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Review Article

A Review on the Role and Impact of Typical Alcohol Additives in Controlling Emissions from Diesel Engines

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Abstract. Today, most of the essential energy needs of humans and production are met by fossil fuels that are expected to be exhausted in the next century. Furthermore, fossil fuels are not renewable and sensitive to the environment. In particular, there is growing concerned about the negative impact of internal combustion engine emissions on climate change and global environmental pollution. Fuel and alcohol-based additives are being considered as good candidates for sustainable alternative fuels used on compression ignition engines. In this review, the different key production pathways and properties of each of the five alcohol additive candidates were discussed. Besides, their effects on the emission characteristics of diesel engines when alcohol additives are added to diesel fuel are also carefully considered. Five candidates including methanol, ethanol, propanol, butanol, and pentanol have been shown to control pollutants from combustion engines while using alcohol-based additives. This is of great significance in the strategy of coping with the threats of pollution and climate change caused by the operation of transport vehicles.

Keywords: Alcohol additives; emission characteristics; diesel engine; controlling emissions.

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1. Introduction

Pressure on energy demand stems from the growth of the global population. Energy consumption demand by 2050 is expected to increase by about 50% compared to current energy consumption (Chu *et al.*, 2020). In particular, Asia is accounting for half of the world's population, so the energy demand in this continent is increasing sharply. Furthermore, there is a close relationship between the increasing levels of energy use and the increasingly severe environmental pollution occurring in Asian countries (Le *et al.*, 2021). The excessive use of fossil fuels is causing two sides of a global problem, the depletion of fossil fuels, and the excessive levels of PM (Nayak *et al.*, 2020), and carbon dioxide (CO_2) emissions (Ölçer *et al.*, 2021). The common danger of both sides is to seriously damage the mother earth's ecosystem as well as increase global climate change (Sharifi and Yamagata, 2016)(Łosiewicz, 2017)(Viet and Tuan, 2018).

There are many effective solutions to reduce emissions from diesel engines such as improving combustion quality, adjusting the engine structure, and installing an exhaust gas treatment system, as described in Figure 1. Although the community of researchers has implemented emission treatment solutions such as exhaust gas recirculation (Hoang, 2018), catalytic reduction selectivity (Pham, 2019), optimization for injection strategy (Hoang, 2020a)(Nayak and Mishra, 2016)(Hoang, 2020b) and piston-top geometry (Tran *et al.*, 2020) and adding nanoparticles (Hoang, 2021). However, those solutions are complex and have to accept their trade-offs for other emissions or engine modifications. In that context, energy transition to clean and sustainable energy is considered to be the end of the above concern (Huynh *et al.*, 2021). Although clean energy offers many opportunities to mitigate global climate change (Xuan and Viet, 2021), it also faces several technological and policy challenges (Thomas *et al.*, 2021). Indeed, a recent research trend is to reduce the use of traditional fossil fuels by using solutions using alternative and renewable fuels such as biofuels (Tabatabaei *et al.*, 2021)(Nayak *et al.*, 2017) and hydrogen (Murugesan *et al.*, 2021). Moreover, the process of adapting the decarbonization strategy (Dong *et al.*, 2021) has promoted new technologies in biofuel production from lignocellulosic biomass sources (Chin Kui *et al.*, 2021). More interestingly, lignocellulosic biomass pre-treatment techniques have achieved new advances and optimizations in improving yields and reducing pollutant

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waste chemicals (Chong *et al.*, 2021). Advanced technologies in pyrolysis, gasification, fermentation, thermochemical conversion, and anaerobic digestion have improved the production efficiency of biorefineries (Ong *et al.*, 2021). Value-added products from biorefineries can be bio-alcohol, biodiesel, syngas and hydrogen, all of which are making significant contributions to the sustainable energy transition.

However, the use of biofuel does not avoid some disadvantages stemming from their properties (Le and Hoang, 2017). For example, the biofuel itself has a higher viscosity, higher density (Anh, 2018)(Nayak *et al.*, 2019), and lower volatility properties than that of conventional fuels and therefore can adversely affect atomization efficiency and combustion quality (Le and Pham, 2019). Serious problems caused by biodiesel for compression ignition (CI) engines can be the formation of deposits in the combustion chamber (Le and Hoang, 2019) or lubricant degradation (Pham and Hoang, 2019a), or the corrosion behavior of metal/alloys in engines (Tabatabaei and Aghbashlo, 2020). In addition, a lower energy density of alcohol-based fuels results in higher fuel consumption to achieve the same power (Al-Tawaha *et al.*, 2019). Therefore, one of the emerging strategies is to partially replace liquid biofuels with organic-derived fuel additives to increase the efficiency of biofuel use. Emerging organic additives such as 2,5 dimethylfuran (DMF) (Aykut and Sandro, 2021)(Engel *et al.*, 2021)(Le *et al.*, 2021), 2-Methylfuran (MF) (Van and Anh, 2021), and alcohol (Darmayanti *et al.*, 2020) additives are considered excellent alternative candidates for this energy shift. With advantages such as being non-toxic, environmentally friendly, and bringing energy efficiency and higher economy, they are more feasible when applied on internal combustion engines (Sandro and Viet, 2020)(Ölçer and Nižetić, 2021).

