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Evaluation of key fuel properties of three generation biodiesel fuels: an experimental investigation of feedstock type

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ABSTRACT

Transportation sector is one of the most important causes of environmental problems such as global warming and acid rain. To reduce transportation's negative impacts on the environment, it should be made carbon neutral. Although electrification has been very popular in recent years, internal combustion engines will continue to dominate transportation for a long time. Biodiesel can be produced from varios feedstocks and is classified into three generation according to its feedstock origin. However, the fuel properties of biodiesel fuels of different generations vary significantly depending on feedstock. Biodiesel fuels' physico-chemical fuel properties greatly influence the engine characteristics and exhaust emissions. In this experimental study, 13 different biodiesel fuels' (including three generations) some key fuel properties such as kinematic viscosity, density, flash point, cold filter plugging point and cetane index were determined and compared with each other. The highest and the lowest kinematic viscosities were measured for Waste Cooking Oil Biodiesel and Soybean Oil Biodiesel, respectively. Among the biodiesel fuels, only Waste Cooking Oil Biodiesel and Waste Chicken Fat Biodiesel could not meet the viscosity specification in EN 14214. The density values of test fuels were very similar (between 875.83 $kg.m^{-3}$ and 891.46 $kg.m^{-3}$) and all were within the required specification range. The lowest flash point (142 °C) was measured for Algae Oil Biodiesel. It was considerably lower than other fuels. The highest flash point (184 °C) belonged to Hazelnut Oil Biodiesel. Waste Fleshing Oil Biodiesel and Waste Cooking Oil Biodiesel had the highest (58.80) and lowest (50.54) cetane values, respectively. However, all biodiesels met the minimum cetane value given in European biodiesel standard. The most significant differences (ranged from -10 °C and 10 °C) between the fuel properties of biodiesels of different origins were observed in CFPP. The viscosity and poor cold flow properties of waste feedstock-based biodiesels may cause critical problems in diesel engines. Nevertheless, they can be blended with other biodiesels or petro-diesel in certain amounts. Among the biodiesel fuels of different origins tested in this study, algae oil biodiesel has the best physico-chemical fuel properties and technical potential.

Keywords: Transportation, carbon neutrality, biodiesel, fuel properties, feedstock.

1. Introduction

Energy can be defined as the ability to do work in its most fundamental meaning. Nevertheless, energy is the most basic input for many different sectors and a critical requirement for sustainable development, and it is as important as life itself in today's conditions. Because of the rapid increase in the world population, rising living standards, growing industrial production and logistics needs, world energy consumption is constantly increasing. By the end of 2035, total world energy consumption is estimated to be 779 EJ [1]. Despite important academic and industrial scale studies on many different alternative energy sources, most of the world's energy needs are still met by fossil energy sources and petroleum is still the primary energy source. However, fossil energy resources are generally obtained from certain regions of the world and countries like Türkiye, which do not have sufficient fossil resources, have to spend large amounts of money for energy imports. When today's world is thought geo-politically and geo-strategically, considering the effects of energy dependency in terms of only economically is insufficient. Countries exporting fossil energy sources may use these advantages as a means of blackmail in international relations. In addition to the economic and geopolitical effects of increasing fossil energy consumption, its environmental impact should also be strongly highlighted. Intensive energy usage causes vital environmental problems including global warming, acid rain, etc. These environmental problems are getting worse every year and cause consequences that directly affect human life, such as climate change and decreasing water resources.

Energy consumption is mainly driven by three sectors: industry, households and transportation. In spite of many economic and geopolitical uncertainties in the world, energy demand of the transport sector has been increasing each year, accounting for about 30% of all energy consumed globally. To satisfy this energy need of transportation is crucial for the continuity of the people's mobility and the global supply chain. As an inevitable result of this huge energy consumption, the transport sector accounts for more than a third of CO_2 emissions from end-use sectors and is one of the biggest GHG emissions reasons [2]. According to Net Zero Emissions by 2030 Scenario, CO_2 emissions from transport must be decreased by about a quarter, even though transport demand continues to increase. To achieve this target (sustainable and environmentally friendly transportation), policies need to encourage shifting to less carbon-intensive transport options. Nowadays, electrification of transportation is a very popular issue, and electric vehicles are generally considered as the key technology for the decarbonization of transport sector, especially road transport that accounts for about one-sixth of global emissions. Nevertheless, there are many technical problems and economic uncertainties that should be solved for the widespread electrification of transportation. Because of these reasons, electric vehicles are not yet a global phenomenon in the transport sector. For example, the share of electric cars (battery electric vehicles and plug-in hybrid vehicles combined) in total car sales was about 14% in 2022 (about 10 million worldwide) [3]. One of the most important issues to be emphasized in terms of electrification of transportation is that the electrical energy, which is used to charge the vehicle battery, should be obtained from domestic, renewable, sustainable and environmentally friendly sources. The production of electricity used in battery charging from imported fossil resources by polluting the environment will have a positive effect on reducing imported energy dependency nor on environmental pollution. Considering IEA report stating that only 29% of electricity used for transport sector in 2023 was obtained from renewable sources [4], it can be said that electromobility powered by renewable sources will not be able to solve this problem on its own. It should be strongly stressed that the electrification of transport sectors other than road transport (such as aviation, maritime, rail and even medium- and heavy-duty road vehicles) is very difficult and in some cases impossible. Thus, a very big portion of transportation depends substantially on internal combustion engines running generally on fossil fuels. Among internal combustion engines, diesel engines have higher torque and better thermal efficiency superiorities over gasoline engines. Thus, in comparison to gasoline engine vehicles, diesel engine vehicles have been dominantly used in all transportation sectors. Apart from transport, diesel engines are also used extensively in many different areas such as construction machinery and generators. Because of this situation, diesel fuel consumption is much higher than that of gasoline.

