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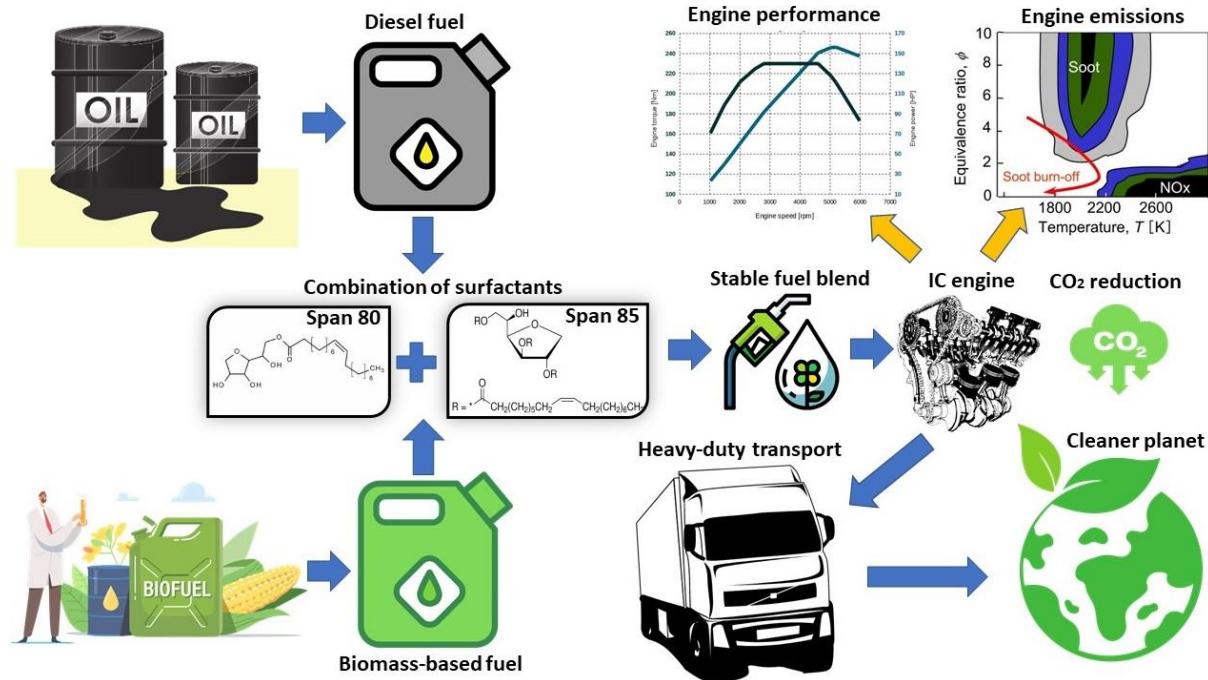
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# Biomass-based fuel blends as an alternative for the future heavy-duty transport: A review.

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## Graphical abstract



## Abstract

This paper analyses the current trends in application of biomass-based fuels as a valid option for heavy-duty transport and discusses their technology readiness levels, cost and emphasizes on these fuels to be applied as drop-in fuels in heavy-duty engines to minimize potential green-house and toxic gas emissions. Through the extended analysis, this study has identified that ethanol could be the best candidate for application in heavy-duty transport in terms of sustainability, cost, and emission reduction. Ethanol can be used in high concentrations as an additive or blended with the conventional diesel, which still remains a main type of fuel for heavy-duty transport. However, in order to completely adapt ethanol-diesel fuel blends to heavy-duty transport, a few challenges have to be resolved. The first challenge is the phase separation when high-concentration ethanol is blended with neat diesel. This can be fairly resolved by using certain types of surfactants, which will not negatively affect, but on the contrary, result in engine performance improvements as well as emission reductions. The second challenge is the ignition quality of the blends, as the cetane number of an ethanol-diesel blend decreases when high-concentration ethanol is blended with neat diesel. This can be resolved by using certain types of cetane improvers, as highlighted in this paper. The third challenge is the sustainable production and supply of ethanol without competing with food producers and minor impact on the indirect land use. This challenge can be resolved by producing ethanol from different types of organic waste, wastewater and biomass.

**Keywords:** Heavy-duty transport; Biomass-based fuels; Diesel-ethanol fuel blends; Surfactants; Phase separation; Internal combustion engines; Exhaust gas emissions.

**Word Count:** 9130 words (excluding title, author names and affiliations, keywords, abbreviations, table/figure captions, acknowledgments and references)

### List of abbreviations

BEV: Battery Electric Vehicle	HLB: Hydrophilic Lipophilic Balance
BSFC: Brake Specific Fuel Consumption	HRR: Heat Release Rate
BTE: Break Thermal Efficiency	HVO: Hydroprocessed Vegetal Oil
BTL: Biomass to Liquid	ID: Ignition Delay
CBG: Compressed Biogas	ILUC: In Land Use Change
CD: Combustion Delay	LBM: Liquefied Biomethane
CI: Compression Ignition	LDV: Light Duty Vehicle
CNG: Compressed Natural Gas	LNG: Liquefied Natural Gas
CO: Carbon Monoxide	LPG: Liquefied Petroleum Gas
CP: Combustion Pressure	NOx: Nitrogen Oxide
DME: Dimethyl Ether	OME: Oxymethylene Dimethyl Ether
EC: European Commission	PM: Particulate Matter
EU: European Union	PN: Particulate Number
EV: Electric Vehicle	PODE: Polyoxymethylene dimethyl ethers
FT: Fischer Tropsch	RED: Renewable Energy Directive
GHG: Green House Gas	RES: Renewable Fuel Standard
HC: Hydrocarbon	SI: Spark Ignition
HDV: Heavy Duty Vehicle	TRL: Technology Readiness Level
HEFA: Hydroprocessed esters and fatty acids	WHSC: World Harmonized Stationary Cycle
HFC: Hydrofluorocarbon	W/O: Water in Oil
	WTT: Well to Tank

### 1. Introduction

Nowadays our societies are facing a critical challenge which is to fight against global warming. In this context, all the countries around the world, and especially the European Union, took several measures in the aim to reduce the greenhouse gas emissions [1]. As an example, the European Energy directive aims to achieve 20% renewable energy by 2020 and 10% for the transport with a global cut of 6% for the GHG emissions [2]. As the transportation sector is one of the largest global transmitters of GHG, the sector has become a priority to mitigate the emissions we release in the atmosphere by finding alternative to fossil fuels with cleaner fuels,

gas or electric vehicles. In this aim, several norms and laws have also been introduced to obligate countries to reduce their emissions [3].

However, many countries and especially developing countries, are far behind the most developed nations on topic of the production of renewable energy and green transport [4; 5]. Currently, the European Union is a leader with their Energy Directive which should be very crucial for driving the utilisation of low-emission transport and renewable fuels in the next years up to 2030 [1]. To achieve these goals, the Euro 6 norms [6] were introduced to fix emission limits to the vehicle and engine manufacturers. Table 1 shows that new emission norms of Euro 6 standards have been introduced for passenger cars and light-duty vehicles effective from 2018-2020. Emission norms of Euro 6 standards for heavy-duty vehicles are quite outdated, as shown in Table 2, however, the European Union is revising its main law limiting pollution from heavy-duty vehicles, vans and trucks in 2021-2022. A new emission standard called Euro 7 is under development and is expected to be implemented from 2025. New standards are urgently needed, as a failure to address the causes of the toxic air that millions of us are forced to breathe daily, will result in more health problems and deaths.

Table 1. Euro 6 norm requirements for passenger cars and light duty vehicles

Data in g/km	TIER	Date (Type Approval)	Date (First Registration)	CO	NOx	HC+NOx	PM	PN (#/km)
PASSENGER CARS	EURO 6C	-	September 2018	0.50	0.080	0.170	0.0045	6x10 <sup>11</sup>
	EURO 6D	September 2017	September 2019	0.50	0.080	0.170	0.0045	6x10 <sup>11</sup>
LIGHT COMMERCIAL VEHICLES < 1305 kg	EURO 6C	-	September 2018	0.50	0.080	0.170	0.0045	6x10 <sup>11</sup>
	EURO 6D	September 2017	September 2019	0.50	0.080	0.170	0.0045	6x10 <sup>11</sup>
LIGHT COMMERCIAL VEHICLES 1305 - 1760 kg	EURO 6C	-	September 2019	0.630	0.105	0.195	0.0045	6x10 <sup>11</sup>
	EURO 6D	September 2018	September 2020	0.630	0.105	0.195	0.0045	6x10 <sup>11</sup>
LIGHT COMMERCIAL VEHICLES > 1760 kg (max 3.5 T)	EURO 6C	-	September 2019	0.740	0.125	0.215	0.0045	6x10 <sup>11</sup>
	EURO 6D	September 2018	September 2020	0.740	0.125	0.215	0.0045	6x10 <sup>11</sup>

Table 2. Euro 6 norms for heavy-duty vehicles

Data in g/kWh	TIER	Date	Test Cycle	CO	HC	NO <sub>x</sub>	NH <sub>3</sub> (ppm)	PM	PN (#/kWh)
Heavy-Duty Diesel Engines	EURO 6	31 December 2012	WHSC	1.5	0.13	0.4	10	0.01	8x10 <sup>11</sup>
	EURO 6	31 December 2012	WHTC	4.0	0.16	0.46	10	0.01	6x10 <sup>11</sup>

Unfortunately, even if these regulations help to improve the situation, it is still not enough, and alternatives need to be found and CO<sub>2</sub> emissions must be reduced to reach the 95g CO<sub>2</sub>/km target for new passenger cars manufactured by 2021 and 147g CO<sub>2</sub>/km for light commercial vehicles by 2020. Regarding the heavy duty vehicles, the European Commission stated to introduce regulations with the intention of reducing the average CO<sub>2</sub> emissions for new trucks by 2025 to 15% as well as a further reduction of 30% by 2030, compared to 2019 [2]. Biofuels and electric vehicles have started to be used to reach these targets. Biodiesel is currently the major biofuel used in the EU for transport and it represents 80% of all biofuel use in 2017 and accounts for 12.5 Mtoe. It is followed by ethanol which represents 18% and see its production increase year-by-year as it represents a promising alternative [2; 7]. Moreover, we can see in the Fig.1 the proportion of electric and hybrid vehicles across Europe in 2017 [8]. EU is still at

the beginning of the electrification of its fleet. However, we can see that Norway is already reaching a really high portion of these electric and hybrid vehicles and is in a way of showing to the rest of EU the future, as many experts foresee it nowadays.

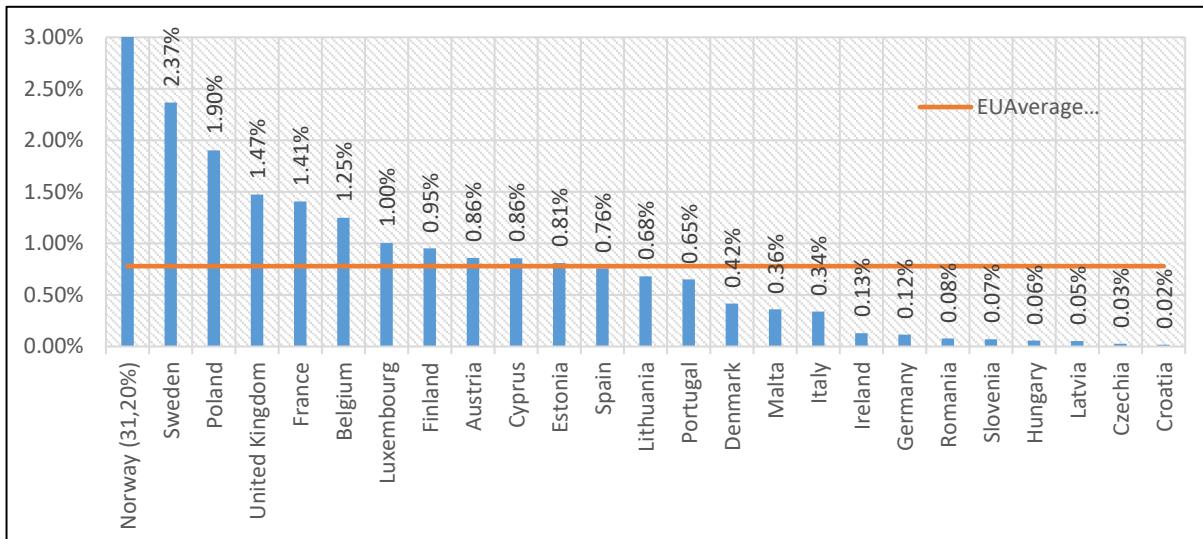


Fig. 1. Share of Electric and Hybrid vehicles across Europe in 2017, Norway (2018 data), Italy (2016 data), Romania (2015 data)

Battery electric vehicles are expected to be the future of our power source for transport and some countries and manufacturers have announced that they want to move to a full electric fleet in the next 30 years. As an example, Mercedes-Benz announced that the group aims to produce a fleet of new carbon-neutral passenger and make plug-in hybrid or all-electric vehicles more than 50% of their sales by 2030. This includes extending the electrification of vehicles to their vans, trucks and buses [9].

However, even if electrification seems to be fairly achievable for passenger cars and light duty vehicles, lots of questions are still to be answered for heavy duty vehicles. Niculescu et al. [10] highlighted the fact that electrification needed to reach a better cost and performance level but also a better well-to-wheel efficiency. In the Future Transport Fuels report (2011) [11], European Commission summarized the ability of different alternatives to fossil fuels to cover different transport modes, as shown in Table 3. They established this coverage by considering that replacing actual fuels with biofuels for different transportation means, while still using the current technologies and fuelling infrastructures was possible.

Table 3. Different modes of transportation based on different alternative fuels. (BEV - Battery Electric Vehicles; HFC - Hydrogen Fuel Cell; CNG - Compressed Natural Gas; CBG - Compressed Biogas; LNG - Liquefied Petroleum Gas; LPG - Liquefied Petroleum Gas) [11]

		Road/passengers			Road/freight			Rail	Water		Air
		Short	Medium	Long	Short	Medium	Long	Inland	Short Sea Shipping	Maritime	
<b>Electric</b>	BEV										
	HFC										
	Grid										
<b>Biofuels (liquid)</b>											
<b>Synthetic fuels</b>											
<b>Methane</b>	CNG										
	CBG										
	LNG										
<b>LPG</b>											

As seen in the table, electricity cannot give us enough guarantees as a power source now concerning heavy duty transport. In addition, many manufacturers consider EV as zero emission vehicles, but they forgot to consider the carbon footprint of their batteries [12]. Indeed, during the life cycle of batteries, the greenhouse gas emissions they produce is close to zero, but they require a lot more energy than traditional power units to be manufactured [13]. The problem with that point is that it depends on many factors to evaluate the GHG emissions of the battery such as where the energy comes from to manufacture the battery, which chemical components are used for the assembly, etc. If it seems that batteries may have a limited impact on the environment when they are produced with renewable energy, but it becomes problematic when produced in countries where fossil energy is used to produce electricity for their production. There is as well two other problems to highlight, which are: first the fact that batteries contain certain amount of cobalt and nickel. It is important to take into account that cobalt is in the list of critical so called “conflict mineral” for the European Industries (European Commission, 2017) [14]. This means that cobalt is extracted from mines own by armed groups and purchasing from these mines is forbidden. Furthermore, as nickel and cobalt are quite scarce to come across nowadays, it could be problematic to meet the market demand in the future with a full electric fleet of vehicles.

A great improvement needs to be done as well regarding the recycling of the batteries. Indeed, the current legislation in the EU states that 95% of batteries introduced on the market need to be collected and that 50% of the total weight of them needs to be recycled. However, a large part of the batteries is not recycled today as it is extremely complex and therefore more expensive to recycle than just produce their components again [15]. In the end, even if electricity seems to be the best alternative to fossil fuels in the future, it would not be realistic to think that the full transition from fossil energy to full electric vehicles could be achieved in the future and even less for heavy-duty vehicles. It could then be interesting to focus on alternative fuels, which some of them are really promising and could reduce emissions from vehicles currently on roads and even be used with future heavy-duty vehicles.

Therefore, this review explores the challenges and opportunities for biomass-based fuel blends to be used as alternatives for the future heavy-duty transport as an efficient and sustainable way to mitigate exhaust gas emissions. We have analysed the current trends in application of biomass-based fuels and introduced criteria showing the strengths and weaknesses of each fuel based on their technology readiness levels, cost and emphasizes on these fuels to be applied as drop-in fuels in heavy-duty engines to minimize potential green-house and toxic gas emissions. The structure of this review is shown in Figure 2.

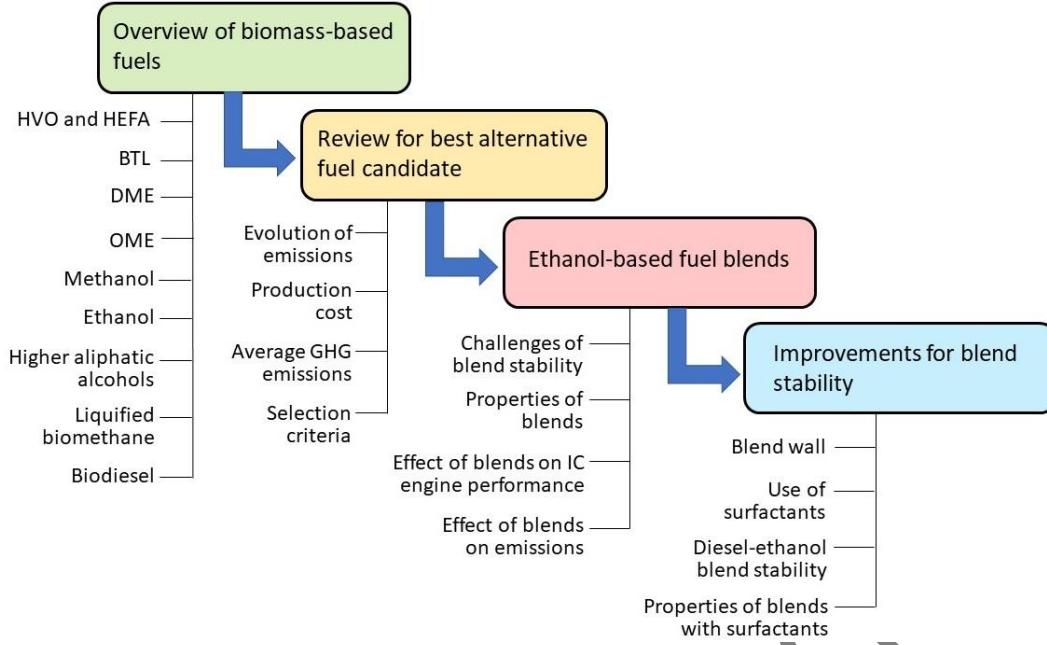


Fig. 2. Structure of this review paper.