Recent review works of the literature indicated that organic additives, especially alcohol-based additives, are being used more on direct injection and compression ignition engines (Norhafana *et al.*, 2018). More clearly, alcohol-based compounds are used in many spark injection engines as biofuel (Chau *et al.*, 2020). However, there are many barriers to be found when using alcohol directly for a diesel engine or blending with diesel fuel. Alcohol-based compounds are used in many spark injection engines as biofuel (Mamat *et al.*, 2019). However, there are many barriers to be found when using alcohol directly for a diesel engine or blending with diesel fuel. Some studies suggest that complications occur when mixing alcohol with diesel fuel due to poor lubricity and high spontaneous ignition temperature. Furthermore, to avoid the separation of alcohol and diesel in the mixture it is necessary to use emulsifiers or solvents. However, recent studies have provided effective solutions to overcome alcohol-diesel blending problems (Shanmugam *et al.*, 2021). The addition of alcohol-based additives to diesel fuel has many advantages in engine performance as well as emissions characteristics. For example, OH-based additives when blended with diesel fuel can improve overall engine performance, specific fuel consumption, and increase in-cylinder pressure. In addition, exhaust components from diesel engines such as particulate matter (PM), nitrogen oxides (NOx), unburnt hydrocarbon (HC), and carbon oxide (CO) are often reduced if alcohol additives are added to the fuel (Zaharin *et al.*, 2017).

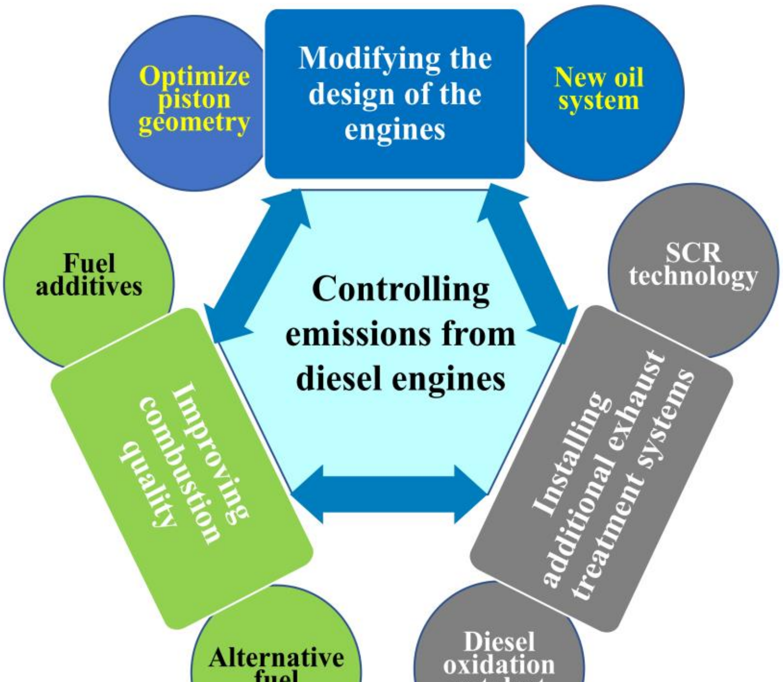

Fig.1. Solutions to control emissions from diesel engines

Although, the role of alcohol additives in biofuels has been confirmed by the results of simulation and experimental studies for compression ignition engines. However, the review literature often reports both engine performance and combustion properties when diesel engines use typical alcohol additives. That means a more in-depth assessment of the relevant properties is required. The content presented in this review can be summarized as follows: (i) The process of converting bio-alcohol from biomass is also briefly discussed to clarify the role and potential of this additive; (ii) Comprehensive assessment of the chemical and physical properties of typical alcohol additives as well as their impact on the emission characteristics in diesel engines; (iii) Conclusions and prospects for alcohol additives.

2. The pathway of bio-alcohol production

Bio-alcohol production plays a key role in stabilizing the biofuel supply chain. Bio-alcohol is defined as a fuel that is either biologically produced or converted from biomass to alcohol. The prominent products of bio-alcohol can be listed as methanol, ethanol, propanol, butanol, etc. In the first-generation bio-refineries, the fermentation of starch or cellulose is considered the traditional method for bio-ethanol production. However, in the second-generation bio-alcohol, bio-methanol and bio-ethanol are purified mainly through the pyrolysis of agro-forestry waste or algal biomass. In particular, bio-methanol is easier to recover from biomass. Fast pyrolysis is considered a typical technique among the three current biomass pyrolysis techniques (slow pyrolysis, fast pyrolysis, and flash pyrolysis) (Demirbaş, 2000). The fast pyrolysis for bio-alcohol production usually takes place in a fixed bed reactor at 400-500^{o}C, 10-200^{o}C/s, and inert gas support (Kasmuri *et al.*, 2017).

The conversion efficiency is an output parameter that determines the scale of bio-alcohol production. The bio-methanol production process is easier and reduces greenhouse gas emissions. It can be prepared by biochemical as well as thermochemical processes (Chan *et al.*, 2019). Moreover, the applications of methanol are expected to expand, it can be a medium material for the synthesis of other useful organic compounds as well as a promising hydrogen carrier. The path of methanol production by thermochemical conversion is carried out through three stages (1) gasification, (2) pyrolysis, and (3) liquefaction (Raheem et al., 2015). In this process, a syngas mixture of H_2 and CO is formed by pyrolysis of biomass in an oxygen-deficient environment. Next, liquid biofuel is formed when the syngas is catalytically treated. Conditions to maintain the activity of the catalyst in the reaction of converting syngas to methanol are high temperature (200-400^{o}C) and high pressure (300bar) (Chin Kui *et al.*, 2021). The conversion efficiency to methanol depends on the cleanliness of the syngas or the CO_2 removal capacity which depends on the gas purification technology. In addition, the catalysts used in the process are expensive, and the equipment operates under extreme conditions (high temperature and pressure). The conversion efficiency to methanol depends on the cleanliness of the syngas or the CO_2 removal capacity which depends on the gas purification technology. In addition, the catalysts used in the process are expensive, and the equipment operates under extreme conditions (high temperature and pressure). These characteristics have made the production cost of methanol from syngas very high. Therefore, a low-cost methanol production method is the biochemical conversion through anaerobic digestion, fermentation, and photoconversion. During the biochemical conversion process, methane gas is considered an essential raw material for heterotrophic bacteria (Jin *et al.*, 2021). All processes take place at ambient temperature and pressure. The processes for the production of methanol from biomass are depicted in Figure 2.