Taking into consideration these conditions, the importance of renewable, environmentally friendly, sustainable and domestically producible alternative diesel fuel is clearly seen. Biodiesel, which is an alternative diesel fuel, can be utilized in all areas of transport sector including railway, seaway and airway. It is compatible with the existing fuel distribution and station network and does not require significant modification in the engine and fuel injection system of the vehicle [5]. When considered on an industrial scale, factory and infrastructure investment can be realized with smaller budgets compared to other alternative energy types. Since biodiesel fuels can be produced from domestic feedstocks, it can alleviate the dependence on the import energy sources causing account deficit problems. Moreover, biodiesel fuels of all generations, regardless of its feedstock type, have better exhaust emission profile (apart from NO_x) than petro-diesel [6-8]. At finally yet importantly, biodiesel fuel contains carbon absorbed by plants through photosynthesis. When this biodiesel fuel is used in a diesel engine, the carbon released during combustion returns to the atmosphere. Therefore, biodiesel can be accepted as a near zero-emission fuel, and it can be stated that biodiesel fuel will not increase the carbon footprint of the transportation sector [9].

Powering a diesel engine with biodiesel is not a new concept. Considering that Rudolph Diesel, the inventor of the diesel engine, powered his engine on peanut oil, it can be said that the history of biodiesel as a diesel engine fuel is as old as the diesel engine itself. Although powering a diesel engine with crude vegetable oils gave positive results in the short term, serious engine problems were encountered when the operating times were prolonged [10]. The most important reason for these vital engine failures is the unacceptably high viscosity of vegetable oils. The viscosity values of vegetable oils are approximately 15 times higher than that of petro-diesel. This unacceptably high viscosity negatively affects the fuel injection process, resulting in incomplete combustion [11]. In addition, glycerin in the chemical structure of vegetable oil causes the injector needle to stick to the injector nozzle. Coking occurs in the injector and the injector nozzle becomes clogged over time. Cooked and clogged injector nozzles lead to poor atomization quality [12]. Especially during cold starts, unburned fuel adheres to the cylinder walls and as it flows under the cylinder wall, it washes the lubricating oil film here and breaks the lubricating film. As a result of dry friction on the cylinder walls, scratches and abrasions occur on the cylinders over time [13]. Furthermore, unburned fuel goes down to the crankcase and mixes with the lubrication oil and deteriorates the chemical structure of the lubricant. Due to the deterioration of the lubricant quality, the above-mentioned serious engine failures occur in a very short time [14].

In order to avoid the aforementioned serious engine failures, the unacceptably high viscosities of vegetable oils should be decreased to a value close to that of petro-diesel. This viscosity reduction is achieved through the transesterification reaction. As shown in Figure 1, in the transesterification reaction, the triglyceride molecule, which is the main constituent of vegetable oils and animal fats, is chemically broken down by reacting with alcohol in the presence of a catalyst and its glycerin is replaced by the alkyl radical of the alcohol used in the reaction [15]. In a transesterification reaction, three moles of alcohol are theoretically used for one mole of feedstock, but since this is an equilibrium reaction, more alcohol should be used to force the reaction to the products side. The transesterification reaction is also an alcoholysis reaction, and any alcohol can be used in this reaction. However, for both technical and economic reasons, methanol is predominantly used [16]. Therefore, biodiesel is commonly defined as methyl ester (as seen in many fuel standards). Although alkaline, acid or enzyme catalysts can be used to catalyze the reaction, alkaline catalysts (mostly hydroxide catalysts such as KOH or NaOH) are generally preferred as they are much faster than other catalysts [17]. However, when alcohols other than methanol (such as ethanol) are used, better results can be obtained with alkoxide catalysts such as CH_3ONa, CH_3OK

