-1}, respectively. The calorific value of binary biodiesel blends prepared from these biodiesels falls within this range. However, the addition of butanol to binary biodiesels has decreased the calorific value. The increase in oxygen content with the addition of butanol has resulted in a decrease in the calorific values of the fuels. Similar results have been reported in the studies of Lebedevas et al., (2010); Örs et al., (2019).

It has been observed that the flash points of C_{100}, W_{100}, and binary biodiesel blends comply with EN 14214 and ASTM D6751 standards. The flash point of biodiesel not only influences its volatility but also a higher flash point reduces the risk of fire, making the transportation and storage processes of biodiesel safer compared to diesel fuels (Odega et al., 2021). The addition of butanol to binary biodiesel blends has lowered their flash points, falling outside the standard range. As the ratio of butanol in binary biodiesel blends increases, the flash point has decreased between 64% and 73%. This can be attributed to the fact that the flash point value of pure butanol is 34 °C. Ramuhaheli et al. (2022) reported a 40% decrease in flash point by adding 15% ethanol to a binary biodiesel blend in their study. Researchers have indicated that the addition of alcohol to fuel blends lowers the flash point (Álvarez et al., 2019; Örs et al., 2019).

Copper strip corrosion values have been determined as 1a for all fuels, showing similarity according to Ciubota-Rosie et al., (2013); Eryilmaz et al. (2022).

The water content in the biodiesel matrix is a critical parameter that significantly affects the quality and performance of biodiesel. Excessive water content can lead to a decrease in the oxidation stability of biodiesel, a shortened shelf life, and a potential for microbial growth; contributing to the clogging of fuel filters and corrosion in the fuel distribution system (Jalil et al., 2022). The water content in canola biodiesel is found to be 30% higher than in waste cooking oil biodiesel. Biodiesel derived from waste cooking oil is obtained from previously used cooking oil that has undergone a cooking process. In the course of this process, the water present in the oil is evaporated, leading to a reduced water content in the waste cooking oil (Singh et al., 2022). All biodiesels, binary biodiesel blends, and ternary fuel blends have complied with the standards. The addition of butanol to binary biodiesel blends has increased the water content. Similar results have been reported by Yeşilyurt et al. (2018) in their studies.

The term "ester yield" in biodiesel fuels refers to the percentage of fatty acid methyl esters (FAME) obtained from the transesterification process of vegetable oils or animal fats. This yield is a crucial factor in biodiesel production as it directly impacts the efficiency and effectiveness of the fuel. The highest ester yield within biodiesels was measured at 98% in C_{100}. The lower ester yield of W_{100} compared to C_{100} has reduced the ester yield of binary biodiesel blends. However, the volumetric increase in the ratio of butanol in binary biodiesels has increased the ester yield.

Conclusion and Recommendations

In this study, the fuel quality and relationships of biodiesel produced from canola raw oil (C_{100}), biodiesel produced from waste frying oil (W_{100}), binary biodiesel blends ($C_{25}W_{75}$, $C_{50}W_{50}$, and $C_{75}W_{25}$), and ternary fuel blends with added butanol ($C_{47.5}W_{47.5}B_5$, $C_{45}W_{45}B_{10}$, and $C_{40}W_{40}B_{20}$) were investigated.

Physicochemical properties of all fuel samples (C_{100}, W_{100}, $C_{25}W_{75}$, $C_{50}W_{50}$, $C_{75}W_{25}$, $C_{47.5}W_{47.5}B_5$, $C_{45}W_{45}B_{10}$, $C_{40}W_{40}B_{20}$) respectively; density (g cm^{-3}) (15 °C) 880; 882; 880.8; 881; 881.6; 877.6; 874.2 and 867.2, viscosity kinematics (mm^2 s^{-1}) (40 °C) 4.8; 4.9; 4.88; 4.86; 4.82; 4.48; 4.15; 3.69, flash point (°C) 140; 160; 152; 150; 143; 53; 47; 40, calorie value (Mj kg^{-1}) 39.9; 39.73; 39.76;

39.82; 39.87; 39.45; 39.09; 38.89, cold filter clogging point (°C) -15; -5; -6; -10; -14; -9; -10; -10, water content (ppm) 445; 310; 417; 414; 427; 439; 475; 486, ester yield (%) 98; 95; 93; 96; 95; 97; 98; 99, copper strip corrosion (3h 50°C) determined as 1a in all fuels.

The addition of butanol to binary biodiesels can be employed to improve high values, such as viscosity; however, caution should be exercised due to the reduction in flash point caused by the addition of butanol, especially in terms of transportation and storage. Optimized studies can be conducted to determine the interactions of various factors in the biodiesel production process and identify optimal conditions. These studies can be considered significant steps towards enhancing efficiency and reducing costs in biofuel production. The current research has presented some fundamental characteristics of biodiesel and ternary fuel blends; however, a more detailed analysis of the emission profiles of these fuels is possible. Such an analysis contributes to understanding environmental impacts and identifying the advantages of transitioning to low-carbon energy sources.

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