## 2. Biomass based fuels, an efficient and sustainable way to mitigate emissions

As mentioned in the introduction, over the last few years, a great improvement has been made over the alternative fuels. They represent an interesting solution to help to reduce the emissions in an effective way and most of them are relatively easy to produce. Moreover, most of them could be blend with or used as substitutes to diesel or petrol without requiring any modifications on the engines. Therefore, it could represent from an economical point of view a real advantage in comparison to electric vehicles. We decided to summarize below a list of the most promising biofuels based on the report: “Survey on advanced fuels for advanced engines” published by the IEA Bioenergy in October 2018.

For each of these biofuels, they took seven criteria into account, but we will only keep the relevant ones for our study as seen below:

- Typical feedstock for biofuel production that was commercialised or under investigation.
- Fuel production processes on typical or potential co-products.
- Technology readiness levels (TRL) [16], by European Commission (EC 2010).
- Fuel production costs as an indicator of economic competitiveness.
- GHG emissions for fuel production and distribution as an indicator of environmental impact (e.g. EU RED or US RFS).

### 2.1 Hydrotreated vegetable oils (HVO) and hydroprocessed esters and fatty acids (HEFA)

HVO/HEFA is a relatively easy to produce. Lipid feedstock, like vegetable oils, used cooking oils or even animal origin is converted renewable diesel using hydrogen [17]. As its characteristics are close to traditional diesel, it is compatible with current engine design. Also, the heating value is close to fossil diesel and one of the main benefits is that it enables reducing the soot emissions from the engines. However, HVO/HEFA presents high cetane number with low viscosity and lubricity, which needs to be corrected by blending [18].

HVO/HEFA is a fuel already in production which is classed as 9/9 on the TRL [16]. Its production cost is evaluated between 19 and 47 USD per GJ [30]. The emission

characteristics of HVO has analysed as a neat fuel as well as diesel blend [19-24]. On average, it shows a reduction of about 35% of carbon monoxide (CO) and hydrocarbons (HC) emissions with a decrease of 25% of the particulate matter. Unfortunately, it didn't show a positive trend about the nitrogen oxide ( $\text{NO}_x$ ) emissions. According to RED, the typical GHG for HVO/HEFA is going from 5 to 76 kg  $\text{CO}_{2\text{eq}} \text{ GJ}^{-1}$  [25].

## 2.2 Biomass-to-liquid (BTL)

BTL is a fuel that requires a lot of treatment to be processed. The typical feedstock for BTL is wood, stalk material (straw, plants, etc.) but also municipal waste. Although, there are three main processes to obtain BTL, it is interesting to note that these processes can be also used to obtain other type of alternative fuels as we will see further. Each of these processes have advantages and disadvantages. The first is a gasification process [26], with no restriction to a plant-based feedstock. This means that most lignocellulosic biomass can be considered. Nevertheless, if the water content is high and too many impurities appear during the Fischer-Tropsch process [27], it could be problematic as it would require cleaning the gas. The second one is a pyrolysis process, its main advantage is that it is an inexpensive process which allows a total utilisation of the feedstock. However, the final bio-oil obtained from this process is not good for use as fuel, as it is a relatively low energy density liquid with corrosive properties which could affect the reliability of internal combustion engines [28]. The third one is a liquefaction process, similar to the pyrolysis one. However, even if liquefaction is a more expensive process, the bio-oils obtained have much better properties than the one obtained through pyrolysis [29]. Therefore, regardless its cost, the liquefaction process seems to be the most suitable one to obtain a fuel with properties close to actual fossil fuels.

Even if BTL is a fuel with promising qualities, it is currently considered as 5/6 on the TRL scale [16]. The typical production cost of BTL is going from 18 to 62 USD per GJ [30]. Concerning the emissions, when the fuel is produced via the FT process, it tends to see the emissions decreasing. A few studies [31-33] showed a decrease of 25% for HC, 40% for CO, 10% for  $\text{NO}_x$  and 20% for PM. According to RED, the typical GHG emissions for BTL are between 7 and 100 kg  $\text{CO}_{2\text{eq}} \text{ GJ}^{-1}$  [25].

## 2.3 Dimethyl ether (DME)

Dimethyl ether is a biomass-based fuel, that was first used as a source of energy in several countries around the world and can be produced via catalytic conversion of methanol [34] with the main advantage of resulting in a high octane fuel [35]. However, as DME present a relatively low lubricity it needs to be blended to be working efficiently in engines and will still require a modification of the injection system. Similar to BTL, DME is still in a development phase and classified as 4-6 on the TRL scale [16]. One of its other main advantages is that it is relatively cheap to produce with a typical production cost going from 16 to 30 USD per GJ [30].

Regarding the emissions from engines running on DME, as it requires modifications, it is therefore not possible to make a direct comparison with fossil fuels. However, few studies [36-44] have been carried out but they have not shown a clear trend. According to RED, the typical GHG emissions of DME are between 1 and 72 kg  $\text{CO}_{2\text{eq}} \text{ GJ}^{-1}$  [25]. Therefore, further engineering investigations needs to be carried out to obtain clear trends and to fully exploit DME potential.

## 2.4 Oxymethylene Dimethyl Ether (OME)

Oxymethylene dimethyl ether is also a biomass-based fuel. It is considered as a diesel oxygenated and six variations of OME exist ( $\text{CH}_3(\text{OCH}_2)_n\text{OCH}_3$  ( $n=1-6$ )). However, only OME1 is being produced in commercial quantities now [45]. When OME is produced, a few impurities such as methyl can appear and result in problems for the efficiency and the performance of an engine. From OME2, the fuel shows acceptable properties to be used in CI engines, even if it presents some limitations such as high density and lower

viscosity than those of traditional diesel. The boiling point is in an acceptable range (from 42 to 280°C) and shows a high-octane number [46-49].

As there is a range of different OMEs and as only OME1 is being commercialised nowadays, this fuel is only ranked as 3 on the TRL scale [16]. One of the main benefits of OME would be its production costs which is between 33 and 50 USD per GJ (for coal based biomass) [30] but a study from Oyedun et al. [50] shows that following the type of biomass feedstock used to produce OME, daily production cost of 500 tons of dry biomass ranges from 1.66 to 1.93 USD per litre.

Regarding the emissions level, OME shows great improvement for soot emissions regardless the variation of OME is used. Studies [46-55] show that OME tends to reduce PM, CO and HC emissions with an average of 36%, 44% and 52%, respectively. Unfortunately, no clear trends appeared for NO<sub>x</sub> and some of the studies revealed a minor increase.

## 2.5 Aliphatic alcohols

Methanol and ethanol are both simple aliphatic alcohols (straight carbon chain) which have been used since the beginning of engine development. Both are produced from biomass feedstock. They have the main advantage of reducing in an effective way particulate emission and GHG. However, as they contain a high volume of water they need to be blended with diesel and emulsifiers are required to prevent phase separation [56-58].

### - Methanol

Methanol is produced [59] from biomass but mainly from methane and coal. Methanol can either be used in the process of DME and biodiesel, for example, or as a blend component for gasoline and diesel. The main advantage is that methanol has a high-octane number, which allows a higher compression ratios. However, methanol do have a massive disadvantage, which is that it is a toxic substance and therefore could not be accepted as a fuel due to blend walls, DME and OME. Regarding its technology readiness level, methanol is classified as 6 on the EU TRL scale [16] and its typical production cost is estimated between 14 and 54 USD per GJ [30].

Regarding the emissions, most of the studies have been carried out for methanol with gasoline blends but very few studies were carried out with diesel blends [60-62]. These studies have not shown a clear trend, they mostly shown a reduction in HC and CO emissions but some of them found a reduction in NO<sub>x</sub> when others found an increase up to 50%. Therefore, standardized tests need to be carried out to elaborate clear trends on exhaust gas emissions when methanol is used as fuel. According to RED [25] the typical GHG emissions are in a range from 2 to 58kg CO<sub>2eq</sub> GJ<sup>-1</sup>.

### - Ethanol

Ethanol is currently the most known biofuel around the world and keeps being improved in every of its aspects year after year. Initially produced from sugar or starch crops, a new generation came, and ethanol can now be produced from biomass [63] and therefore having a minor impact on the “indirect land use change” (ILUC) [64-66]. Ethanol is currently being blend with gasoline, but a few countries have been using it as a pure fuel, such as Brazil [67]. Some blends of diesel with ethanol started to be developed [67; 68] and seems to be more promising than gasoline blends especially regarding emission trends where gasoline did not give clear trends on the dependant of the emissions characteristics on the engine technology and operation. Therefore, it could be interesting to investigate previous studies on diesel-ethanol blends to define a trend as results are usually promising. One of the other advantages of ethanol is its ease to be produced. It is interesting to note as well that ethanol is already being commercialised and that lignocellulose ethanol is classified as 8 on the TRL scale [16]. Its typical production costs are going from 21 to 46 USD per GJ [30] and according to RED [25], the average GHG emissions range goes from 4 to 32 kg CO<sub>2eq</sub> GJ<sup>-1</sup>. Also, ethanol appears to be a promising alternative as it does not have impact on the ILUC when produced from lignocellulose and its close properties to diesel

could allow him to be added to diesel fuel in high concentrations without requiring any modifications of CI engines.

#### *- Higher aliphatic alcohols*

Tests have been conducted on higher aliphatic alcohols such as propanol, butanol and pentanol. However, for most of these studies [69-75], even if a decrease of PM and CO were documented, an increase in NO<sub>x</sub> an HC has been also seen. Some studies showed the average GHG emissions for butanol [76] were between 18 and 89 kg CO<sub>2eq</sub> per GJ.

Liu et al. [77] investigated blending effects of gasoline fuel with *n*-butanol on the fuel consumption and harmful emissions in a GDI vehicle. They showed that when compared to pure gasoline fuel at steady-state operation, blending *n*-butanol could reduce CO<sub>2</sub>, CO, total hydrocarbon (THC), and NO<sub>x</sub> emissions, which were also decreased by employing a higher blending ratio of *n*-butanol. Zheng et al. [78] conducted experimental studies on combustion and emissions of *n*-butanol/biodiesel under both blended fuel mode and dual fuel RCCI mode. They found that blended fuel mode can maintain high efficiency at all test loads and *n*-butanol ratios.

Higher aliphatic alcohols are in the early development phase with TRL of 5 [16], for example for butanol. Even if higher aliphatic alcohols are showing some interesting properties, including a high cetane number, they are not developed enough yet to be considered as alternative transportation fuels in an immediate future.

#### *2.6 Liquefied Biomethane (LBM)*

Liquefied natural gas (LNG) and since few years its renewable substitution, liquefied biomethane (LBM) has gained a popular interest especially in the HDV sector [79]. It can be easily produced from Biomass following a similar process of the DME process. It is interesting to note that LNG and LBM are fully ready to be commercialised and therefore classified as 9 on the TRL Scale [16]. These fuels present several strong advantages, they are cheap to produce (between 13 and 16 USD per GJ [30]), they have low GHG emissions (11 to 21 kg CO<sub>2eq</sub> per GJ [25]) and they show significant decrease in NO<sub>x</sub>, PM and CO emissions [80-81]. However, this fuel is not efficient when used with CI engines because LNG and LBM have a low cetane number which is fundamental for CI engines. Also, their use can have an impact on the reliability of engines since no additives are used as it would normally be with a more traditional fuel and the high combustion temperature puts the engines under hard running conditions. It also requires high energy needs for production. Even if LNG and LBM are promising when used in SI engines, improvements still need to be done to show promising results when used with CI engines.

#### *2.7 Biodiesel*

To conclude the review on biomass-based fuels, we will finalize with the most used one nowadays, the biodiesel. The first generation of biodiesel ended having a bad reputation due to many disadvantages it was showing. Firstly, biodiesel had a major impact on the ILUC by being produced mostly from virgin oil. Secondly, the fact that it did not show great improvement in exhaust gas emissions. New generations of biodiesel which tend to have a minor impact on ILUC and improved emissions have shown more promising results [82]. New generations have more oxygen, higher cetane number, higher density and viscosity but reduced lower heating value compared to diesel [83]. However, the use of biodiesel, depending on its feedstock and impact on ILUC, allows reducing the GHG emissions by approximatively 40% in comparison to neat diesel [84]. It is also important to note that it is an extremely cheap fuel to produce with an average production cost of 9,80 USD per GJ [85; 86]. However, studies [87-92] have shown that an increase in the biodiesel composition of blends increases NO<sub>x</sub> emissions. Therefore, it is difficult to see biodiesel as an alternative for the future heavy-duty transport as it has already reached the blend wall and also the public opinion for its 1<sup>st</sup> generation and that 2<sup>nd</sup> and 3<sup>rd</sup> generations still need to show improvements for NO<sub>x</sub> emissions.

### 3. Which fuel is the best alternative?

Fig. 3 shows the conversion process for renewable transport fuels [93] and summarizes the data presented above where we highlighted the strengths and weaknesses of the different fuels.

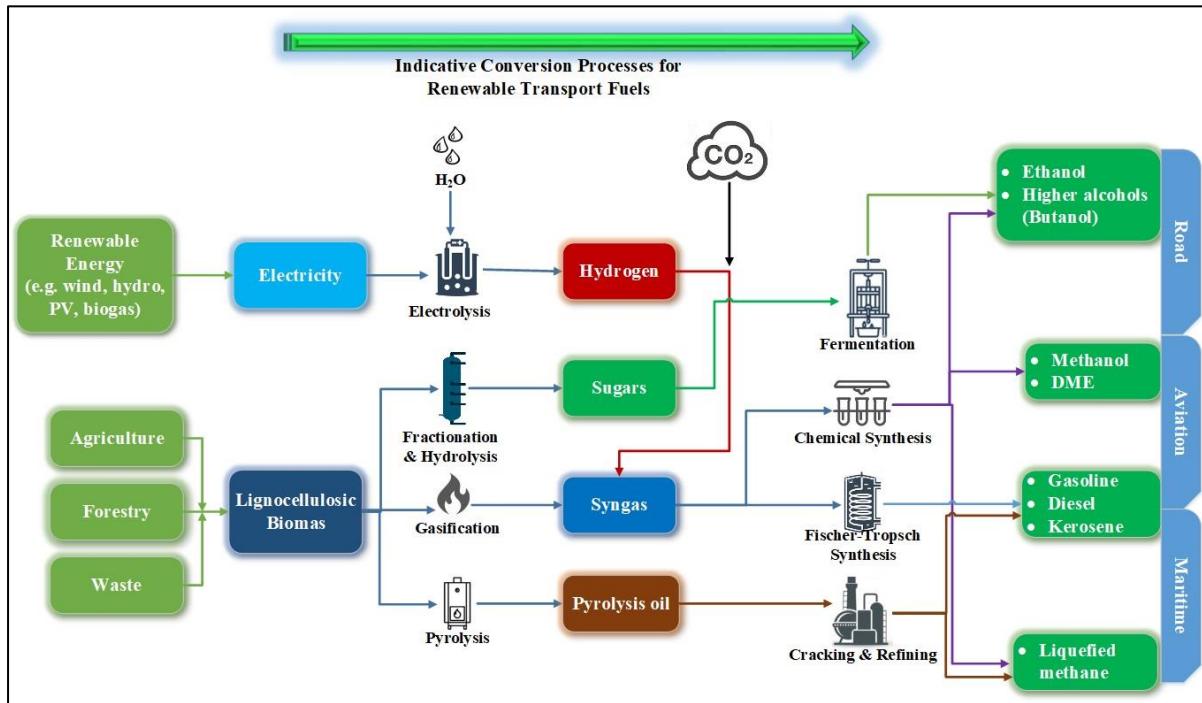


Fig. 3. Conversion process for renewable transport fuels [93]

Fig. 4 displays the evolution of emissions for some alternative fuels in comparison to neat diesel. BTL and especially OME show a great improvement in CO and PM emission reduction. These are oxygenated fuels with increased oxygen content, which improves the combustion in CI engines and allows to eliminate most of the CO and PM. So, it is viable to use high oxygen concentration fuels in CI engines in the aim of reducing in an efficient way the typical emissions that we find with the use of neat diesel.