instead of hydroxide catalysts [18]. It should be strongly emphasized that if the free fatty acid (FFA) content of the feedstock used in the transesterification reaction is above 1%, the alkaline catalyst causes saponification problem. In this case, the FFA content of the feedstock can be reduced to below 1% by the acid-catalyzed pre-treatment reaction/s and then the alkaline-catalyzed transesterification reaction (main reaction) can be carried out [19].

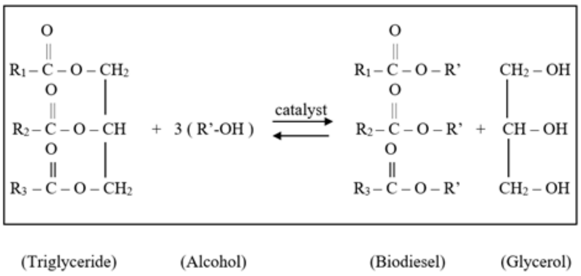

Figure 1. Stoichiometric transesterification reaction

When the chemical structure of any biodiesel fuel is analyzed, it will be seen that it consists of the fatty acid moiety coming from the feedstock from which it is produced and the alkyl radical moiety coming from the alcohol used in transesterification. Although alcohol has effects on the fuel properties of biodiesel, the main determining factor at this point is the biodiesel feedstock [20]. In a transesterification reaction, the fatty acid distribution remains essentially the same and shows itself in the biodiesel fuel obtained. In other words, the fatty acid composition of the biodiesel fuel reflects the fatty acid structure of the feedstock from which it is produced. Because of this, the fuel properties of different origin biodiesels show significant differences. For example, as the fatty acid chain length and saturation level increases, properties such as cetane number, heating value, lubricity and oxidative stability improve, while viscosity and cold flow properties are adversely affected [21-23].

Biodiesel can be obtained from many different feedstocks. Biodiesel fuels produced from conventional edible vegetable oils are classified as the first-generation biodiesel. If a worldwide investigation is conducted, it will be seen that edible vegetable oils including soybean oil, rapeseed oil, palm oil, etc. are the main feedstocks in the global biodiesel production industry. However, since approximately 75-80% of the total cost of biodiesel fuel is the feedstock price, the unit price of first-generation biodiesel fuel produced from food-grade high quality, but expensive feedstocks are quite high in comparison to petro-diesel [24]. Due to the unacceptably high break-even price of first-generation biodiesel fuels, their marketing is so difficult and consequently the biodiesel industry needs economic incentives, tax reductions, and subsidies. It can be said that first-generation biodiesel industry is not affordable and sustainable.

Waste frying oils, waste animal fats and inedible oils can also be used as feedstock in biodiesel production. Waste feedstock or inedible oil origin biodiesel fuel is classified as the second-generation biodiesel fuel. The usage of waste frying oils and fats as feedstock in transesterification may be beneficial for a sustainable biodiesel industry by alleviating the high-cost problem of biodiesel. Furthermore, by using the waste frying oils and waste animal fats as biodiesel feedstock, the probable environmental problems caused by the disposal of the waste materials may be prevented and also these waste feedstocks will be transformed into a very important and strategic product such as energy. However, this important point should be stressed that FFA content of second-generation feedstocks may be quite high. Thus, alkaline-catalysed transesterification may lead to saponification problem. This can hinder the biodiesel production process and result in significantly lower product yields [25].

Another biodiesel feedstock that is more recent than other feedstocks is algae. A biodiesel fuel obtained from algae oil is defined as the third-generation biodiesel. Compared to other biodiesel feedstocks, algae have many advantages such as not requiring soil for algae production, can be grown in a very short time and can be produced even in wastewater. There are many different algae species and the literature in this field is getting richer every year. However, studies on changing the fatty acid composition of algae according to the fuel properties of the biodiesel to be produced are quite remarkable [26]. For example, in order to produce algal oil-derived biodiesel, which can be mixed with jet fuel at certain ratios in the aviation sector, the algae feedstock should consist of fatty acids with low freezing point. However, while modifying the genetics of the fatty acid structure of the algae feedstock for this purpose, other critical fuel properties such as kinematic viscosity, calorific value, volatility, etc. should not be adversely affected by this intervention.