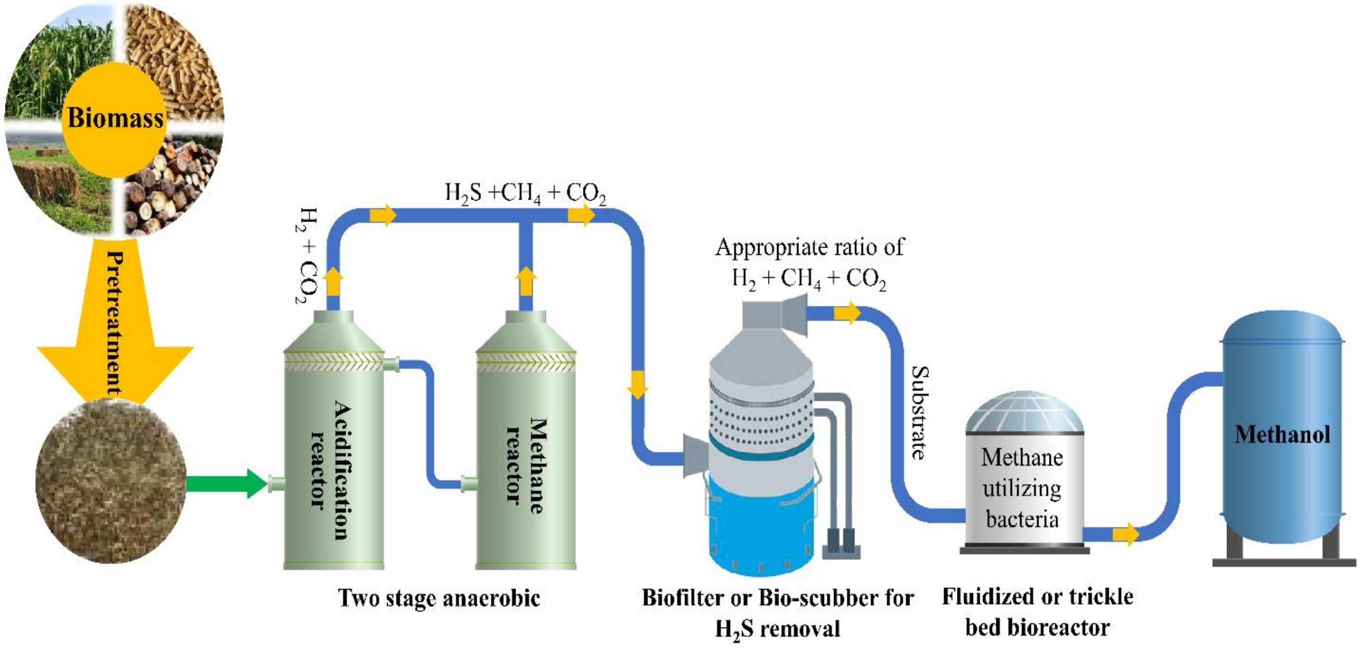

Fig. 2. The processes of the methanol production from biomass (Jin *et al.*, 2021)

3. Properties of alcohol additives

Typical alcohol-based additives can be listed such as methanol, ethanol, propanol, butanol, and pentanol. These alcohol additives have the characteristic properties shown in Table 1. Methanol has the most basic molecular structure of all alcohols. It has four hydrogen atoms, one oxygen atom, and one carbon atom. It is flammable at atmospheric temperature and burns with a blue smokeless flame. Properties of pure methanol: chemical structure CH_3OH; The low calorific value of 19.9 MJ/kg is about half the low calorific value of diesel and heavy fuel; Flashing point of 12 °C; Auto-ignition temperature of 470 °C is the lowest temperature at which a substance will automatically ignite and burn under normal atmospheric conditions without any external influence; High auto-ignition temperatures can be difficult to burn in conventional diesel engines (Verhelst *et al.*, 2019)(Dong *et al.*, 2019). A diesel engine burns only methanol fuel with high-temperature, high-compression air; and density at 15 °C is 0.796 kg/dm^3, which is lower than that of diesel fuel. Methanol burns at a lower heat release rate than conventional fuels, about one-third of that of diesel. It has a sulfur content of 0.5 ppm, which is significantly lower than that required by the fuel used in Sulfur Emission Control Areas (Svanberg *et al.*, 2018)(Tran *et al.*, 2020). Therefore, methanol can become a potential fuel or fuel additive for internal combustion engines. However, methanol is corrosive, so care should be taken when choosing the material for the fuel system (stainless steel is the material recommended for use with methanol). Methanol is only compatible with some plastics and rubbers, so the materials used in gaskets, o-rings, etc., should be checked for compatibility with methanol (Li *et al.*, 2020). On the other hand, methanol is completely soluble in water, and methanol solutions do not ignite when the methanol concentration is below 25% in water. This means that if water is used to control the combustion temperature, the volume of water should be at least four times the volume of methanol (Gupta and Mishra, 2019)(Lamas *et al.*, 2015).