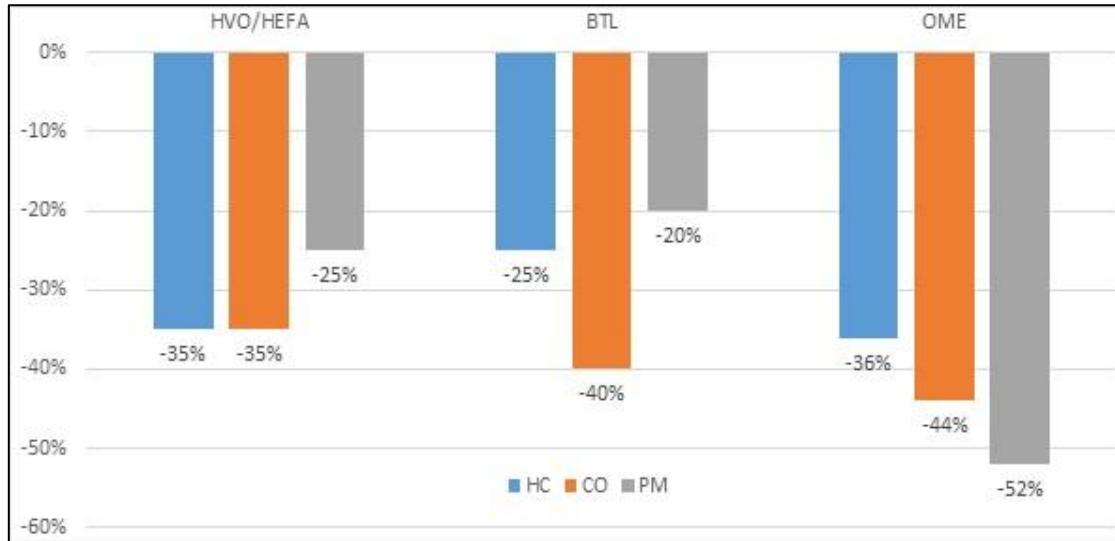


Fig. 4. Evolution of emissions for different alternative fuels in comparison to neat diesel [19-24]; [31-33]; [46]; [51-55]

Fig. 5 shows the minimum (blue) and maximum (orange) production costs of different renewable fuels in 2016 extracted from the DBFZ database [30]. The prices are dependent on the technology readiness level of the fuel and the difficulty of the production process. This means that fuels which are still in an early development stage would cost more money to produce than a fully commercialised fuel such as biodiesel. Therefore, we could expect the production costs of some of fuels to decrease in the future with the increased production efficiency.

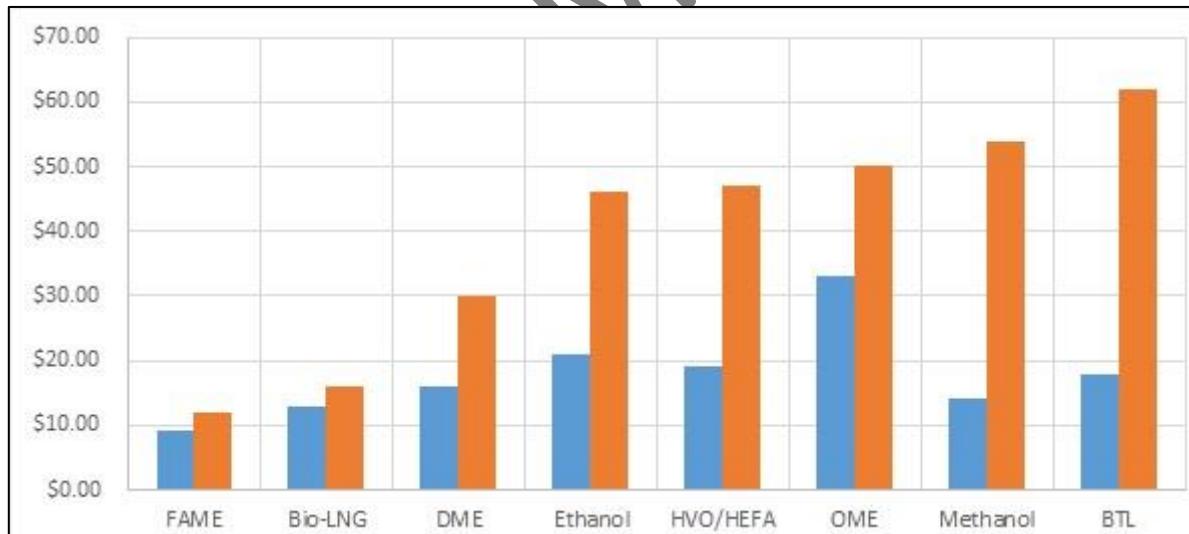


Fig.5. Minimum (blue) and maximum (orange) production costs of alternative fuels in 2016 in USD/GJ [30]

Fig. 6 shows the average greenhouse gas emissions of the different alternatives reviewed previously [25]. A difference between the minimum (blue) and the maximum (orange) values of emission levels is noticeable. This could be explained in different ways. Firstly, the presented data depends on the conducted test cycle and engine. Secondly, the formulation of the fuel used can also have a great influence on the results. Nevertheless, as for the average price, we can see that the highest values correspond mostly to fuels which are in an early development stage and therefore, their production cost is expected to decrease in the future.

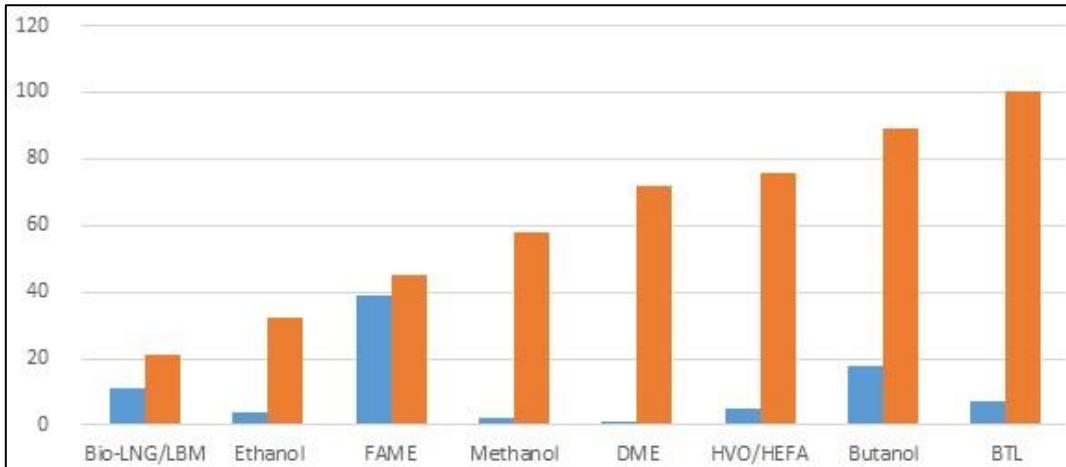


Fig. 6. Average GHG emissions of alternative fuels (kg CO<sub>2</sub>eq/GJ) [25]

Tables 4 and 5 provide summary for each fuel based on the cost, emission and adoptability as drop-in fuel. For each criterion, the best option is highlighted as green and the worst option is as red. As we can see in Table 4, each of the fuels show advantages regarding some of the criterion, which makes it very difficult to evaluate. Table 5 allows us to see which alternative fuel seems to be more promising (as 1 is for the most favourable and 3 is for the least favourable). Once we weight each fuel for each criterion, as shown in Table 5, it allows us to get a trend and therefore to see which biomass-based fuel could potentially be the most favourable. We can see that ethanol and bio LBM/LNG are getting the best average score of 1.5. Unfortunately, the lack of information on the bio LBM/LNG does not give us the most accurate results, but it is important to note that even if it shows promising results, bio LBM is a relatively new technology, and currently LNG is the mostly used.

Table 4. Summary of criteria showing the strengths and weaknesses of each fuel. (5 as major positive impact, 3 as no impact, and 1 as major negative impact)

Fuel	Minimum Production Price [30]	Maximum Production Price [30]	Minimum GHG Emissions [25]	Maximum GHG Emissions [25]	TRL [16]	Exhaust Gas emissions [93]	Drop In Fuel [93]	Raw Material Base [93]
HVO/HEFA	\$ 19,00	\$ 47,00	5 kg CO <sub>2</sub> eq/GJ	76 kg CO <sub>2</sub> eq/GJ	9	4	5	4
BTL	\$ 18,00	\$ 62,00	7 kg CO <sub>2</sub> eq/GJ	100 kg CO <sub>2</sub> eq/GJ	6	4	5	5
DME	\$ 16,00	\$ 30,00	1 kg CO <sub>2</sub> eq/GJ	72 kg CO <sub>2</sub> eq/GJ	6	5	--	methanol
OME	\$ 33,00	\$ 50,00	-	-	3	5	3	methanol
Methanol	\$ 14,00	\$ 54,00	2 kg CO <sub>2</sub> eq/GJ	58 kg CO <sub>2</sub> eq/GJ	6	3	2	5
Ethanol	\$ 21,00	\$ 46,00	4 kg CO <sub>2</sub> eq/GJ	32 kg CO <sub>2</sub> eq/GJ	8	5	4	5
Butanol	-	-	18 kg CO <sub>2</sub> eq/GJ	89 kg CO <sub>2</sub> eq/GJ	5	4	4	5
Bio LNG/LBM	\$ 13,00	\$ 16,00	-	-	9	3	5	4
Biodiesel	\$ 9,00	\$ 12,00	39 kg CO <sub>2</sub> eq/GJ	45 kg CO <sub>2</sub> eq/GJ	9	4	4	3

Table 5. Weighted table of the data of each fuel for each criterion (3 as the best score and 1 as the worst score)

Fuel	Minimum Production Price [30]	Maximum Production Price [30]	Minimum GHG Emissions [24]	Maximum GHG Emissions [24]	TRL [16]	Exhaust Gas emissions [93]	Drop In Fuel [93]	Raw Material Base [93]	Average Score
HVO/HEFA	2	2	3	2	3	2	3	2	1,625
BTL	2	1	3	1	2	2	3	3	1,875
DME	2	2	3	2	2	3	1	3	1,75
OME	1	2	-	-	1	3	1	3	2,167
Methanol	3	1	3	3	2	1	1	3	1,875
Ethanol	1	2	3	3	3	3	2	3	1,5
Butanol	-	-	2	1	2	2	2	3	2
Bio LNG/LBM	3	3	-	-	3	1	3	2	1,5
Biodiesel	3	3	1	3	3	2	2	1	1,75
3= x≤14\$		3= x≤25\$		3= x≤10 kg CO <sub>2eq</sub> /GJ	3= x≤60 kg CO <sub>2eq</sub> /GJ	3= 6< x	3= 5	3= 5	3= 5
2= 14\$<x≤19\$		2= 25\$<x≤50\$		2= 10< x≤20 kg CO <sub>2eq</sub> /GJ	2= 60< x≤80 kg CO <sub>2eq</sub> /GJ	2= 3< x≤6	2= 3	2= 3	2= 3
1= 19\$< x		1= 50\$< x		1= 20< x kg CO <sub>2eq</sub> /GJ	1= 80< x kg CO <sub>2eq</sub> /GJ	1= x≤3	1= 1	1= 1	1= 1

As a conclusion, ethanol is our most promising alternative, especially because it does have close physical properties to neat diesel fuel which could be used in high concentrations for CI engines. Numerous experimental studies have been completed to investigate the effect of high concentrations of ethanol in biodiesel and diesel fuel blends on performance and emission characteristics of IC engines and showed promising results. The following sections of this paper will elaborate on the specifics of the application of ethanol as an addition to biodiesel and diesel and blends with the specific emphasis on high concentration use of ethanol.

#### 4. Ethanol-based fuel blends

As was previously highlighted, ethanol has lots of advantages (close properties to neat diesel, low price, renewable feedstock, etc.), which makes this fuel a promising alternative for the future heavy-duty transport. Nowadays, ethanol is mostly used in gasoline blends with up to 5%, 10% or 20% [94] and we can even see high-level blends of ethanol in gasoline (E85) [95]. The common application of ethanol in gasoline engines is due to its high octane number and high combustion speed, which result in the increase of thermal efficiency and engine torque with decrease in CO<sub>2</sub>, NO<sub>x</sub> and THC emissions [96]. It has been challenging to use ethanol in diesel engines due to its low cetane number, which is fundamental for these engines. However, by adding cetane improvers to the fuel blend allows using ethanol in diesel engines without requiring any engine modifications [97]. Furthermore, using ethanol in diesel engines instead of gasoline engines is preferable because diesel engines combustion efficiency is better than gasoline engines with lower emissions. However, even if adding a cetane improver allows using ethanol without needing any engine modifications, there is another problem associated with using ethanol in high-concentrations. Due to the high water content, phase separation appears between ethanol and neat diesel when ethanol used in high concentrations. To overcome this problem certain types of surfactants have to be used to ensure stable fuel blends without phase separation **Error! Reference source not found..** It could be then interesting to have a look at the different surfactants that can be used to avoid phase separation in blends before to review few studies led on high ethanol concentration blends.

##### 4.1 Types of surfactants to improve ethanol diesel blends

Surfactants are chemical substances that have the ability to alter the properties of two immiscible phases in their interfaces. In other words, they create self-assembled micelles in a solution to absorb the interface between different fluid groups. The first one needs to be a hydrophobic group and the second one is a hydrophilic group [101]. In many cases, surfactants are used as emulsifiers in the aim of absorbing the separation surface between two phases.

Fig. 7 shows a surfactant added to a water-oil (W/O) solution. The circular part of the surfactant molecule represents a hydrophilic portion and the tail represents a hydrophobic portion. First, surfactant molecules are absorbed into the oil surface then, surfactant molecules are being surrounded by oil once it is removed to prevent its re-deposition in the bottom and to prevent phase separation.

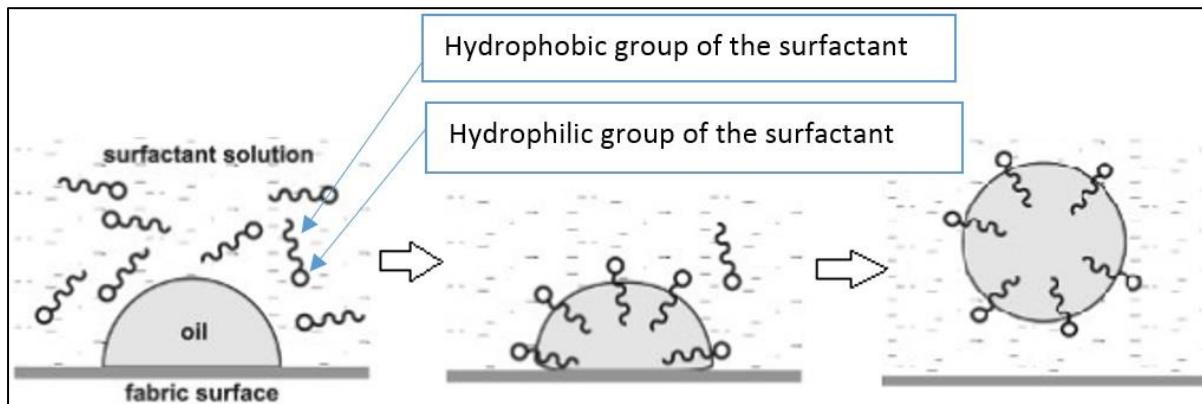


Fig. 7. Effect resulting from addition of surfactants into a W/O solution [101]

There are four major groups of surfactants: 1) biosurfactants, 2) polymeric surfactants, 3) synthetic surfactants and 4) non-ionic surfactants. Biosurfactants are surface-active biomolecules produced by microorganisms, their main advantages come from the fact that they have low toxicity, they are easy to produce, and they are renewable [102]. Salek et al. [103] reviewed the use of biosurfactants to create water in oil emulsions for cosmetic and food use and showed some interesting results. Therefore, it could be useful to study the effect of biosurfactants on ethanol-diesel blends because they possess both hydrophilic and hydrophobic properties causing them to locate themselves at interfaces between fluids such as hydrocarbons and water [104; 105]. Polymeric surfactants present the benefits of increasing the viscosity of water and decreasing the interfacial tension which allows water to be miscible in oils. However, they are relatively expensive to produce [106] but they could fit for a use in ethanol-diesel blends as they are already used to treat polluted soils with crude oil [107]. Synthetic surfactants or animal-derived surfactants are nowadays being replaced by biosurfactants as they are fully renewable and cheaper to produce.

Non-ionic surfactants are relatively non-toxic [108], produced from esters which makes them easy and cheap to produce and are already widely used to create ethanol-diesel blends. SPAN (hydrophobic) and TWEEN (hydrophilic) are non-ionic surfactants. They offer several advantages such as the increased stability, full biodegradability and are based on natural fatty acid and sugar alcohol sorbitol. These are the most commonly used surfactants to create emulsions between ethanol and diesel. SPAN is considered as a sorbitan ester and is produced by dehydration of sorbitol and by esterification with fatty acids. The chemical formula of SPAN 80 is  $C_{64}H_{124}O_{26}$ . TWEEN is ethoxylated span which means that ethylene oxide is added to the SPAN during a chemical reaction. The chemical formula of TWEEN 65 is  $C_{100}H_{194}O_{28}$ . When used for blending fuels, they allow reducing corrosion and act as emulsifiers to improve the water tolerance of the blend [109]. SPAN 80 is one of the most common non-ionic surfactants because its HLB value is close to the optimal value for ethanol-diesel blend which is located at around 4.3 [110]. Tables 6 show the properties of SPANS and TWEENS [111]. For mineral oils (diesel) SPANS 80 and higher are perfectly soluble. These surfactants could allow producing diesel-ethanol

blends with high-concentration of ethanol and therefore to ensure achieving even more promising results in decreasing exhaust gas emissions.

Table 6. HLB values and solubility of Span and Tween at 10% w/w in a number of common solvents and oils at 25° C [111].