In the literature, there are various studies comparing the fuel properties of biodiesel fuels produced from different feedstocks. However, when these studies are examined, it is seen that either the test fuels do not cover all three biodiesel generations or most of the fuel properties are quoted from other studies. When the transesterification reaction parameters and conditions change, the fuel properties of the obtained biodiesel will also change. When the related literature is studied from this perspective, it will be realized that the number of studies comparing the critical fuel properties of biodiesels produced from different feedstocks under the same transesterification reaction conditions, covering all three generations, is quite limited. This experimental study aims to fill this gap in literature partially.

2. Experimental

The physico-chemical fuel properties of different generation biodiesels produced from various feedstocks will also be different. This will significantly affect combustion efficiency, engine performance characteristics, exhaust emissions and engine life. For this reason, in the current study, biodiesel fuels were produced from different feedstocks (including 3 generations) to determine some key fuel properties of biodiesels of different generations. Sunflower oil, hazelnut oil, rapeseed oil, corn oil, soybean oil, olive oil, safflower oil, palm oil as the first-generation feedstocks. Waste frying oil, waste chicken fat, waste fleshing oil and cottonseed oil as second-generation feedstocks. Algae oil (*chlorella protothecoides*) is the third-generation feedstock.

In the production of biodiesel fuels, transesterification reaction conditions were selected as 6:1 molar ratio of methanol to feedstock, 1% KOH, 1 hour, 60 °C. At the end of the reaction, the mixture in the reaction vessel was taken into the separation funnel and waited overnight for a complete phase separation. After draining the glycerol, the biodiesel fuel was washed 3 times with distilled water at 50 °C and then heated at 101 °C for one hour to dehydration.

Biodiesel fuels are coded as follows: sunflower oil biodiesel (SoB), hazelnut oil biodiesel (HoB), rapeseed oil biodiesel (RoB), corn oil biodiesel (CoB), olive oil biodiesel (OoB), safflower oil biodiesel (SfB), palm oil biodiesel (PoB), soybean oil biodiesel (SyB), cottonseed oil biodiesel (CtB), waste cooking oil biodiesel (WOB), waste chicken fat biodiesel (WCB), waste fleshing oil biodiesel (WFB), algea oil biodiesel (AlB). In order to symbolize the determined fuel properties FP, CFPP, KV, D, and CI stands for flash point, cold filter plugging point, kinematic viscosity, density and cetane index, respectively.

Table 1 shows the measured properties, equipment, and methods used in this study.

Property	Equipment	Method	Accuracy
Density	DMA 38	EN 12185	0.1
Kinematic Viscosity	HVM 472	EN 3104	0.001
Flash Point	HFP 339	EN 3679	0.5
Cold Filter Plugging Point	FPP 5Gs	EN 116	0.1
Distillation Temperatures	Herzog	EN 3405	0.1

In order to calculate the CI values of biodiesel fuels, ASTM D4737-21 method was used:

$$CI = 45.2 + (0.0892)(T_{10N}) + [0.131 + (0.901)(B)][T_{50N}] + [0.0523 - (0.420)(B)][T_{90N}] + [0.00049][(T_{10N})^2 - (T_{90N})^2] + (107)(B) + (60)(B)^2$$

where:

CI= Cetane Index, D= Density at 15 °C, DN= D-0.85, B= $[e^{(-3.5)(DN)}]-1$, T_{10}= 10% recovery temperature (°C), T_{10N}= T_{10}-215, T_{50}= 50% recovery temperature (°C), T_{50N}= T_{50}-260, T_{90}= 90% recovery temperature (°C), T_{90N}= T_{90}-310.

The determination of distillation temperatures of test fuels can be seen in Figure 2.

3. Results and discussion

The physico-chemical properties of the biodiesel fuels were determined through a series of standardized tests and measurements.

The results obtained for these fuel properties are presented in Table 2 for reference and comparison.

Figure 2. Determination of distillation temperatures of test fuels

3.1. Flash point

Flash point can be described as the minimum temperature at which fuel emits vapor in sufficient concentration to form an ignitable mixture with air near its surface. In other words, the lower the flash point, the easier it is to ignite a fuel [28]. It is generally evaluated in terms of transportation and storage safety; however, flash point is an important fuel property for the operability of a diesel engine. Namely, flash point of a fuel can be useful for providing insight into that fuel's volatility and flammability both of which directly influence easy start in cold weather, combustion and exhaust emissions. A diesel fuel with high viscosity and flash point may cause difficulties in easy cold-start and also ignition delay time. Moreover, in the literature, there are some studies reporting that high flash points may result in soot formation inside the engine cylinders [29].