Ethanol, also known as ethyl alcohol, is an organic compound, in the homologous series of methyl alcohol, flammable, colorless, is one of the common alcohols in the composition of alcoholic beverages (Sun and Cheng, 2002). Ethanol alcohol is aromatic, the colorless flame that evaporates at quite a low temperature, is very hydrophilic, can indefinitely dissolve in water and many other organic and inorganic substances. The high latent heat leads to a cooling effect of the intake air (Minh and Anh, 2018). Therefore, the engine when using alcohol fuel or additives is filled with more mixture into the cylinder. Ethanol has high octane number, which means good anti-knocking ability, thus allowing to increase compression ratio as well as engine efficiency. In recent times, the use of ethanol as an efficient blending fuel on internal combustion engines has been widely deployed in countries with abundant ethanol sources such as Brazil. However, a lower flash point than diesel fuel and its corrosion potential also requires safety measures in material storage and compatibility (Shahir *et al.*, 2014)(Dung and Anh, 2020).

Propanol is being seen as another successful alternative fuel to ethanol. In terms of chemical isomers, propanol comes in two forms, n-propanol and isopropanol. The cost of producing propanol is higher than that of ethanol, leading to a lower number of studies on this alcohol-based fuel (Kim and Park, 2018). However, propanol has superior advantages compared to ethanol and methanol in terms of higher cetane coefficients, greater calorific values, better solubility, and lower corrosion risks. Propanol is being seen as another successful alternative fuel to ethanol. In terms of chemical isomers, propanol comes in two forms, n-propanol and isopropanol (Walther and François, 2016). The cost of producing propanol is higher than that of ethanol, leading to a lower number of studies on this alcohol-based fuel. However, propanol has superior advantages compared to ethanol and methanol in terms of higher cetane coefficients, greater calorific values, better solubility, and lower corrosion risks (Al-Tawaha *et al.*, 2018). In addition, the higher flash point and density support an increase in energy value. Furthermore, the lower ignition heat of propanol is due to the higher evaporation latency heat, resulting in a reduced delay in burning time compared to lower alcohols (Putri *et al.*, 2018).

Table 1
Physicochemical and combustion properties of typical alcohol additives

Additive	Molecular formula	Density (kg/m^3)	Viscosity (cSt)	Latent heat (kJ/kg)	Boiling point (°C)	Cetane number	Oxygen content (%)	Lower heating value (MJ/kg)	Auto-ignition temperature (°C)	References
Methanol	CH_3OH	729	0.54	1178	64	5	50	20	470	(Verhelst *et al.*, 2019)
Ethanol	C_2H_5OH	792	1.04	840	78	7	34	27	434	(Shahir *et al.*, 2014)
Propanol	C_3H_7OH	804	1.74	728	97	12	26.6	30.6	350	(Putri *et al.*, 2018)
n-Butanol	C_4H_9OH	810	2.2	585	118	25	22	33	385	(Brandão and Suarez, 2018)
Pentanol	$C_5H_{11}OH$	814	2.89	308	138	20	18.2	34.7	300	(Zhu *et al.*, 2016)

Butanol has attracted the most attention recently in alcohol-based fuels or additives. In terms of chemical structure, butanol has 4 OH groups that are attached to different carbon chain positions that form four isomers (Chen *et al.*, 2015). Butanol has a higher cetane number and is less toxic and corrosive than ethanol. More importantly, its volatility and higher flash point made it easy to integrate with the fuel system in internal combustion engines. Furthermore, the viscosity and density of butanol are close to that of diesel fuel, thus meeting the requirements of fuel economy as well as lubricity (Brandão and Suarez, 2018). With higher heat value and lower vaporization calorific value than isopropanol and ethanol, resulting in better flammability. However, the slightly higher latent heat increases the ignition delay time, resulting in a higher amount of fuel accumulating in the cylinder (Kumar *et al.*, 2013).

The fifth member of the alcohol family is pentanol, which is seen as a potential fuel because of its properties and production advantages. The methods of producing pentanol require less energy since it is based on natural fermentation mechanism. Furthermore, the stability of pentanol when blended with conventional fuels is better than those four alcohols (Zhang *et al.*, 2008). Better yet, the cetane number and energy content are higher while the density, latent heat, and viscosity are asymptotic to the traditional diesel fuel. In addition, pentanol is a corrosion inhibitor and therefore is well compatible with the metallic materials of the fuel system composition (Zhu *et al.*, 2016). Recent studies have focused on the combustion behavior of pentanol isomers in the diesel engine combustion chamber with updated chemical kinetics models (Togbe *et al.*, 2011).