Surfactant	HLB Value	Rapeseed Oil	Diesel	Water	Kerosene
Span 20	8.6	Partly Soluble	Soluble	Partly Soluble	Soluble
Span 40	6.7	Insoluble	Gel Formed	Partly Soluble	Insoluble
Span 60	4.7	Insoluble	Gel Formed	Partly Soluble	Insoluble
Span 80	4.3	Soluble	Soluble	Partly Soluble	Soluble
Span 83	3.7	Soluble	Soluble	Partly Soluble	Partly Soluble
Span 85	1.8	Soluble	Soluble	Partly Soluble	Soluble
Span 120	4.7	Soluble	Soluble	Partly Soluble	Partly Soluble
Tween 20	16.7	Insoluble	Insoluble	Partly Soluble	Insoluble
Tween 40	15.6	Insoluble	Insoluble	Soluble	Insoluble
Tween 60	14.9	Partly Soluble	Insoluble	Partly Soluble	Insoluble
Tween 65	10.5	Insoluble	Partly Soluble	Gel Formed	Partly Soluble
Tween 80	15	Partly Soluble	Insoluble	Soluble	Insoluble

#### 4.2 The effect of ethanol-diesel blends on the performance and emissions of combustion ignition engines

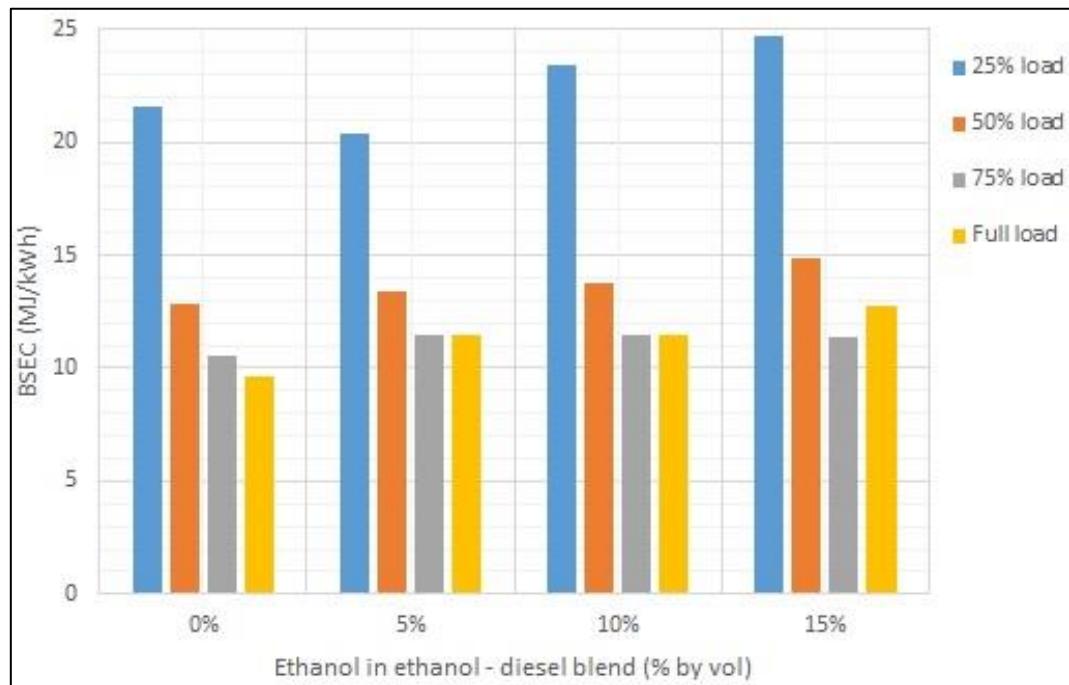
Ethanol-diesel blends have been studied for the last 25 years. The processes of combustion and exhaust gas emission for different ethanol-diesel blends were reviewed. To examine the overall performance of these engines, different studies with different ethanol concentrations, different additives and surfactants have been analysed. Hansdah et al. [100] performed experiments on diesel engine with ethanol-diesel emulsions up to 15% ethanol including the use of SPAN 80. They used ethanol based on Madhuca Indica flower, which has been emulsified with diesel using SPAN 80. They tested 3 different diesel-ethanol blends of 5%, 10% and 15% on a Kirloskar TAF1 single cylinder 4 stroke diesel engine with a rated power of 4.4 kW at 1500 rpm to analyse the engine performance and emissions. Table 7 shows the properties for different fuel blends used for these experiments.

Table 7: Physical properties of diesel, ethanol and its emulsions [100]

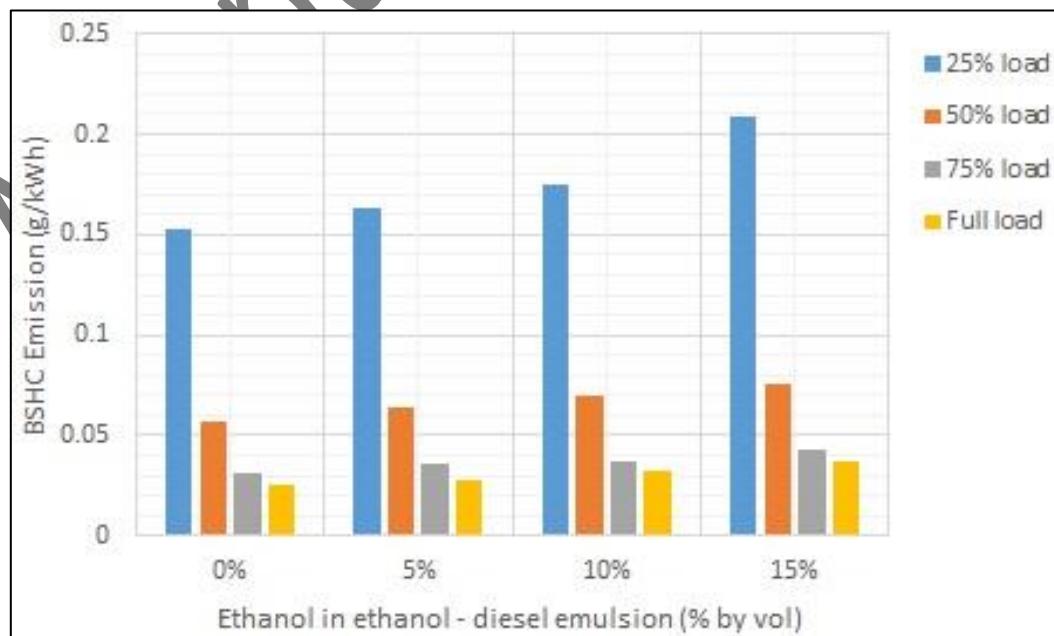
Properties	ASTM standard	Diesel	Ethanol	BMDE5	BMDE10	BMDE15
Lower heating value (MJ/kg)	D 4809	43.8	29.38	38.21	37.02	35.34
Flash point (°C)	D 2500	49	24	30	29	26
Pour point (°C)	D 97	-15	-103	-9	-12	-36
Boiling point (°C)	D 7169-11	180–360	80	165–342	142–326	114–298
Kinematic viscosity at 40 °C (cSt)	D 445	2.58	1.73	2.31	2.01	1.95
Surface tension at 20 °C (N/m)	D3825	N/A	N/A	0.016	0.019	0.02

The results displayed an increase in the combustion pressure and a peak appearing later for diesel when the concentration of ethanol increased. They also showed that the long ignition delay is as a result of the high concentration of ethanol. This increase in both combustion pressure and ignition delay could be associated with low viscosity and boiling point of ethanol, therefore, due to the increased ignition delay, fuel is better premixed with the in-cylinder air which improves the rate of heat release of the engine due to a more complete combustion. The improvement of the combustion also affects the engine performance with an increase in the BSEC as seen in Fig. 8 (a), and a better efficiency of the engine when 15% ethanol was used. However, the engine performance improvement does have side effects on the emissions. For example, an increase in the

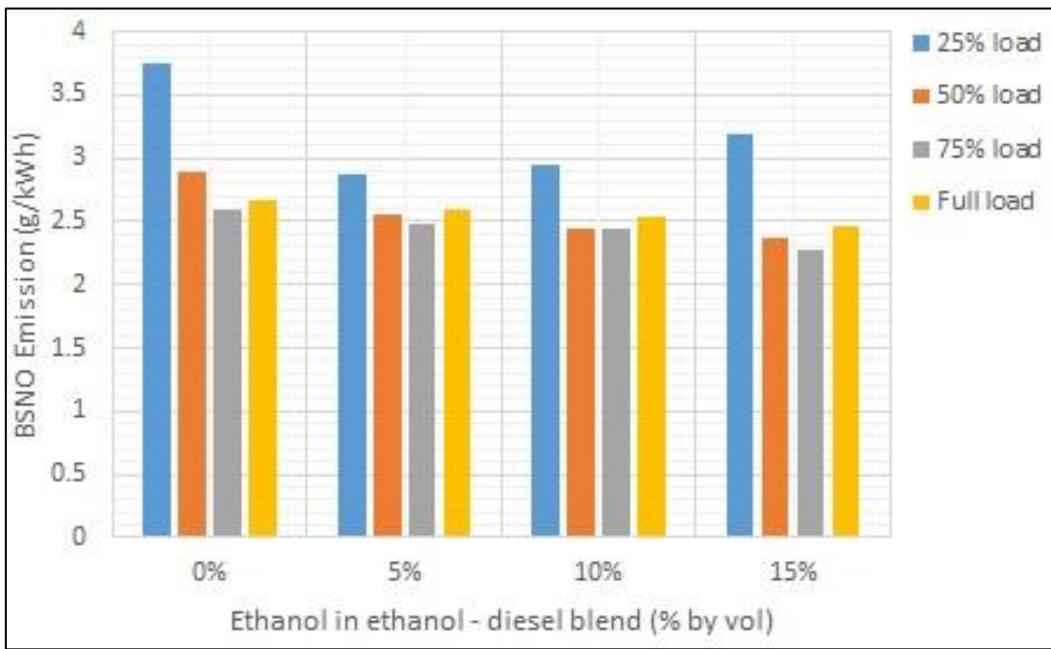
heat of evaporation with the increase of ethanol concentration resulted in an increase of HC by 22% in comparison to neat diesel for a blend of 15% ethanol at full load. NO<sub>x</sub> emissions decrease more than 4% and smoke emissions by 20% at full load, as illustrated in Fig. 8 (b), (c) and (d), respectively. Therefore, it can be concluded that these results were quite promising despite a slight increase in HC emissions.



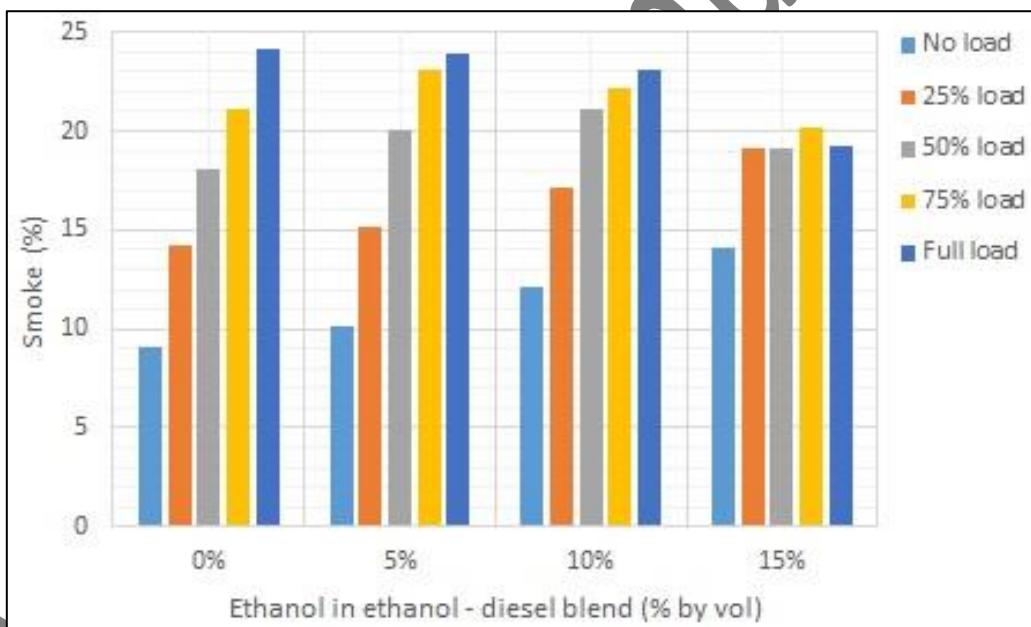
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(b)



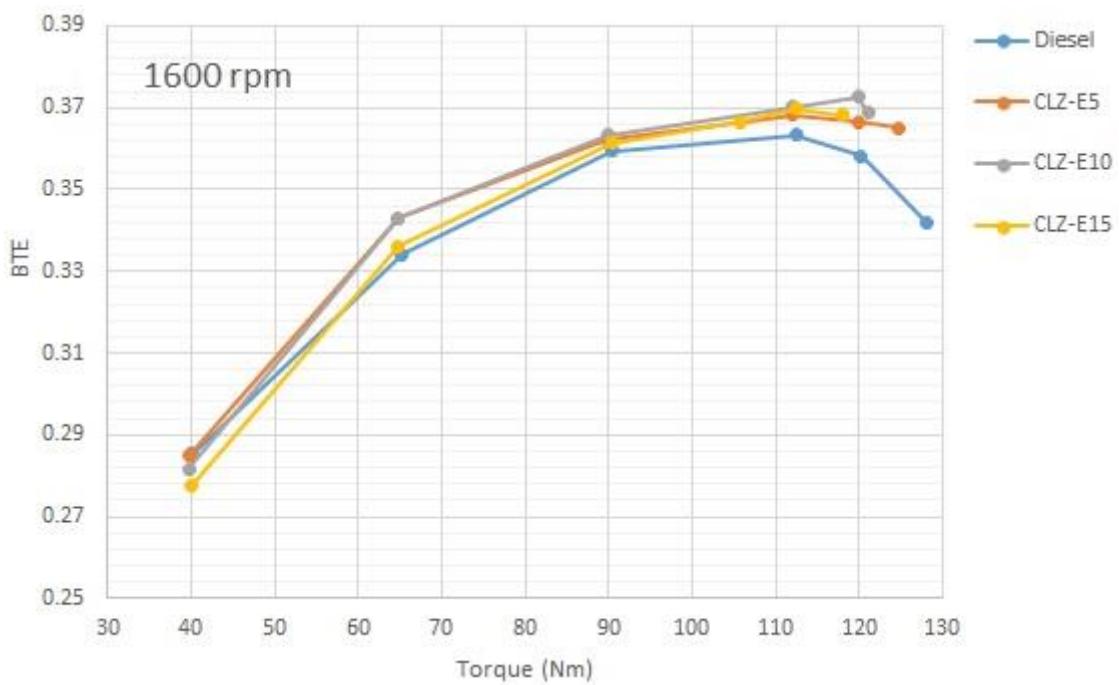
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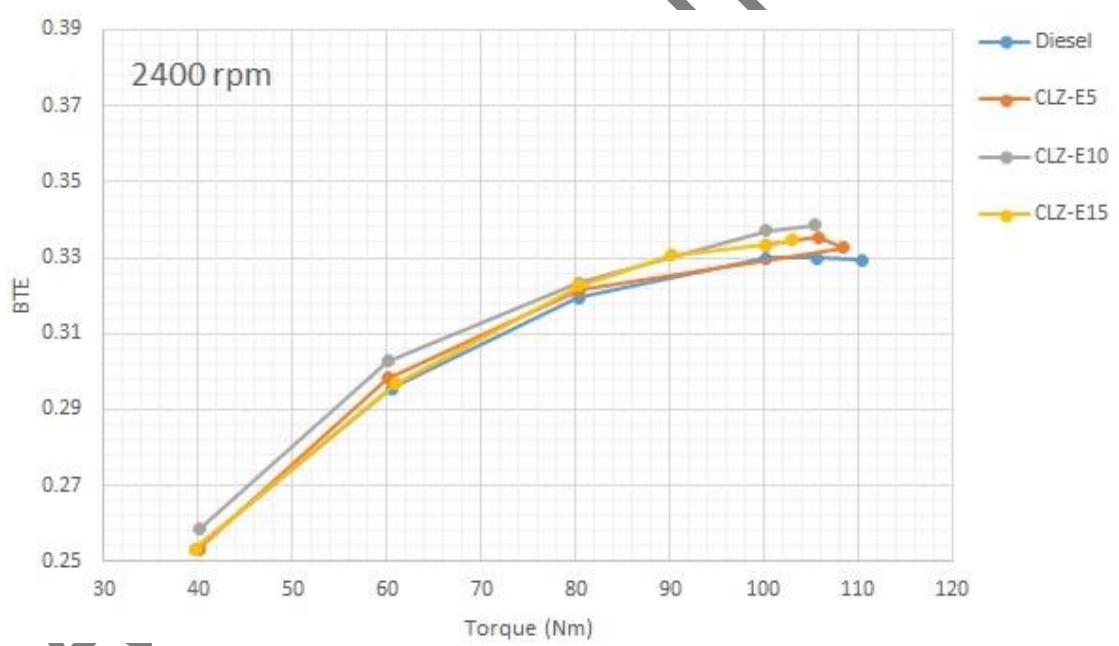
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Fig. 8. Brake specific efficiency of combustion and emissions variations for the different diesel-ethanol blends at different loads for: (a) BSEC, (b) unburnt hydrocarbons (HC); (c) nitric oxides (NO); (d) smoke [100]

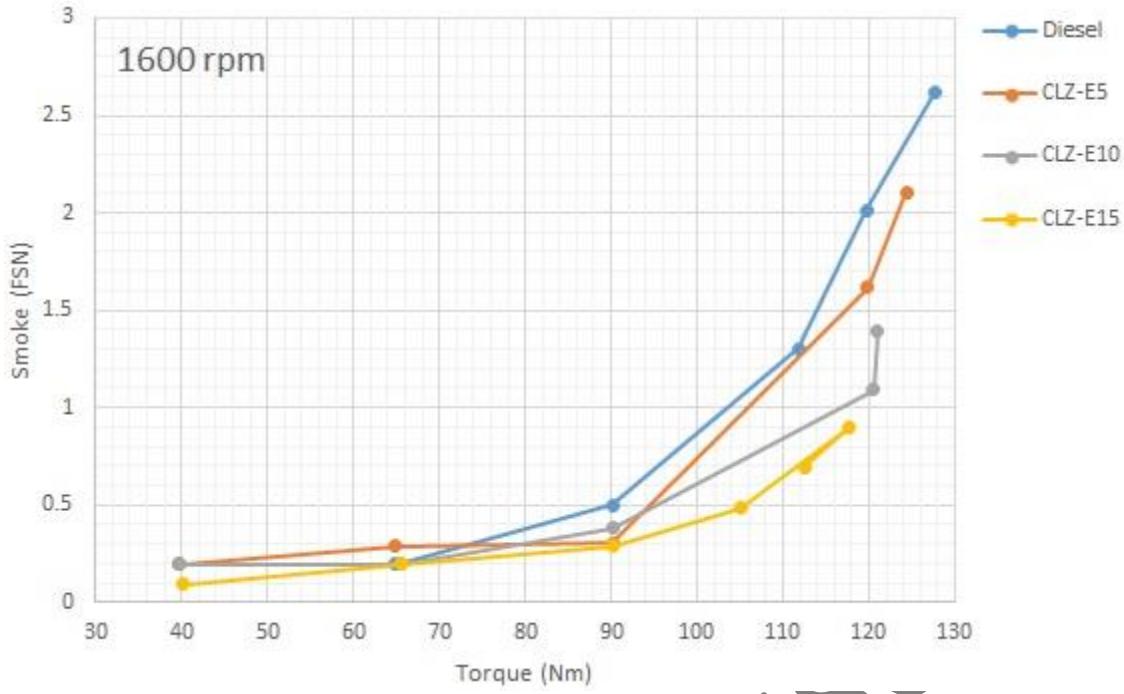
Lei et al. [112] studied the use of CLZ emulsifier with ethanol-diesel blends and analysed its effect on performance and emissions of a CI engine with up to 15% ethanol. They ran experiments with three different blends 5%, 10% and 15% ethanol on a traditional 4 cylinders, 4 strokes CI engine at two different speeds of 1600 rpm and 2400 rpm. The experiments showed that the break thermal efficiency (BTE) of diesel-ethanol blend is higher than that of neat diesel for two different loads, as shown in Figs. 9 (a) and (b). It can be explained by the high oxygen contents in the blend allowing a better combustion. However, they found out that the highest BTE was seen with the blend containing 10% ethanol meaning that the optimal ratio of ethanol and diesel for this engine would be around 10% and that adding more ethanol would not improve it.