Among the fuel properties defined in EN 14214 and ASTM D6751, the only property that has the same specifications is the flash point. Both biodiesel standards require a flash point of minimum 101 °C. This flash point value is considerably higher than that of petro-diesel. The flash point specification required for petro-diesel is min 55 °C in EN 590, while min 52 °C in ASTM D975. This significant difference between the flash points of petro-diesel and biodiesel fuels shows that the logistics and storage of biodiesel are safer than diesel fuel. This is one of the important superiorities of biodiesel over diesel fuel.

As can be seen from Table 2, the FP values of all biodiesel fuels were well above the limit value of 101 °C. The highest (184 °C) and lowest (142 °C) FP values were measured for HoB and AlB fuels, respectively. The most remarkable result here is that there is a significant difference of 26 °C between the FP value of AlB, which has the lowest temperature, and the FP value of WFB (168 °C), which has the second lowest temperature. Similar situation was not observed among biodiesel fuels with high FP values. The difference between the highest FP value (184 °C, HoB) and the second highest FP value (182 °C, OoB) is only 2 °C. The FB values of WCB and WFB, which were biodiesel fuels of waste animal fat origin, were almost the same (169 °C and 168 °C, respectively).

Table 2. Some key fuel properties of biodiesel fuels

Biodiesel	FP (°C)	CFPP (°C)	KV ($mm^2.s^{-1}$, 40 °C)	D ($kg.m^{-3}$, 15 °C)	CI
AlB	142	-10	4.49	880.25	55.90
WOB	180	9	6.65	891.31	50.54
WCB*	169	3	5.30	889.70	52.30
WFB*	168	10	4.70	876.70	58.80
CtB	178	3	4.36	884.36	55.82
HoB	184	-10	4.77	879.94	57.70
RoB	180	-10	4.84	885.24	55.83
CoB	176	-9	4.32	879.52	55.48
OoB	182	-6	4.84	890.48	57.09
SfB	174	-9	4.13	887.85	53.74
PoB	172	8	4.55	885.15	56.40
SyB	170	-9	4.11	875.83	51.50
SoB	176	-6	4.45	891.46	51.39

3.2. Cold filter plugging point

Among the physico-chemical properties of a fuel, cold flow properties are the most important features that affect the ability of that fuel to be used without any problems in regions with different weather conditions. A vehicle fuelled in a hot region can move to a region with much lower temperatures during the day and the engine must continue to run smoothly. Although biodiesel is generally used in road transport, projects are being developed for the use of biodiesel fuels in air transportation by blending with jet fuel in certain proportions. This is very important in terms of the usability of biodiesel in all transportation sectors. At this point, the cold flow properties of biodiesel fuels are very critical. The fatty acid distribution of a biodiesel fuel has a significant effect on its cold flow properties. As the fatty acid chain length and saturated fatty acid content increase, the cold flow properties deteriorate. In addition, the alcohol type used in transesterification reaction also affects the cold flow quality. For example, in comparison to methanol, when branched-chain alcohols such as 2-propanol are used, the cold flow properties improve, but when alcohols such as ethanol, n-propanol, n-butanol are used, the fuel begins to freeze at relatively higher temperatures [30].

In general, three concepts (cloud point, pour point, and cold filter plugging point) are used to characterize the cold-flow quality of a liquid fuel. The cloud point is the temperature at which the crystal cloud (wax crystals), which is an indicator of the onset of freezing, first appear on the fuel surface. As the temperature drops, crystallization increases, and consequently the fluidity of the fuel decreases. The pour point is defined as the lowest temperature at which fuel can maintain its fluidity and still be pumped. However, the fuel pumped by the fuel pump can continue to flow in the fuel line, but as a result of gel formation, it cannot pass through the small pores of the fuel filter and clogs the filter, causing the engine to stop. Therefore, pour point may be misleading in order to assess the usability of an engine fuel in cold weather. The cold filter plugging point (CFPP) is the temperature at which the pumped fuel cannot pass through the fuel filter as a result of crystallization, and it is more accurate to use the CFPP value for internal combustion engines [31].

There is no specification for CFPP value in EN 14214 and ASTM D6751. A similar situation is also seen in petro-diesel standards. While ASTM D975 does not specify a CFPP limit, EN 590 has a limit of -15 °C for winter and 5 °C for summer. As can be seen from Table 2, there were significant differences between the CFPP temperatures of biodiesel fuels. The difference between the highest and lowest CFPP values was 20 °C. The lowest CFPP temperature (-10 °C) was measured for RoB, HoB and AlB fuels. The CFPP value (-9 °C) of CoB, SyB and SfB is quite low and ranked second. The CFPP temperatures of SoB and OoB (-6 °C) were acceptable and the last temperature value below zero. It is noteworthy that the CFPP values (3 °C) of WCB, which is of animal fat origin, and CtB, which is of vegetable oil origin (both second generation biodiesels) were the same. WFB had the highest CFPP (10 °C) value among 13 different biodiesel fuels tested. CFPP temperatures of WOB (9 °C) and PoB (8 °C) were also too high and close to that of WFB. The high CFPP results may be caused by these biodiesels' high saturation level.