4. Impact of alcohol additives on emission characteristics

The combustion characteristics and the chemical reactions that create soot are affected mainly and directly by the content of oxygen and the additive's molecular structure (Nabi and Hustad, 2010)(Xuan *et al.*, 2021). Improvement of combustion process and decline of emission are possible with the help of alcohol compound additives (i.e., ethanol, methanol, n-butanol, etc.) which have rich oxygen content (Kumar *et al.*, 2018). Additionally, Khalife et al. (Khalife *et al.*, 2017) indicated that the PM and HC emissions were gone down as integrating alcohols and diesel because the amount of oxygen is sufficient for the combustion process. On the other hand, the engine exhausted more emissions of CO and NOx. Due to the rich content of oxygen, high ratio of stoichiometric air/fuel, low sulfur content, and high ratio of hydrogen/carbon, the concentration of emission are declined. Some similar discussions have also been shown in the comparative studies using the DMF additive by Tran et al. (Tran *et al.*, 2021)(Chan, 2018) and Bui et al. (Balasubramanian *et al.*, 2021a). Moreover, findings from the study by Akcay et al. (Akcay and Ozer, 2019) observed that the use of fusel oil mixtures declined CO, NOx, and smoke emissions. In contrast, a negative impact on the engine performance and a decline of the maximum in-cylinder pressure appeared as the amount of the fusel oil additive raised (Cao *et al.*, 2020)(Vinh *et al.*, 2018). A general picture of the application of bio-alcohol additives in reducing pollutant emissions from diesel engines has been described in this section. Table 2 presents a summary of the prominent effects of alcohol additives on the emission characteristics (CO, HC and NOx) of CI engines.

Table 2
Effect of typical alcohol additives on emission characteristics in diesel engine

Additive	Concentration	Test Condition	NO_X	CO	HC	Ref.
Methanol	5 (M5)	Injection times: 15^o, 20^o, 25^o CA BTDC Loads: 5, 10, 15, 20 Nm at a constant speed	↓		↑	(Sayin *et al.*, 2009)
	10 (M10)		↓		↑ (↓ by 20.12% at 20Nm)	
	15 (M15)		↓		↑	
Methanol	5 (M5)	Speeds: 1000, 1800 rpm and fixed torque at 30 Nm		↓ 27.3% compared to Diesel fuel	↓ 31.5% compared to Diesel fuel	(Sayin, 2010)
	10 (M10)			↓ 2.4% compare to M5	↓ 5.7% compare to M5	
Methanol	20% vol.	Injection times: 350, 360,370,380,390,400 CA Loads: 5. 10, 15, 20 Nm at constant speed.	↓ at low load	↑ compare to Diesel fuel	↑ compare to Diesel fuel	(Tutak *et al.*, 2015)
	50% vol.		↓ at low load	↑ compare to Diesel fuel	↑ compare to Diesel fuel	
	75% vol.		↓ at low load	↑ compare to Diesel fuel	↑ compare to Diesel fuel	
	90% vol.		↓ at low load	↑ compare to Diesel fuel	↑ compare to Diesel fuel	
Methanol	5% (DM5)	Loads: 25, 50, 75, 100%	↓ compare to diesel fuel	-	-	(Datta and Mandal, 2016)
	10%(DM10)		↓ compare to DM5	-	-	
	15%(DM15)		↓ compare to DM10	-	-	
Ethanol	5% (E5)	BMEP: 1.40, 2.57, 5.37 bar; Injection times: -20^o to 80^o CA	↓ compare to Diesel fuel	↓ compare to Diesel fuel	↑ compare to Diesel fuel	(Rakopoulos *et al.*, 2007)
	10%(E10)		↓ compare to Diesel fuel and E5	↓ compare to Diesel fuel and E5	↑ compare to Diesel fuel and E5	

Ethanol	15%(E15)	Speeds: 1200, 1500 rpm; BMEP: 3.56, 7.04, 10.52 bar	↓ compare to Diesel fuel and E10	↓ compare to Diesel fuel and E10	↑ compare to Diesel fuel and E10	(Rakopoulos *et al.*, 2008)
	5% (E5)		↓ compare to Diesel fuel	↓ compare to Diesel fuel	↑ compare to Diesel fuel	
	10%(E10)		↓ compare to Diesel fuel and E5	↓ compare to Diesel fuel and E5	↑ compare to Diesel fuel and E5	
Ethanol	5% (E5)	Injection time: 27°, 30°, 33° BTDC; The constant speed at 2200 rpm The constant load of 30Nm	↑ compared to Diesel fuel	↓ compared to Diesel fuel	↓ compared to Diesel fuel	(Uslu and Sayin, 2008)
	10%(E10)		↑ compared to Diesel fuel and E5	↓ compared to Diesel fuel and E5	↓ compared to Diesel fuel and E5	
	15%(E15)		↑ compared to Diesel fuel and E10	↓ compared to Diesel fuel and E10	↓ compared to Diesel fuel and E10	
Ethanol	5% (E5)	Speeds: 1000÷1800 rpm; with a step of 200 Fix load at 30 Nm	↑ compared to Diesel fuel	↓ compared to Diesel fuel	↓ significantly compared to Diesel fuel	(Sayin, 2010)
	10%(E10)		↑ compared to Diesel fuel	↓ compared to Diesel fuel and E5	↓ significantly compared to Diesel fuel and E5	
Ethanol	10% by vol. (E10)	Main injection time: -60º to 60º ATDC Pilot injection time: -30° to -10° ATDC	↓ compared to Diesel fuel	↑ compared to Diesel	↑ compared to Diesel	(Park *et al.*, 2010)
	20% by vol. (E20)		↓ compared to Diesel fuel and E10	↑significantly compared to Diesel fuel and E10	↑significantly compared to Diesel fuel and E10	
Ethanol	85% (E85)	Fixed speed of 1500 rpm at full load of 24 kW	↓ compared to Diesel fuel at low load	↑ compared to Diesel	↑ compared to Diesel	(Tutak *et al.*, 2015)
Ethanol	11mg/cycle	Injection time: 350, 400 and 450 CA EGR rates: 10 and 25%	↓ 88% compared to Diesel fuel	-	-	(Nour *et al.*, 2017)
Ethanol and isopropanol	Ethanol 15% (DE15) isopropanol 15%vol (DIPA15)	Speeds: 1500, 2000, 2500 rpm Loads: 3.3, 5.0, 6.6, 8.3 bar	↑ 7.1% compared to Diesel fuel but lower than DIPA15	↑ 6% compared to Diesel fuel	↑ compared to Diesel fuel but lower than DIPA15, only ↓ at 2500 rpm and BMEP of 6.6 bar	(Alptekin, 2017)
Butanol	5% (DB5)	Loads: 25, 50, 75, 100%; Speeds: 1500, 2500, 3000, 3500 rpm	At 1500 rpm: ↑ compared to Diesel fuel, ↓ at 100% load. At 3000 rpm: ↓ compared to Diesel fuel, ↑ at 50% load	At 1500 rpm: ↓ compared to Diesel fuel, ↓ at 100% load. At 3000 rpm: ↑ compared to Diesel fuel at 25% load	At 1500 rpm: ↑ compared to Diesel fuel, At 3000 rpm: ↓ compared to Diesel fuel at 50% load	(Siwale *et al.*, 2013)
	10% (DB10)		At 1500 rpm: ↑ compared to Diesel fuel, ↓ at 100% load. At 3000 rpm: ↓ compared to Diesel fuel,	At 1500 rpm: ↓ compared to Diesel fuel, At 3000 rpm: ↓ compared to Diesel fuel at 75 and 100% load and ↑ at 25 and 50% load	At 1500 rpm: ↑ compared to Diesel fuel, At 3000 rpm: ↓ compared to Diesel fuel at 50% load	
	20% (DB20)		At 1500 rpm: ↑ compared to Diesel fuel, only ↓ at 100% load. At 3000 rpm: no significant change comparedd to Diesel fuel	At 1500 rpm: ↓ compared to Diesel fuel. At 3000 rpm: ↓ compared to Diesel fuel at 75 and 100% load and ↑ at 25 and 50% load	At 1500 rpm: ↑ compared to Diesel fuel, At 3000 rpm: ↓ compared to Diesel fuel at 100% load	