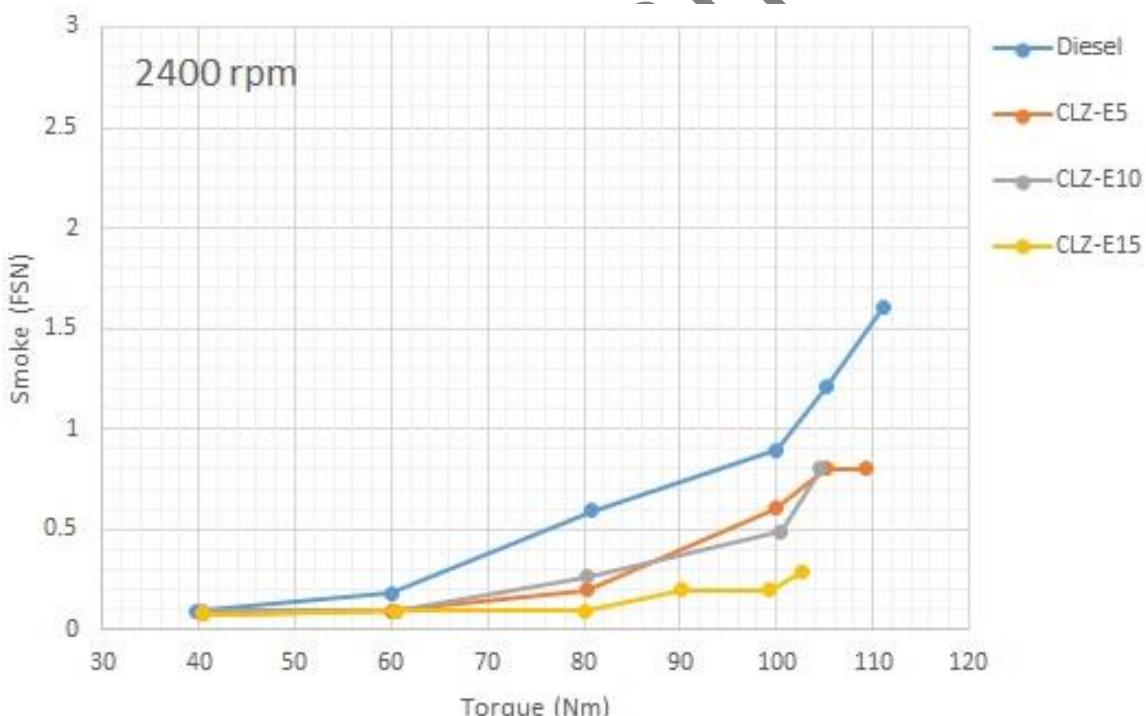
(a)



(b)



(c)



(d)

Fig. 9. Performance and emissions of diesel engine fuelled with diesel-ethanol fuel blends, (a) BTE at 1600 rpm, (b) BTE at 2400 rpm, (c) smoke at 1600 rpm, (d) smoke at 2400 rpm [112]

It has been found that CO emissions increase at low loads and decrease at high loads for blends compared to neat diesel. CO emissions increased at low loads due to high latent heat of vaporization of ethanol that reduces the temperature of the cylinder and prevents the oxidation of CO. However, the high temperature and oxygen content of ethanol improves combustion at high speed and heavy loads, and therefore, the oxidation of CO tends to decrease. As for Hansdah et al. [100], minor decrease was observed in NO<sub>x</sub>

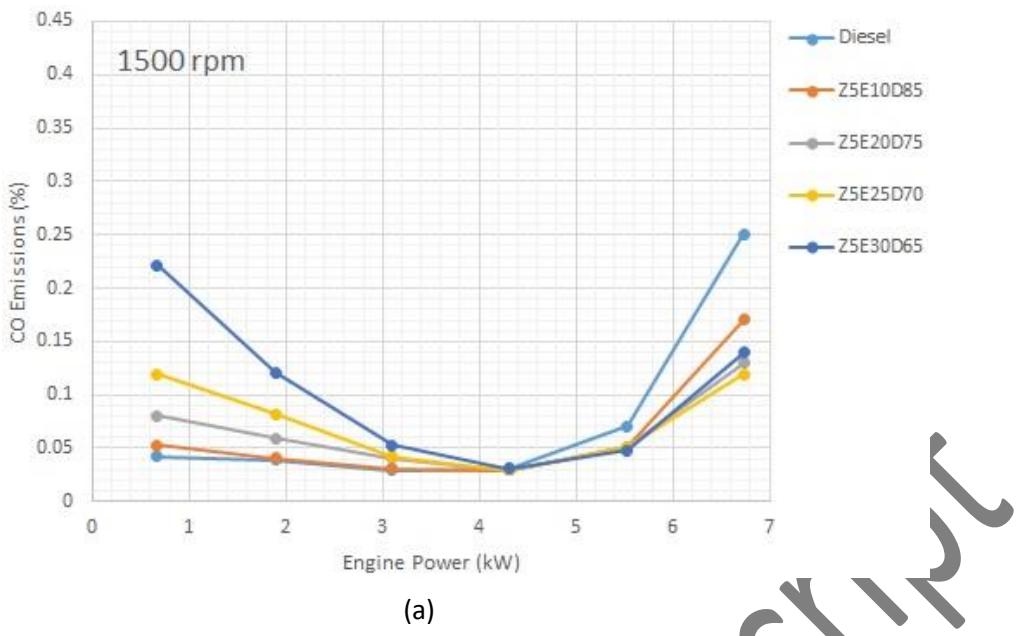
emissions at low and high loads but the speed have no impact the NO<sub>x</sub> emissions. Due to the same reasons, as for CO emissions, the reduced in-cylinder temperature led to an increase in HC emissions for the ethanol-diesel blends. Specifically, at heavy loads and high speeds the use of ethanol helps to reduce smoke emission as illustrated in Figs. 9 (c) and (d).

Huang et al. [113] undertook a different approach than the previous studies and used 5% of butanol in the solution instead of an emulsifier to stabilise the blend. The results showed that the blends lasted between 11 and 14 days before were separated. Then, they ran experiments with four different blends containing respectively 10%, 20%, 25% and 30% of ethanol at different engine speeds of 1500 rpm and of 2000 rpm on a S195 4-stroke single cylinder engine. Table 8 summarizes the fuel properties used in these experiments.

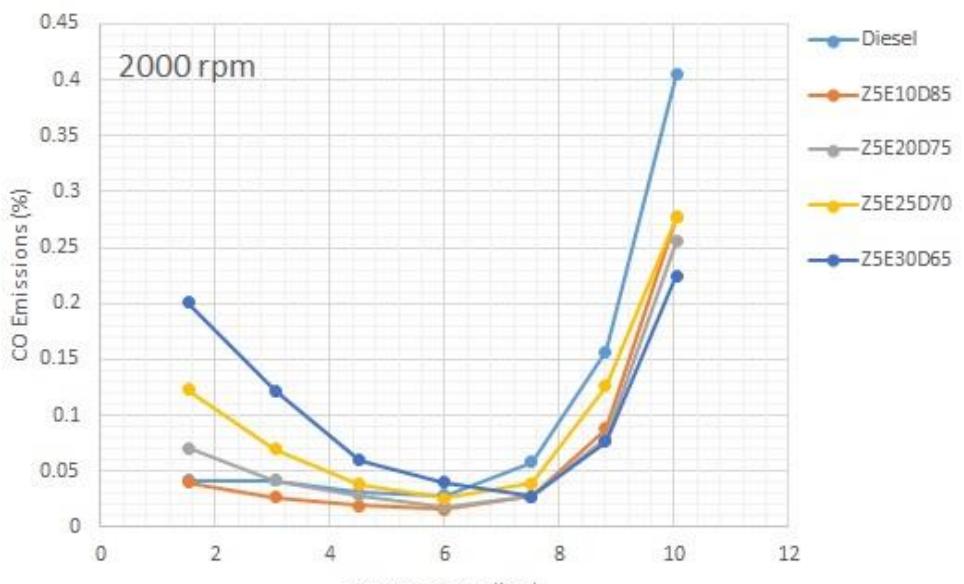
Table 8. Properties of diesel, ethanol and n-butanol [113]

	Formula	Molecular weight	Density at 20 °C	Oxygen content (wt %)	Carbon content (wt %)	Hydrogen content (wt %)	Viscosity at 20 °C (10 <sup>-6</sup> v/m <sup>2</sup> /s)	Cetane number	Lower heat value (MJ/kg)
Diesel	C <sub>12</sub> H <sub>23</sub>	190–220	0.84	0	~86	~14	3.35	40~50	42.5
Ethanol	C <sub>2</sub> H <sub>5</sub> OH	46.07	0.789	34.8	52.2	13	1.2	8~9	26.4
n-Butanol	C <sub>4</sub> H <sub>10</sub> O	74.12	0.81	21.58	64.82	13.6	3	17	33.2

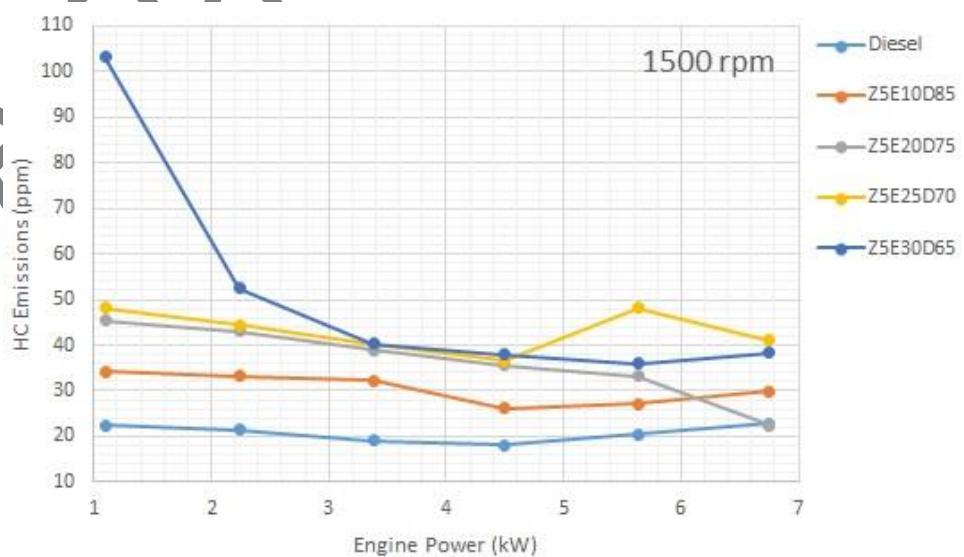
BSFC increased with the increasing amount of ethanol to a maximum 31.5% with the blend containing 30% of ethanol. At low loads, thermal efficiency of the ethanol blends was lower than neat diesel but higher at high loads. This is due to the reduced temperature in the cylinder at low loads. Therefore, it is not surprising to see that CO emissions shown in Figs. 10 (a) and (b) increased at low loads with a reduction at high loads and were much lower for fuel blends rather than for neat diesel with a maximum decrease of almost 44% at high loads and high speed for the blend with 30% of ethanol content. Fig. 10 (c) and (d), showed higher HC emissions at low loads for the blends than for neat diesel but once again, at high loads they become less important than for neat diesel to become almost nil for the blend with 20% ethanol. This is explained again by the high cylinder temperature of the blend at high loads due to the higher oxygen content of ethanol. The test results show a general decrease in NO<sub>x</sub> emissions in Figs. 10 (e) and (f) and especially at lower loads due to the low cylinder temperature with a maximum reduction of 59% for the highest blend at low loads. Smoke emissions in Figs. 10 (g) and (h) show again impressive results for the blends with a decrease up to 87% for 30% ethanol blend. Zheng et al. [114] studied the effect of two-stage injection on combustion and emissions under high EGR rate using blends of diesel/gasoline, diesel/n-butanol, diesel/gasoline/n-butanol. They found that blending gasoline or/and n-butanol in diesel improves smoke emissions while induces increase in maximum pressure rise rate. The variation in fuel properties caused by blending gasoline or/and n-butanol into diesel does not impose obvious influence on post combustion. To conclude, the use of butanol instead of surfactants shows interesting results and it could be useful to investigate more on this topic.



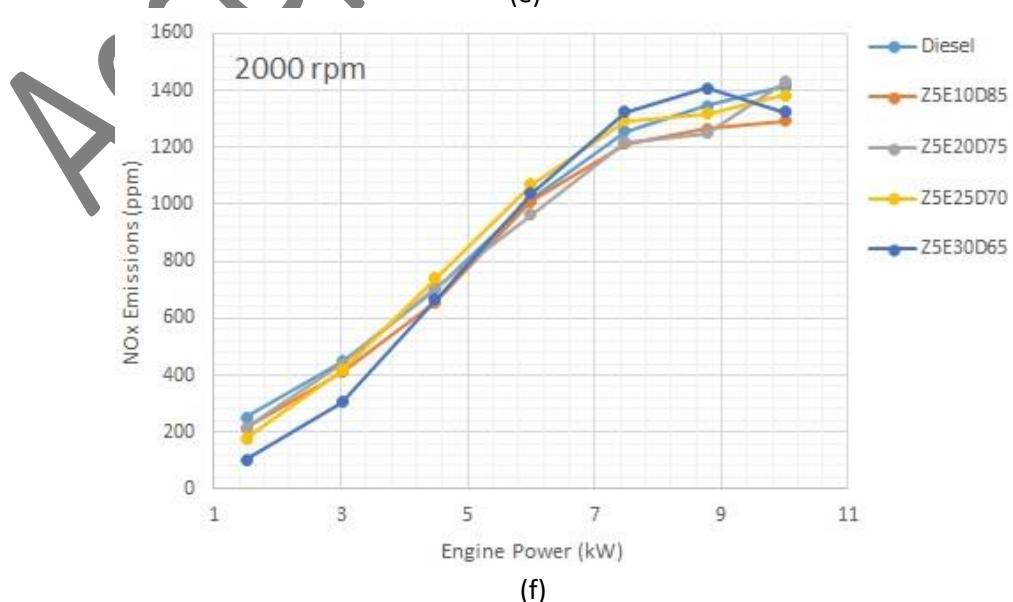
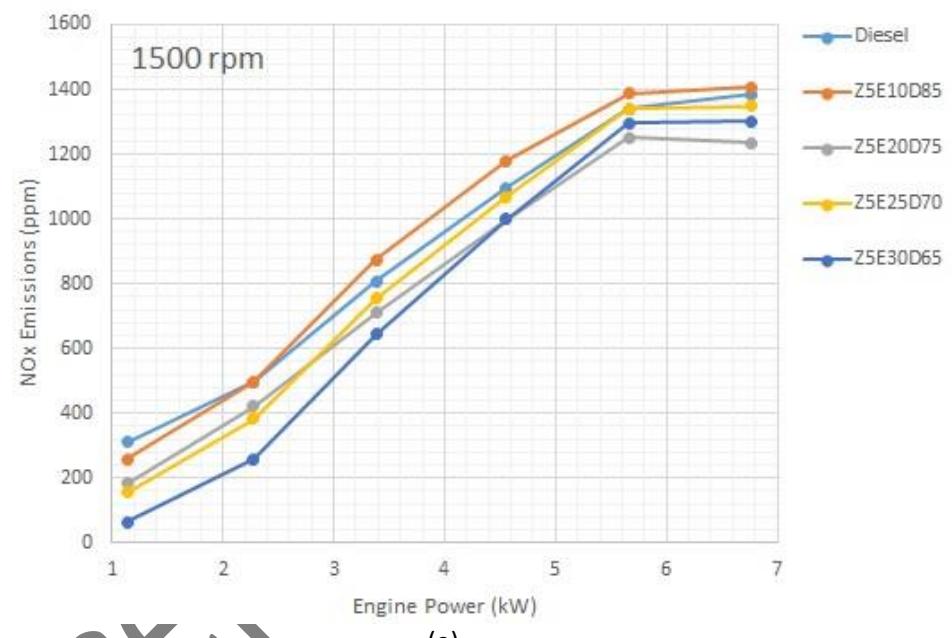
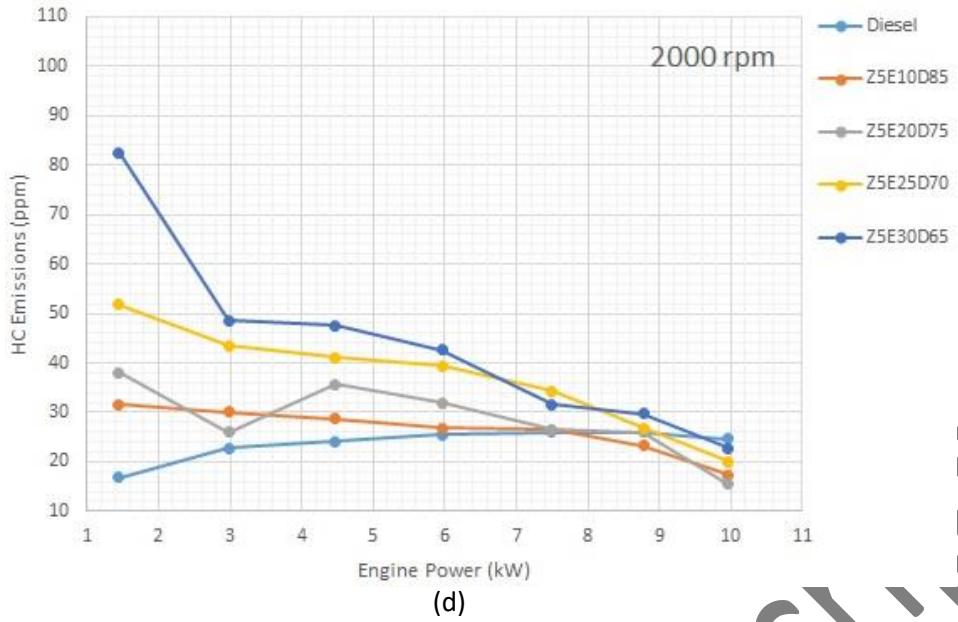
(a)