3.3. Kinematic viscosity

Viscosity, which can be defined as the resistance to flow, is one of the most important physical fuel properties. It should be noted that the main objective of the transesterification reaction is to bring the very high viscosity of triglycerides closer to that of petro-diesel. As the viscosity of the fuel increases, the atomisation quality decreases. Injection of a high viscosity fuel lead to large diameter droplets from the injector nozzle. Since it will take a long time for this relatively large-diameter fuel droplet to evaporate in the cylinder and mix with air, the formation of air-fuel mixture will be insufficient. This will adversely affect combustion efficiency, engine performance and exhaust emissions. In addition, since today’s electronically controlled modern fuel injection systems are extremely sensitive to fuel quality, the use of high viscosity fuels may cause fuel pump, injector and engine failures. Also, higher PM emissions resulted by the incomplete combustion in the engine cylinder may clog the diesel particulate filter of the vehicle in a short time [32].

While the kinematic viscosity specification in EN 14214 is between 3.5-5.0 $mm^2.s^{-1}$, this value is 3.5-6.0 $mm^2.s^{-1}$ in ASTM D6751. The difference of 1.00 $mm^2.s^{-1}$ between the upper viscosity limit of these two biodiesel standards can be critical especially for biodiesel fuels produced from relatively higher saturated feedstocks such as waste animal fats and waste frying oils. It may be useful to remember that the viscosity specification in EN 590 is 2.0-4.5 $mm^2.s^{-1}$, while in the ASTM D975 this range is 1.9-4.1 $mm^2.s^{-1}$. In other words, the upper limit of biodiesel viscosity is higher than that of petro-diesel in both EU and US standards.

Table 2 shows that the viscosity values of 11 biodiesel fuels (except WCB and WOB fuels) were lower than 5 $mm^2.s^{-1}$ and met the EN 14214 standard. The viscosity value of WCB (5.30 $mm^2.s^{-1}$) exceeded the upper limit of the EU biodiesel standard but it met the ASTM D6751. Nevertheless, the viscosity of WOB (6.65 $mm^2.s^{-1}$) exceeded even the upper limit of the ASTM D6751 standard. WOB processed in this study was obtained from a fast-food restaurant and it was palm oil origin. The high saturation level of palm oil and also high molecular weight degradation products (polar materials) resulted by frying process may be possible reasons of this high kinematic viscosity value of WOB.

Another remarkable point in the kinematic viscosity results is that the viscosity value of WFB (4.70 $mm^2.s^{-1}$), a biodiesel of animal fat origin, is lower than the viscosity values of ROB (4.84 $mm^2.s^{-1}$), OoB (4.84 $mm^2.s^{-1}$) and HoB (4.77 $mm^2.s^{-1}$). The viscosity values of vegetable oil based biodiesel fuels varied between 4.11 $mm^2.s^{-1}$ (SyB)-4.84 $mm^2.s^{-1}$ and all of them met the EN 14214 standard. As a third generation biodiesel, the viscosity of AlB (4.49 $mm^2.s^{-1}$) was very close to the first and second generation vegetable oil based biodiesel fuels.

3.4. Density

Density is important in terms of the consecutive breakdown steps of the injected fuel droplet. This will influence the total vaporisation duration and penetration distance of the fuel inside the combustion chamber, inevitably affecting the combustion kinetics. In the literature, there are studies showing that fuel density has an effect on exhaust emission characteristics, especially NO_x and PM emissions [33]. In diesel fuel injection systems, the volume of fuel discharged by the injector needle moving upwards is sprayed into the cylinder. Therefore, if a high density fuel is used, more fuel mass will be sprayed into the engine cylinder, affecting the air excess ratio in the flame front. In addition, in the studies carried out on diesel engines with electronically controlled fuel injection system, it has been observed that as the fuel density increases, the time between the signal input to the injector and the lifting of the injector needle (response time) increases [34]. This delay in the response time may affect both injection characteristics and combustion characteristics such as ignition delay, start and end of combustion.

The density specification in EN 14214 is 860-900 $kg.m^{-3}$ while ASTM D6751 does not specify any density standard. When diesel fuel standards are analysed, it is seen that there is no density specification in ASTM D975 (as in the biodiesel standard) while the density is required to be 820-845 $kg.m^{-3}$ in EN 590.