Butanol	20% (DB20)	EGRs: 10, 20, 30, 40, 50 and 60%	↓ compared to Diesel fuel with increasing EGR percentage			(Liu *et al.*, 2013)
Butanol	20% (BU20)	Injection pressures: 100, 120, 140 and 160 MPa	↓ slightly compared to Diesel fuel,	-	-	(Merola *et al.*, 2014)
n-Butanol	5% (BU5)	Speeds:1000, 2000, 3000 and 4000 rpm	↓ compared to Diesel fuel	↑ compared to Diesel fuel	↑ compared to Diesel fuel	(Choi *et al.*, 2015)
	10% (BU10)		↓ compared to Diesel fuel	↑ compared to B5	↓ compared to Diesel fuel	
	20% (BU20)		↓ compared to Diesel fuel, lower than other blends	↑ compared to B10	↑ compared to Diesel fuel, higher than other blends	
n-Butanol (10, 20 and 30%) and isobutanol (10, 20 and 30%)	10% butanol (NB10)	Loads: 20, 40, 60, 80 and 100%; 10% EGR	↓ compared to Diesel fuel	No significant change at partial load, ↓ compared to Diesel fuel at full load	↑ compared to Diesel fuel	(Kumar *et al.*, 2015)
	20% butanol (NB20)		↓ compared to Diesel fuel NB10 and NB30	No significant change at partial load, ↓ compared to Diesel fuel NB10 and NB30	↑ compared to Diesel fuel and NB10	
	30% butanol (NB30)		↓ compared to Diesel fuel and NB10	No significant change at partial load,↓ compared to Diesel fuel and NB10	↑ compared to Diesel fuel and NB20	
Butanol	2% nB2	Speeds: 2000, 3000 and 4000 rpm Loads: 85, 105, 115, 125, 135 and 145 Nm	↓ 1.47, 2.74 and 5.03% compared to Diesel fuel at 2000, 3000 and 4000 rpm	↑ significantly compared to Diesel fuel	-	(Şahin and Aksu, 2015)
	4% nB4		↑ compared to Diesel fuel	↑ significantly compared to Diesel fuel	-	
	6% nB6		↑ compared to Diesel fuel	↑ significantly compared to Diesel fuel	-	
n-Butanol	10% (B10)	CRs: 17.5, 18.5 19.5 and 20.5 Injection timing: 21, 23 CA BTDC Injection pressures: 200, 210 and 220 bar Engine loads: 12, 16, 20 and 24 Nm The constant speed at 1500 rpm	↓ compared to Diesel fuel	↑ compared to Diesel fuel, higher than B15 but lower than B25.	↑ compared to Diesel fuel, highest among blends	(Nayyar *et al.*, 2017)
	15% (B15)		↓ compared to Diesel fuel	↑ compared to Diesel fuel, higher than B20.	↑ compared to Diesel fuel, lowest among blends	
	20% (B20)		↓ compared to Diesel fuel	↑ compared to Diesel fuel, lowest among blends	↑ compared to Diesel fuel, higher than B15.	
	25% (B25)		↑ compared to Diesel fuel	↑ compared to Diesel fuel, highest among blends	↑ compared to Diesel fuel, higher than B20 but lower than B10.	