(b)



(c)



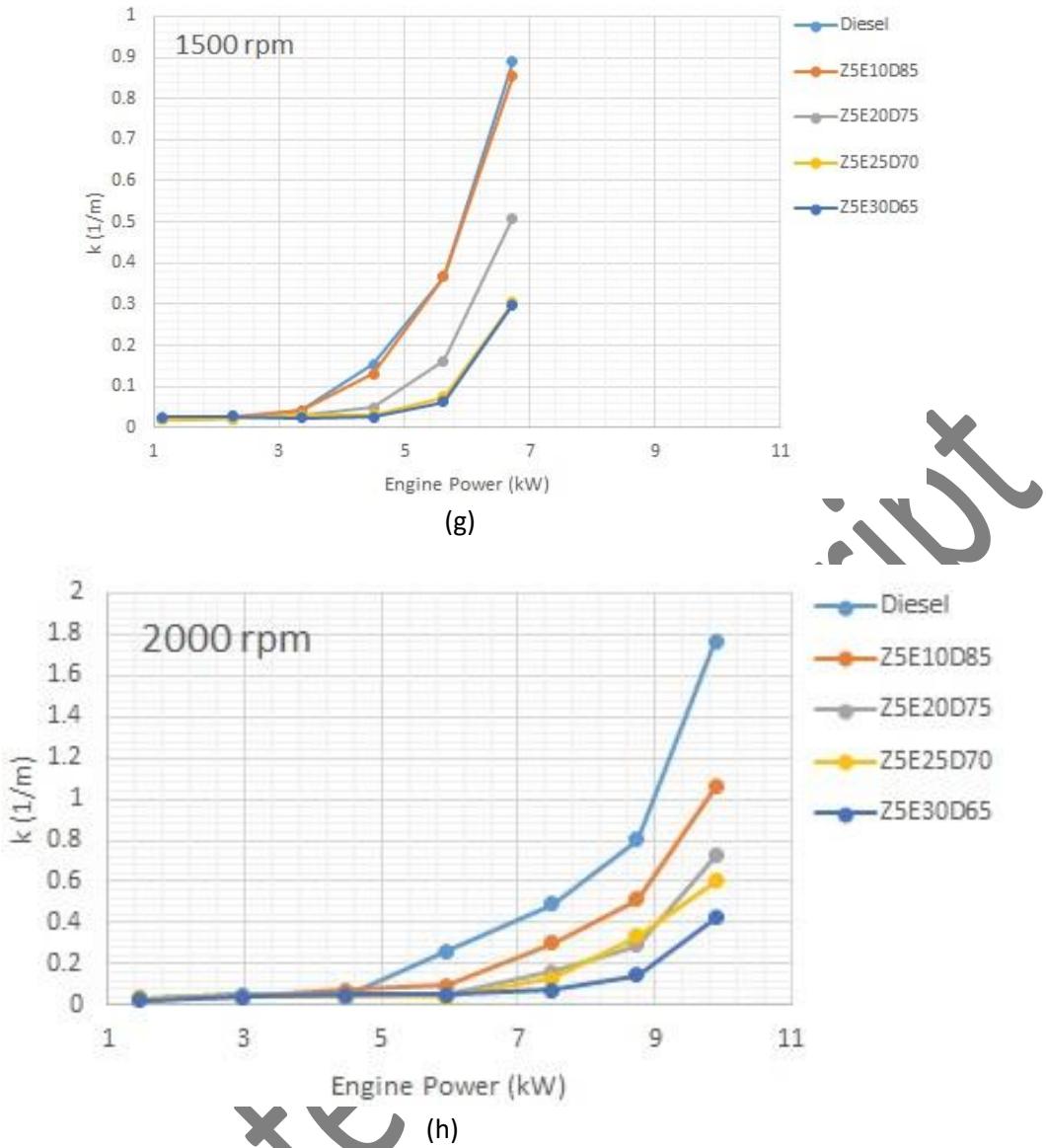


Fig. 10. Engine performance, (a) CO at 1500 rpm, (b) CO at 2000 rpm; (c) HC at 1500 rpm, (d) HC at 2000 rpm, (e) NOx at 1500 rpm, (f) NOx at 2000 rpm, (g) smoke at 1500 rpm, (h) smoke at 2000 rpm [113]

Jin et al. [115] investigated the effects of C3–C5 alcohols on solubility of alcohols/diesel blends. They found that all tested alcohols in their study could blend with diesel at any ratio without water addition. For *n*-propanol, iso-propanol and *tert*-butanol, which were miscible with water, the allowable water concentration was gradually increased with the increase of temperature. But for *n*-butanol, *iso*-butanol, *sec*-butanol, *n*-pentanol, *iso*-pentanol, and *tert*-pentanol, the water concentration admitted in the system gradually decreased with the increase of temperature.

Parthasarathi et al. [116] conducted experiments with diesel-ethanol blends containing up to 40% ethanol on a single cylinder 4 stroke engine of 5.2 kW and at a rated speed of 1500 rpm. To stabilise the blend, they used surfactant called Benzal Konium Chloride ( $C_6H_5CH_2N(CH_3)_2RCI$ ) which have an advantage to provide good solubility properties

when used with ethanol. They tested four different blends each containing 10%, 20%, 30% and 40% of ethanol. Table 9 shows properties for fuel blend with 40% ethanol.

Table 9. Physical chemical properties of diesel, ethanol and D50E40 [116]

Properties	Diesel	Ethanol	D50E40
Lower heating value (MJ/kg)	42.8	26.6	39.1
Flash point (°C)	74	13	13
Cetane number	50	5.8	46
Kinematic viscosity at 40 °C (cSt)	3.4	1.2	2.84
Density at 15 °C in gm/cc	0.83	0.79	0.82

Although, in some studies by other authors BSFC was always higher for the ethanol blends than for neat diesel at any loads, in this study, BSFC is only higher at high loads with a maximum observed for the blends containing 30% of ethanol. Break thermal efficiency in Fig. 11 was high for the blends compared to neat diesel with a maximum of almost 36% for the blend containing 40% of ethanol. The ethanol composition of the blends influences the increase in cylinder pressure and rate of heat release. This rate of heat release significantly increased for the D50E40 blends with a maximum of 162 kJ/m<sup>3</sup> compared to 121.2 kJ/m<sup>3</sup> for neat diesel.

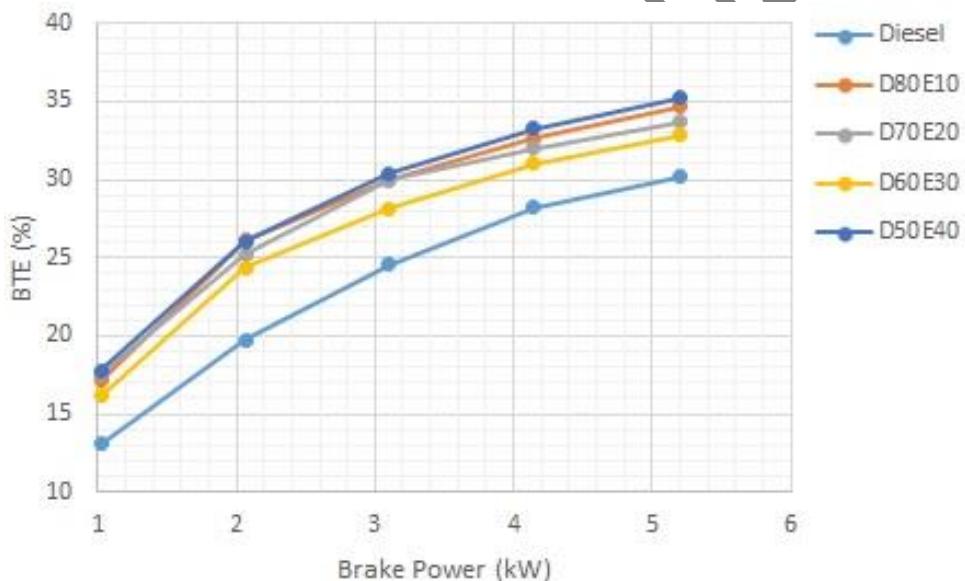


Fig. 11. Break thermal efficiency at different loads of the ethanol-diesel blends [116]

HC emissions significantly increased with increased loads and showed higher values for blends than for neat diesel at medium loads. For the blends containing 40% ethanol, HC emissions increased almost twice than neat diesel at high loads. NO<sub>x</sub> emissions increased as the load increases and were higher for the blends compared to those of neat diesel with a maximum of 612 ppm for the D50E40 blend. Smoke emissions reduced significantly with the addition of ethanol. This study showed promising results for engine thermal efficiency and smoke emissions, but solutions still need to be found to mitigate HC and NO<sub>x</sub> emissions for the blends. These could eventually be improved by using cetane improving additives in the blend.

Ashok et al. [117] tested different cetane improving additives (H<sub>2</sub>O<sub>2</sub>, DEE, DME) with blends of 50% ethanol and 50% diesel emulsified with TWEEN 80. In this configuration, the blend remained stable during approximatively 3 days. The properties of those additives are shown in Table 10.

Table 10. Properties of cetane improving additives, DME, DEE and H<sub>2</sub>O<sub>2</sub> [117]

	Dimethyl ether (DME)	Diethyl ether (DEE)	Hydrogen peroxide
Chemical formula	CH <sub>3</sub> OCH <sub>3</sub>	C <sub>4</sub> H <sub>10</sub> O	H <sub>2</sub> O <sub>2</sub>
Boiling point (°C)	-24.8	34.4	155.5
Cetane number	>125	>87	>56
Self ignition temperature (°C)	125	154	160
Stoichiometric air/fuel ratio (wt/wt)	10.9	11.1	11.2
Lower heating value (kJ/kg)	33.9	36.5	41.25
Specific gravity	1.59	0.714	1.4
Density (kg/m <sup>3</sup> )	734.7	713.4	1442.5

The experiments were conducted at 1500 rpm on a single cylinder diesel with a rated power of 5.4 kW. The results showed increase in the BTE, Fig. 12, due to the addition of cetane improvers in the blend which resulted in a lower self-ignition temperature. Therefore, a longer ignition delay with DME showed the greater results with total 37.8% of efficiency at high loads. Addition of cetane improving additives to the blend reduced the NO<sub>x</sub> emissions. It could be interesting to further investigate the impact of cetane improving additives on ethanol-diesel blends as this has shown promising results.

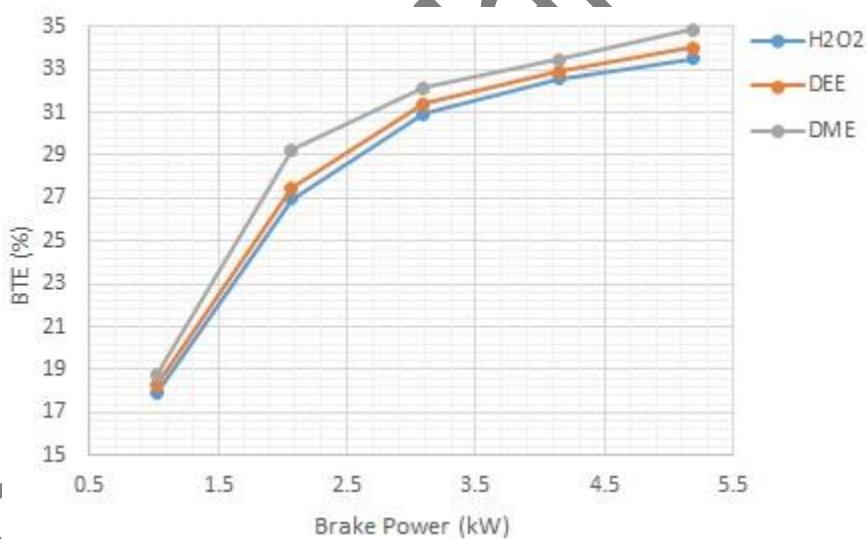


Fig. 12. Brake thermal efficiency [117]

Liu et al. [118] added Polyoxymethylene dimethyl ethers (H<sub>3</sub>CO(CH<sub>2</sub>O)<sub>n</sub>CH<sub>3</sub>) (PODE) in an ethanol-diesel blend and conducted experiments on a heavy duty CI engine. PODE was added to the blend due the high cetane number and clean combustion properties. Three different ethanol blends have been tested: 1) DPE5 (diesel 75%, PODE 20% and ethanol 5%), 2) DPE10 (diesel 70%, PODE 20% and ethanol 10%), 3) DPE15 (diesel 65%, PODE 20% and ethanol 15%). This tests were performed on a six-cylinder diesel engine that produce 243 kW at 2200 rpm. The properties of the tested blends are displayed in Table 11.

Table 11. Properties of DPE5, DPE10 and DPE15 [118]

Properties	DPE5	DPE10	DPE15
			25

Density (kg/m <sup>3</sup> at 20 °C)	874.5	872.1	869.9
Cetane number	56.2	54	51.9
Lower heating value (MJ/kg)	37.07	36.34	35.61
Oxygen content (wt %)	13.05	14.66	16.28

Using ethanol-diesel blends in this experiment helped to reduce HC and CO emissions compared to neat diesel. At high loads, CO can be completely oxidized to CO<sub>2</sub>. It is however interesting to note that more ethanol concentration in the blend caused lower CO<sub>2</sub> emissions. Therefore reducing this oxidation process will in turn reduce CO formation. Soot and NO<sub>x</sub> emissions have been investigated as well. The results showed an increase in NO<sub>x</sub> emissions with higher loads and unfortunately higher values of NO<sub>x</sub> for blend than neat diesel as illustrated in Fig. 13 (a). Soot emissions showed lower values compared to neat diesel with a decrease of 85.7% at the highest load for DPE15 as shown in Fig. 13 (b).