As can be understood from Table 2, the density values of all biodiesel fuels were within the range required in EN 14214. Regardless of feedstock and generation, the density values of all biodiesel fuels were very close to each other. The highest density (891.46 $kg.m^{-3}$) and the lowest density (875.83 $kg.m^{-3}$) were measured for SoB and SyB, respectively. The density value of WFB (876.70 $kg.m^{-3}$) was lower than those of all vegetable oil based biodiesel fuels except SyB (875.85 $kg.m^{-3}$) and AlB (880.25 $kg.m^{-3}$). Similar situation was also detected in the kinematic viscosity values of the related fuels. It can be said that the density value of AlB, the third generation biodiesel, was close to that of the 1_{st} and 2_{nd} generation vegetable oil origin biodiesel fuels. As mentioned before, WOB was of palm oil origin. When WOB and PoB fuels are compared, it is seen that similar to the viscosity values, the density value of WOB (891.31 $kg.m^{-3}$) was higher than that of PoB (885.15 $kg.m^{-3}$). This result can be explained by the chemical groups formed as a result of the degradation reactions in the oil during the frying process and inevitably present in biodiesel fuel.

3.5. Cetan index

Cetane number is one of the most critical fuel properties as it directly affects the combustion process and consequently all engine characteristics. However, before explaining the cetane number and its significant effect on the combustion process, it will be useful to briefly examine the diesel combustion process. Combustion in diesel engines can generally be analysed in 4 stages: 1- Ignition delay, 2- Pre-mixed controlled combustion phase, 3- Diffusion-controlled combustion phase, 4- Post-combustion phase. An ignition delay is the time between the start of fuel injection and the start of combustion. The pre-mixed controlled combustion is the combustion stage in which all of the fuel injected and vaporised into the combustion chamber during the ignition delay period ignites at the same time and can also be referred to as uncontrolled combustion. In the diffusion-controlled combustion, which is the main phase of diesel combustion, the combustion phenomenon (and therefore the temperature and pressure increase) is controlled by the amount of fuel injected onto the flame. Post-combustion is the last stage of the diesel combustion process [35]. In today's modern diesel engines with electronically controlled fuel injection system, a small amount of fuel is injected into the cylinder in this combustion phase in order to reduce HC and especially PM emissions. Cetane number is an indicator of the flammability of a fuel. In other words, fuel with high cetane numbers can burn more easily. The cetane number significantly affects the ignition delay time. As the cetane number increases, the ignition delay decreases. As a result, since the amount of fuel burning at the same time in the pre-mixed combustion phase will decrease, the sudden pressure and temperature rise will also decrease. Thus, the engine runs more quietly and smoothly. In addition, NO_x emission, which is the biggest problem for diesel engines, is also significantly affected by this situation. The increase in cetane number reduces NO_x emission [36-39]. When both biodiesel and diesel standards of the EU and the USA are analyzed, it is seen that higher cetane number is required in EU standards compared to the USA. While a minimum of 51 is required as cetane specification in EN 14214, this limit is 47 in ASTM D6751. While the minimum cetane number should be 46 in EN 590, this value is 40 in ASTM D975.

The cetane number of a fuel is measured by using a cetane engine. However, since the cetane engine is very expensive, it can only be used in a very limited number of research centres. In addition, the determination of the cetane number is time-consuming and labour-intensive. Instead of cetane number, the cetane index (CI) can be calculated by using a fuel’s density value and distillation range. The difference between experimentally determined cetane number and estimated cetane index is negligible. In this study, ASTM D4737-21 method was used to calculate the cetane index values of test fuels. Cetane number values of WCB and WFB fuels were quoted from Ref (27). Distillation temperatures of biodiesel fuels were given in Table 3.

Table 3. Distillation range of biodiesel fuels.