One of the most favorable fuels for conventional fossil-based fuels is methanol. In the last few years, instead of using conventional fossil-based fuels in CI engines, methanol has been applied as an alternative fuel in several types of research to deal with problems of economy and environment (Zhen and Wang, 2015). Moreover, the study of Chen et al. (Chen *et al.*, 2019) presented that the fuel properties of methanol additive and its high molecular oxygen is promising for diesel. It is interesting that due to its low boiling temperature, the volatility is improved and the high content of oxygen promotes blend fuel combustion faster. The additive of n-pentanol may help methanol to be able to dissolve in diesel and to form stable, homogeneous, and transparent microemulsion fuels. According to the results of this study, opacity and soot particles of Diesel/n-pentanol/methanol blend fuels were lower than that of diesel and methanol had significant improvement of the reductions, while a higher concentration of NOx emissions is produced, which showed a trade-off existing between NOx and soot emissions formation (Thu and Anh, 2017). The peak combustion temperature and in-cylinder temperature in the diffusion phase of combustion have a close association with formatting NOx emissions. Besides, the demonstration from Agarwal et al. (Agarwal *et al.*, 2019) indicated that solution of some troubles of diesel engines, reduction in NOx, CO, and HC emissions can be handled simultaneously by methanol–diesel (MD) blends stabilized using suitable additives. Among fuel blends, the lowest amount of CO, HC, and NOx emissions belonged to MD15, the differences shown in Figure 3.

In terms of CO emission characteristics from CI engines, CO emissions are significantly reduced when the fuel fed to the engine is added to the additive methanol, mainly due to the lower C / H ratio and its oxidizing nature. Direct ignition (DI) engines using Exhaust Gas Recirculation (EGR) supercharging were tested by Sato et al. (Sato *et al.*, 1997) to investigate the combustion and NOx emission characteristics of methanol. It is observed that a dramatic reduction of the NOx and unburned HC emissions along with high thermal efficiency maintenance were achieved with heavy EGR during supercharging (Van Pham and Anh Hoang, 2020). Reported from the study of Popa et al. (Popa *et al.*, 2001) demonstrated that increasing concentration of methanol in the fuel blend decreased significantly smoke and NOx levels for whole loads of the engine. More interestingly, It is remarkable that a marked drop in CO and smoke emissions, no large difference in exhaust hydrocarbon, and an upward trend of NOx emissions were found by Huang et al. (Huang *et al.*, 2004).

Prashant et al. (Prashant *et al.*, 2016) observed a longer ignition delay with diesel methanol dual fuel mode as compared to pure diesel mode. Increasing methanol concentration in diesel dropped NOx and soot emissions, however, it is opposite with HC and CO emissions. Recently, Wei et al. (Wei *et al.*, 2017) experimented investigate how combustion and both regulated and unregulated emissions characteristics of diesel-methanol dual fuel (DMDF) engine are affected by pilot injection for optimization of combustion process at high methanol substitution ratio (M0, M10, M30, and M50) and low load condition. Indeed, stability of combustion and economical consumption of fuel in case of high methanol substitution ratio could be enhanced by the pilot injection which also has a positive effect on reductant of regulated emissions CO, THC except for NOx, and unregulated emissions tested in this study except CO_2 on M0 and M10 mode and toluene on M50 mode when compared with single-injection cases.

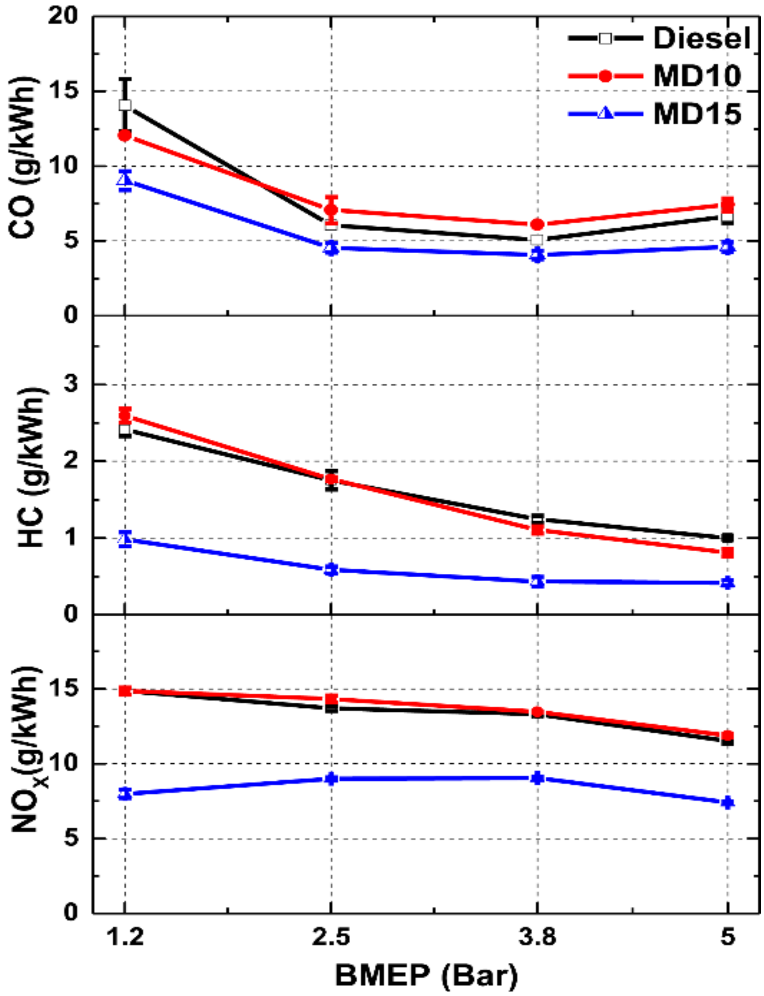

Fig. 3. Change of CO, HC, and NOx emissions from CI engines using diesel-methanol blends (Agarwal *et al.*, 2019)