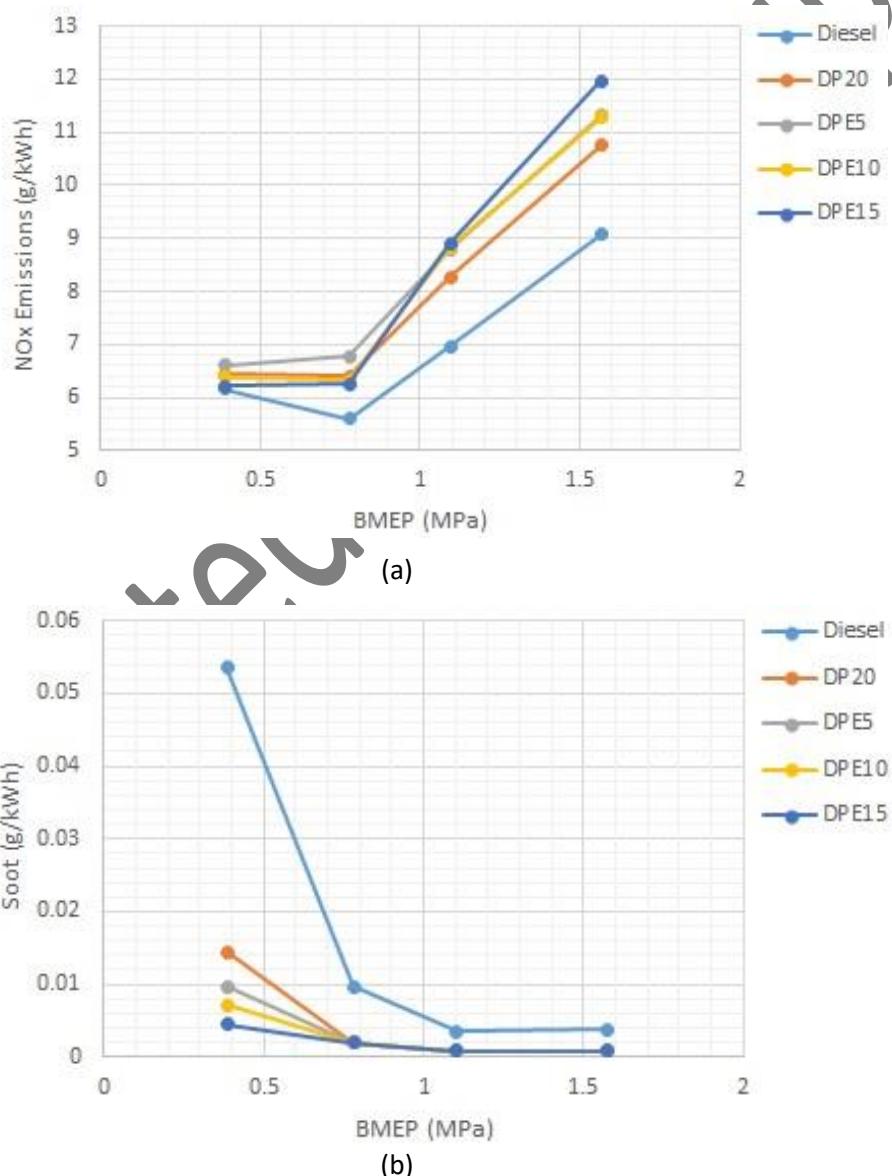
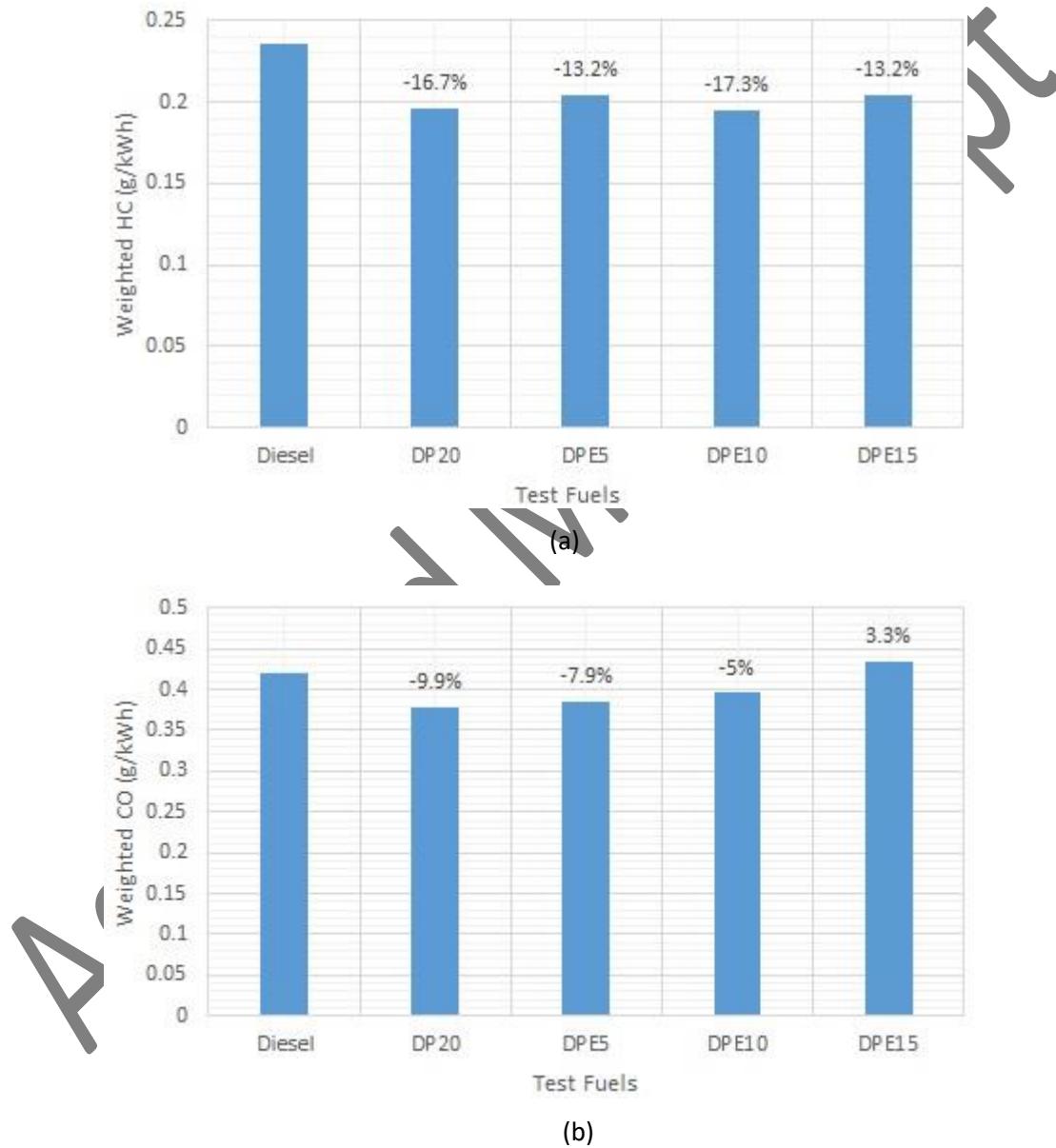
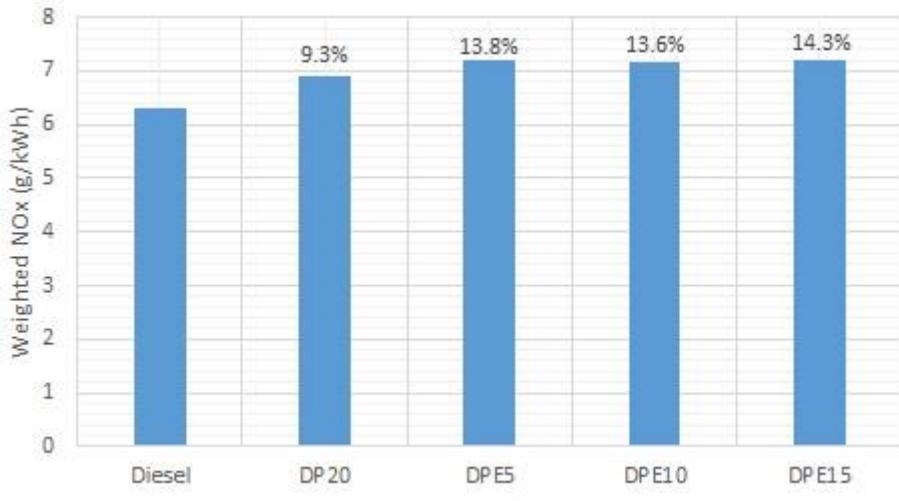


Fig. 13. Soot and NO<sub>x</sub> emissions characteristics for different ethanol-diesel blends at different loads [118]

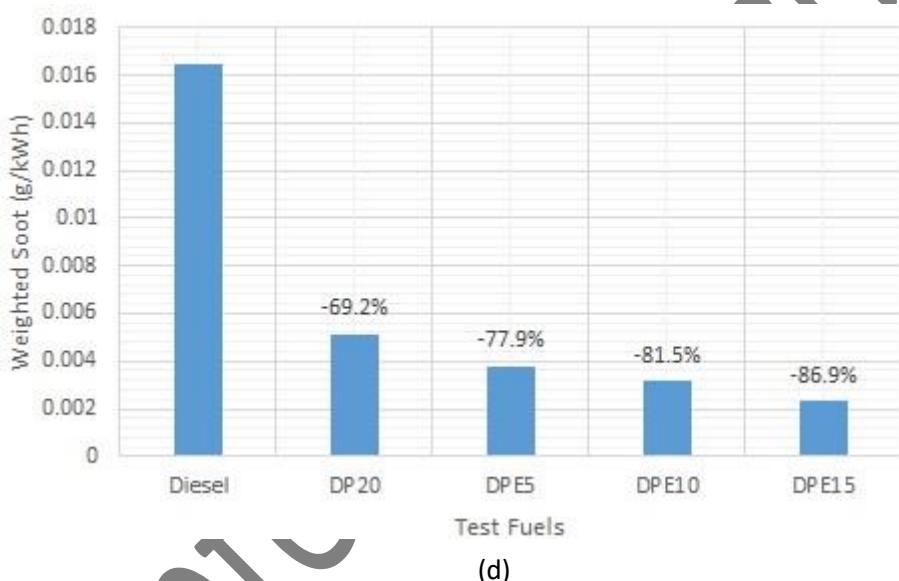
The tests on a Euro IV engine with the blend have been studied for the Euro VI World Harmonized Stationary Cycle (WHSC). The results showed the weighted BSFC for the blends is higher than neat diesel by 21% for the DPE15. All the tested blends showed

decrease in HC emissions when compared to diesel with the largest reduction of 17.3% for DPE10, as illustrated in Fig. 14 (a). CO emissions followed the same trend but CO for the DPE15 blend increased 3.3%, as shown in Fig. 14 (b). This is due to ethanol's high oxygen content, PODE and low LHV resulting in the reduction of combustion temperature and therefore decreased the quality of the CO oxidation under cold idle. Weighted emissions of NO<sub>x</sub> were increased for the blend and the highest increase of 14.3% was monitored for DPE15, as shown in Fig. 14 (c). Fig. 14 (d) illustrates that the WHSC test cycle shows impressive decrease in soot emissions with a maximum decrease of 86.9% for the DPE15, which could be associated with the high composition of oxygen in the blend. Nevertheless, experiments showed promising results on using ethanol blends in HDV engines. However, a solution to improve NO<sub>x</sub> emissions still needs to be found.





(c)



(d)

Fig. 14. Weighted gaseous emissions of the engine for the WSHC test cycle. (a) HC, (b) CO, (c) NOx, (d) Soot, [118]

No clear trend is visible for NO<sub>x</sub> emissions even if small decreases have been reported by other authors. The same conclusion can be made about HC emissions. However, Huang et al. [113] showed great decreases in HC emissions with their blends. Smoke and CO showed good results with a reduction of almost 90% when compared to neat diesel for smoke and almost 45% for CO. Therefore, overall we can see that ethanol-diesel blends are helping to mitigate in an effective way gaseous emissions and some studies have shown great results which are promising for the future. However, certain parameters need to be considered as every study has been performed under different conditions such as the engine specification, the type of additives and surfactants used and especially the composition of the ethanol and its cetane number [119].

#### 4.3 The blend wall of ethanol and the use of surfactants

Over the last few years many automotive manufacturers and governments admitted that blending over 20% of ethanol was not feasible due to a few problems such as the phase separation and the engine compatibility for high-ethanol concentration without engine modifications. Furthermore, as it was mentioned earlier, we know how to use blends with higher concentration of ethanol in CI engines without required modifications and many studies

with up to 30% ethanol used in the blends can be found. But due to the problem with phase separation in the blend, very few studies investigated the blends with ethanol concentrations above 40-50%.

We conducted experimental studies with the aim to achieve high concentration ethanol-diesel blends and obtained promising results, which may lead to new perspectives for application in heavy-duty transport. The stability assessment for these blends was analysed visually, while the samples were kept in closed glass bottles under controlled temperature after mechanical agitation at ambient laboratory conditions. The blends were prepared in airtight bottles and observed for extended period of time for any occurring phase separation. During observations, unstable blends changed into separate phases, some of which were not visible to the naked eye, hence we used UV light to identify the phase separation boundaries.

- *Diesel-ethanol blend (70%-30%)*

This blend was prepared as a starting condition due to the fact that no phase separation was observed for diesel-ethanol blends below 30% ethanol composition. No apparent phase separation was observed during the first week, but by the end of the second week, the two phases have been separated as illustrated in Fig. 15 (a). Different surfactant application processes were carried out in order to decide which surfactant or combination of surfactants would give the best result and at what quantity. This was done by adding Span 80, Span 85, as well as the blend of both surfactants to the phase separated blends in the composition of 2% and 3% consequently, followed by agitation. 3% composition of the surfactant blend of Span 80 and Span 85 gave the best result on stability. The microscopic analysis in Fig. 15 illustrates (b) that the blends with separated phases tend to have bigger droplets sizes, and this was as a result of droplets merging together to form a bigger body. The addition of surfactant followed by agitation was observed to reduce the size of these droplets, as shown in Fig. 15 (c), and create a more evenly distributed disperse phase with significantly reduced droplet sizes from the initial range of 0.26-24.22  $\mu\text{m}$  to 0.26-13.90  $\mu\text{m}$ . Both cases show how the droplet sizes were distributed in both blends without and with addition of surfactants. The blend without surfactant in Fig. 15 (b) has most of its droplet size distribution between the range of 0.26-2.00  $\mu\text{m}$ , while the blend with surfactant in Fig. 15 (c) has its droplet size distribution in the range 0.26-1.00  $\mu\text{m}$ . This shows the effect of the surfactant to reduce concentrated droplets which could cause the phase separation resulting in blend instability.

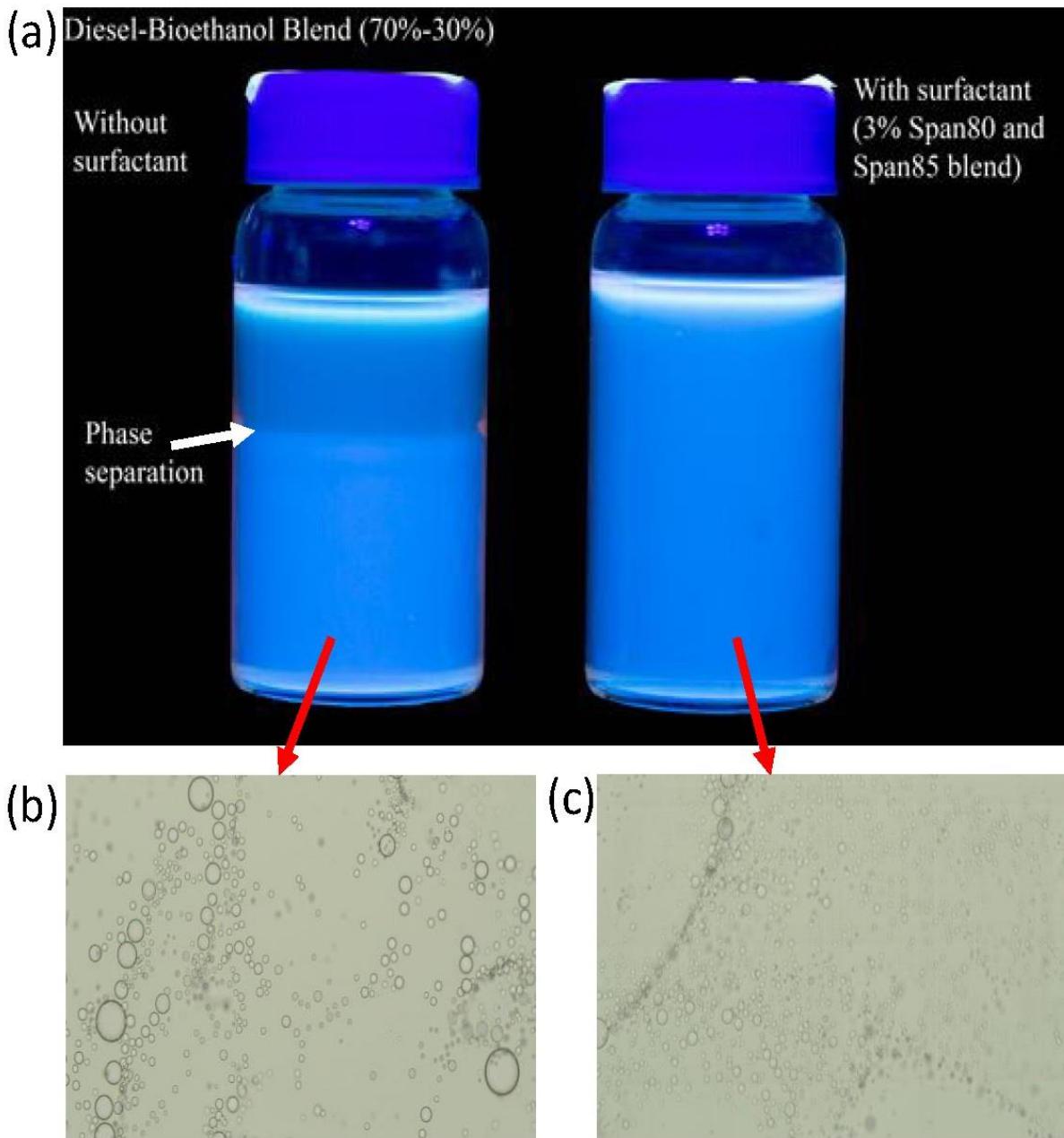


Fig. 15. The effect of SPAN on diesel-ethanol blend (70%-30%). (a) fuel blends before (left image) and after (right image) surfactant application; Microscopic image of the blend, (b) without surfactant, (c) with surfactant;

#### *- Diesel-ethanol blend (60%-40%)*

This blend was tested in order to increase the ethanol composition in the blending structure. After blending, the phase separation illustrated in Fig. 16 (a) was observed after 48 hours. This shorter period for phase separation is associated with the high ethanol composition in the blend. 3% blend of both Span 80 and Span 85 was applied before agitation, which resulted in a stable blend. The microscopic images of the blend before the addition of surfactant as shown in Fig. 16 (b) displayed the ethanol droplets merging together to form bigger droplets which affect the phase separation. It was observed from the second microscopic image in Fig. 16 (c) that a stable blend was formed as a result of the reduction in the initial droplet sizes due to the presence of surfactant. This could be associated with the reduction of the interfacial tension between both phases by the surfactant, which results in a more stabilised droplet network structure. The distributions in Figs. 16 (b) and (c) show the effect of the surfactant on the reduction of droplet size from the range of 0.26-25.30  $\mu\text{m}$  to 0.26-13.95  $\mu\text{m}$ , with more droplets distributed between 0.26-1.00  $\mu\text{m}$  range in the blend with surfactant.