Dist. Temp.	AlB	WOB	CtB	HoB	RoB	CoB	OoB	SfB	PoB	SyB	SoB
IBP	325.3	92.0	312.2	327.5	307.7	244.1	87.3	93.4	281.8	227.2	90.0
5%	326.5	322.7	329.6	330.2	331.8	329.6	327.6	317.8	320.3	325.0	312.5
10%	336.3	323.1	330.4	330.6	331.8	330.5	328.1	326.8	321.7	327.6	320.5
15%	337.1	323.4	330.6	331.0	332.1	330.7	329.0	334.1	321.7	328.4	321.1
20%	337.8	323.8	330.9	334.8	332.5	331.0	330.0	335.4	322.0	329.1	321.8
30%	338.5	324.3	331.1	334.8	332.8	331.3	330.2	335.4	322.3	329.4	322.8
40%	339.6	325.6	332.0	334.9	333.1	331.3	330.3	335.8	322.9	329.9	324.2
50%	340.5	326.5	333.3	335.3	333.2	331.4	330.4	336.4	324.1	330.6	325.6
60%	341.6	327.1	334.4	335.9	333.5	332.2	330.5	337.2	325.0	330.9	365.5
70%	343.8	327.5	336.0	337.0	334.4	333.1	331.4	338.8	326.5	331.6	327.4
80%	345.4	328.4	338.8	338.9	335.8	334.6	333.3	339.3	327.9	333.4	328.5
90%	348.6	336.2	344.6	343.5	340.8	340.2	338.2	343.5	330.0	336.5	337.0
95%	349.8	339.4	345.6	347.8	343.4	344.6	341.7	349.3	332.1	337.8	338.2
FBP	350.6	341.1	347.2	353.9	347.9	349.5	349.0	355.2	339.8	341.3	340.1

As can be seen from Table 2 where fuel properties are given, the difference between WOB and WFB fuels with the lowest (50.54) and highest (58.80) cetane values, respectively, was 16.34%. The highest CI value (57.70) among the first generation biodiesel fuels belonged to HoB. The CI of OoB (57.09) was also very close to this value. The CIs of SoB (51.39) and SyB (51.50) were almost the same. The cetane index of CtB (55.82), which is classified as second generation biodiesel, and AlB (55.90), which is a third generation biodiesel, were almost the same. The cetane index of WCB (52.30), one of the two animal fat based biodiesels in this study, was about 11% lower than the cetane index of WFB (58.80), the other animal fat based biodiesel. However, all of the biodiesel fuels met the minimum cetane specification given in EN14214.

4. Conclusion

Biodiesel can be produced from many different feedstocks. The feedstock type from which a biodiesel fuel produced influences not only the production cost of the obtained fuel but also the sustainability of the biodiesel industry. Furthermore, the physico-chemical fuel properties of biodiesels from different origins are substantially dependent on the feedstock characteristics since the fatty acid distribution of the feedstock does not undergo a significant change during transesterification reaction in which biodiesel fuel is obtained. Biodiesel fuels are classified in three generations according to the type of feedstock they are produced from. First generation biodiesel fuels produced from conventional edible vegetable oil feedstocks, second generation biodiesel fuels produced from inedible vegetable oils, waste frying oils and waste animal fats, and third generation biodiesel fuels based on algae oil.

In this study, some key fuel properties (kinematic viscosity, density, flash point, cold filter plugging point and cetane index) of 13 different biodiesel fuels including three biodiesel generations were compared with each other and with the specification values given in European and American biodiesel standards. Among the biodiesel fuels, only the kinematic viscosity values of WOB and WCB exceeded the upper limit value given in EN 14214 standard and could not meet this fuel specification.

The density values of biodiesel fuels were very close to each other (875.83 kg.m^{-3}-891.46 kg.m^{-3}) and were within the required standard range. AlB had the lowest flash point (142 °C). The differences between this value and the other biodiesels' flash points were significant. The highest flash point (184 °C) was measured for HoB. The cetane values of the biodiesel fuels ranged from 50.54 (WOB) to 58.80 (WFB), but all biodiesels regardless of the feedstock type met the minimum cetane limit specification. Among the fuel properties of different generation biodiesels investigated in this study, the biggest differences were seen in CFPP temperatures. The CFPP value of AlB was -10 °C, while WFB (10 °C), WOB (9 °C) and POB (8 °C) had very high CFPP temperatures. Poor cold flow properties of biodiesel fuels from waste feedstocks and palm oil can cause operational problems, especially in cold regions.

The viscosity and cold flow properties of biodiesel fuels produced from waste frying oil and waste animal fats may cause problems in case of their neat usage in internal combustion engines. However, these biodiesels, which are classified as second generation, can be blended with other biodiesel fuels in certain ratios. Thus, a relatively cheaper biodiesel can be obtained and the properties of other biodiesels such as lubricity, calorific value and oxidative stability can be improved. Among the biodiesel fuels of different generations analysed in this study, AlB has the most remarkable properties (low viscosity, density and flash point values, high cetane number and relatively good CFPP temperature). Therefore, it can be said that algae oil based biodiesel fuels have an important technical potential for being used especially in the aviation sector. In the future studies in this field, it will be useful to investigate how the fuel properties will change by blending the third generation biodiesel fuels produced from various algae oils with different jet fuels at certain ratios. Also, algae-based biodiesel fuels' effects on the characteristics of gas turbines should be studied in detail.

CRediT authorship conribution statement

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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