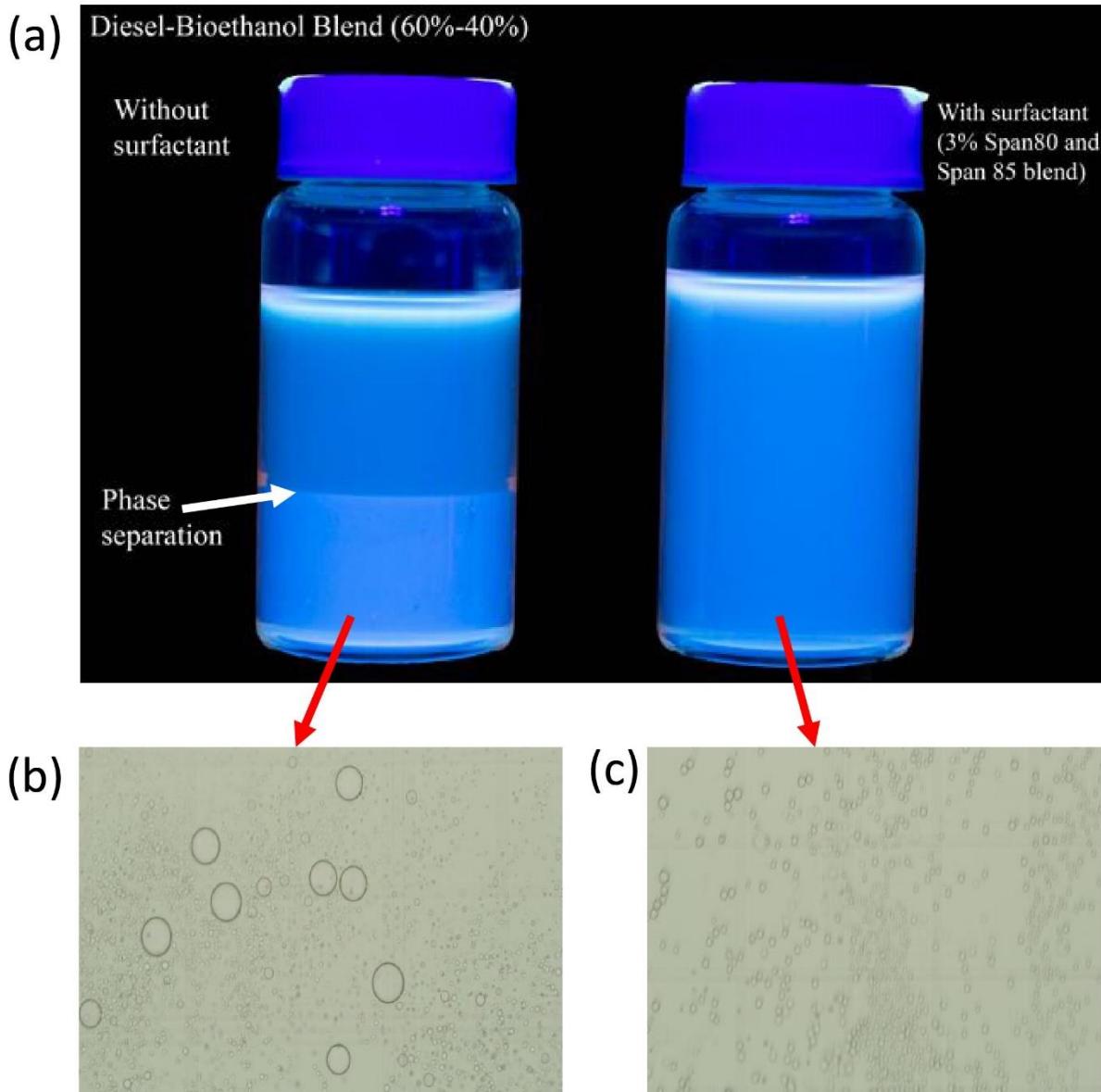


Fig. 16. The effect of SPAN on diesel-ethanol blend (60%-40%). (a) fuel blends before (left image) and after (right image) surfactant application; Microscopic image of the blend, (b) without surfactant, (c) with surfactant;

#### - Diesel-ethanol blend (50%-50%)

This particular blend is the mid-point of the diesel-ethanol blending experiment and it was very hard to achieve stability. The phase separation shown in Fig. 17 (a) was observed 2 hours after the blending operation. 50% concentration of ethanol in the blend really affected stability to the extent that it took 5% blend of Span 80 and Span 85 to achieve a stable fuel blend. The microscopic image of the blend before surfactant application, as shown in Fig. 17 (b), displayed bigger ethanol droplets as a result of the creaming process which leads to the phase separation. The other microscopic image with surfactant, shown in Fig. 17 (c), displayed evenly distributed smaller droplets. This could be associated with blend stability in the way ethanol droplets are dispersed in the continuous diesel phase, with enough rooms for droplets suspension. Stability was further demonstrated with a reduction in the initial droplet sizes from the range of 0.26-29.21  $\mu\text{m}$  to 0.26-22.91  $\mu\text{m}$  with more droplets distributed between 0.26-2.00  $\mu\text{m}$  range in the blend with surfactant.

From this experiment, it was observed that increasing the amount of ethanol beyond 50% of composition will definitely change the current water-in-oil emulsion type to oil-in-water, thereby increasing the lubricity of the blend. This is due to the hydrophilic nature of ethanol

which resulted in the decrease in the viscosity value in the stabilization zone [120]. This decrease made it difficult to get a stable blend for the diesel-ethanol mixture with higher ethanol composition using the calculated surfactant combination. However, after multiple efforts with different surfactant combinations, it was observed that Span 85 can effectively lower the interfacial tensions by increasing the viscosity value in the stabilization zone. Therefore, further increase in the composition of ethanol will require an increase in the surfactant used in order to maintain blend stability.

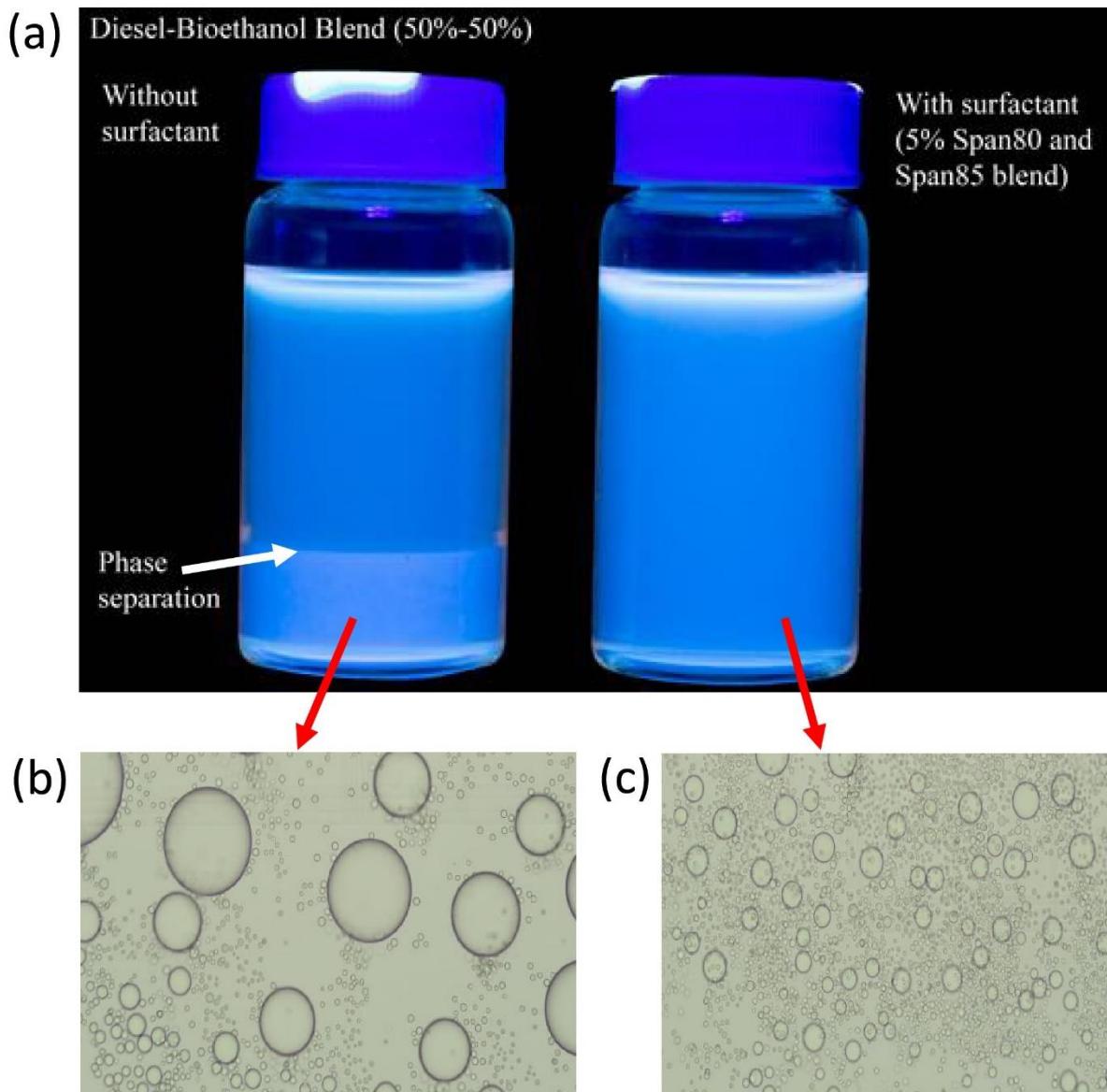


Fig. 17. The effect of SPAN on diesel-ethanol blend (50%-50%). (a) fuel blends before (left image) and after (right image) surfactant application; Microscopic image of the blend, (b) without surfactant, (c) with surfactant;

#### - Diesel-ethanol blend (40%-60%)

This blend has been made of more ethanol component which changes the oil-in-water type of emulsion to the water-in-oil emulsion. The phase separation shown in Fig. 18 (a) was observed after 24 hours of blending operation, whereby a more transparent phase that was believed to be more of the diesel component was formed at the bottom of the bottle. This was due to the composition of the blend which in this case is more hydrophilic, and this type of phase

separation is associated with sedimentation. The diesel droplets which are now dispersed in a continuous ethanol phase are heavier, resulting in a sediment of agglomerated droplets at the bottom of the bottle.

From this experiment it was observed that this particular blend is no longer water-in-oil but oil-in-water. After multiple efforts with different surfactant combinations, it was observed that Span 85 was more effective for this blend stability. The independent use of Span 85 resulted in a stable blend when 4% volumetric composition was added to the blend. The microscopic image of the blend before the addition of surfactant, shown in Fig. 18 (b), displayed diesel droplets merging together to form bigger droplets to affect the phase separation by sedimentation. It was observed from the second microscopic image in Fig. 18 (c) that these droplet sizes are now smaller with more spaces between them. This spacing phenomenon observed is believed to be the effect of the surfactant stabilising the blend, which could be associated with the reduction of the interfacial tension of both phases and creating further stability in the droplet network structure. The surfactant effect on this blend shows a reduction in the droplet sizes from 0.26-28.44  $\mu\text{m}$  to 0.26-15.01  $\mu\text{m}$  with more droplets distributed between 0.26-2.00  $\mu\text{m}$  range in the blend with surfactant.

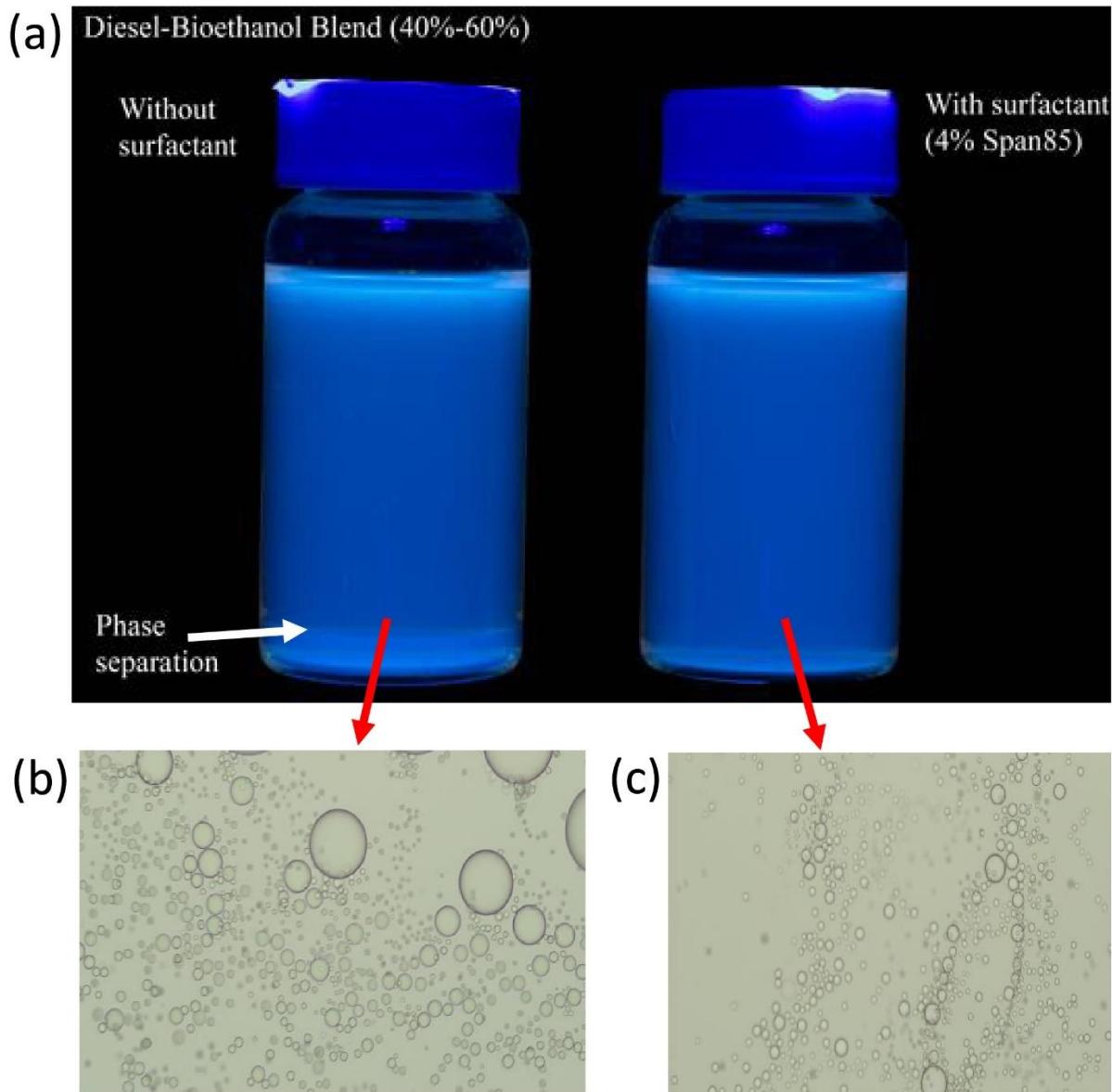


Fig. 18. The effect of SPAN on diesel-ethanol blend (40%-60%). (a) fuel blends before (left image) and after (right image) surfactant application; Microscopic image of the blend, (b) without surfactant, (c) with surfactant;

Fuel blend properties without and with surfactants are summarized in Tables 12, 13 and 14. All these blends were prepared since May 2019 and those with surfactants are still stable except for diesel-ethanol blend (40%-60%) which was only stable for 10 weeks even with surfactant.

Table 12. Fuel properties of each blending component

Fuel Properties	Diesel	Ethanol	Span 80	Span 85
Density (kg/L) at 15°C	0.843 [121]	0.79 [122]	0.99 [123]	0.94 [123]
Cetane number	52 [122]	6 [122]	-	-
Viscosity (mm <sup>2</sup> /s) at 40°C	3.42625	1.12447	2020.2 [123]	319.15 [123]
Oxygen Content (wt %)	0 [124]	34.8 [122]	-	-
Sulphur Content (mg/kg)	7 [125]	0	-	-
Lower Heating Value (MJ/kg)	44.76503	28.22433	-	-

Table 13. Fuel properties of diesel-ethanol blends without surfactant

Fuel Properties	70%-30%	60%-40%	50%-50%	40%-60%	Calculation method
Density (kg/L) at 15°C	0.827	0.822	0.816	0.811	Pearson, et al. [126]
Cetane number	32.11	28.20	24.29	20.38	Pearson, et al. [126]
Viscosity (mm <sup>2</sup> /s) at 40°C	2.34	2.16	1.82	1.61	Experimental result
Oxygen content (wt %)	8.25	11	13.75	16.50	Pearson, et al. [126]
Sulphur content (mg/kg)	4.13	3.54	2.95	2.36	Pearson, et al. [126]
Lower heating value (MJ/kg)	42.0	38.4	38.0	35.9	Experimental result

Table 14. Fuel properties of diesel-ethanol blends with surfactant

Fuel Properties	70%-30%	60%-40%	50%-50%	40%-60%	Calculation Method
	Span 80+85	Span 80+85	Span 80+85	Span 85	
Surfactant applied					
Surfactant composition	3%	3%	5%	4%	
Density (kg/L) at 15°C	0.831	0.826	0.824	0.816	Pearson, et al. [126]
Cetane number	31.17	27.38	23.13	19.59	Pearson, et al. [126]
Viscosity (mm <sup>2</sup> /s) at 40°C	2.47	2.28	2.03	1.82	Experimental result
Oxygen content (wt %)	8.01	10.68	13.09	15.86	Pearson, et al. [126]
Sulphur content (mg/kg)	4.01	3.44	2.81	2.27	Pearson, et al. [126]
Lower heating value (MJ/kg)	39.7	38.6	42.0	35.6	Experimental result

## Conclusion

The future of the transport sector and especially the propulsion of heavy-duty vehicles have been analysed and discussed and a few points have been highlighted. First of all, the feasibility and the viability of a transition to a fully electric transport sector have been analysed. Secondly, different alternatives to fossil fuels such as biomass-based fuels have been explored. And finally, the use of ethanol in high concentrations, as a viable representative of biomass-based fuels in CI engines has been explored. The main conclusions can be summarized as follows:

- Electric vehicles need to reach a better well-to-wheel efficiency and better cost and performance level to be considered as a viable alternative for the heavy-duty transportation sector. Not only the production of batteries but also the collection and recycling need to be improved to reduce the high level of GHG emissions emitted during these steps.
- Among all the biomass-based oxygenated biofuels, ethanol seems to be at the moment the best feasible alternative showing the best characteristics to mitigate the GHG emissions. Ethanol also has advantages of being used without a need for any modifications of engines, and it is cheap to produce.

- When used in CI engines, ethanol-diesel blends show promising results in reduction of smoke and CO emissions. However, further developments are required to improve reduction of NO<sub>x</sub> and HC when high-concentration ethanol is used.
- Surfactants are an efficient way to go beyond the blend walls and allow creating fuel blends of neat diesel with high-percentage ethanol without a need for engine modifications but still maintaining required engine performance.
- Further investigations to create fuel blends with biosurfactants could lead to even more sustainable alternative, and the use of cetane improvers could help boosting engine performance and mitigate NO<sub>x</sub> emissions.

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