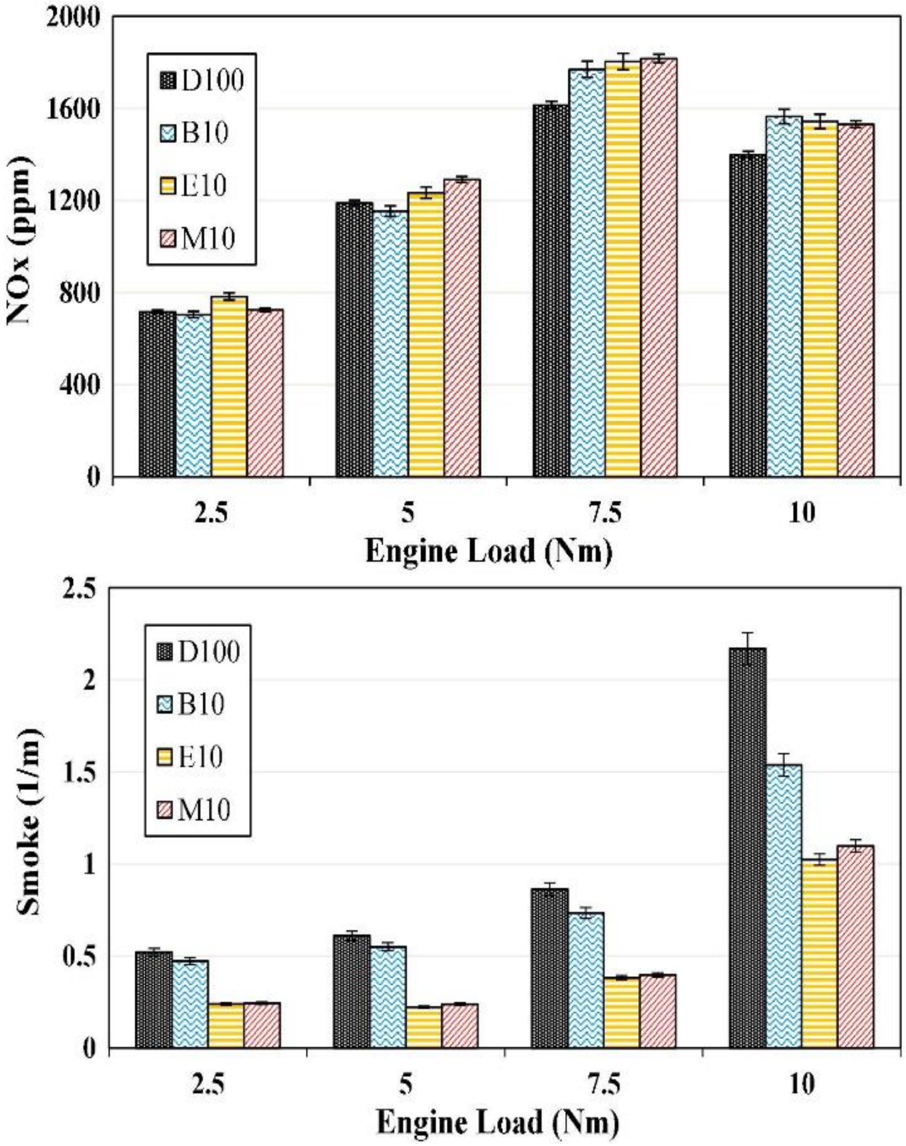

Fig. 4. Change of NOx and smoke emission from CI engine according to various load (Emiroğlu and Şen, 2018)

Ethanol is considered as a biomass-based alternative fuel, which is extracted from the process of pretreatment and conversion of cellulose-rich agroforestry products such as straw, rice husk, food stalks, grass, wood, etc (Chin Kui *et al*., 2021). The incorporation of different alcohols consisting of ethanol and diesel fuel creates a motivation to produce a clean fuel (Kasmuri *et al*., 2017). Although emissions of diesel engines can be decreased by methanol or ethanol, good miscibility is the strong point of ethanol (Jin *et al*., 2021). Some recent studies revealed that emissions like CO and PM can be reduced effectively by ethanol-diesel fuel blends. For example, the study of Emiroğlu et al. (Emiroğlu and Şen, 2018), the blending of 10% methanol, 10% ethanol, and 10% butanol with diesel fuel to supply diesel engines have shown a slight increase in NOx and at the same time a decrease in CO and smoke emissions. The results of this emission characteristic are depicted in Figure 4. On the other hand, HC and CO emissions are increased by the blending of 10%, 20%, and 30% by vol of ethanol in diesel fuels, which was discovered in the study of fuel Xu et al. (Xu *et al*., 2007). The soot and NOx emissions were produced by burning ethanol-blended fuels were higher than by pure diesel fuel. Nonetheless, when the torque output of the engine increased, NOx emission grew up (Noor *et al*., 2018). According to Chen et al. (Chen *et al*., 2008), a fall in emissions (blends E10B, E20B, and E30B obtained a decline of PM by 25%, 50 and 50%; a drop of smoke by 30%, 55 and 85% respectively), however, NOx emissions is almost similar between blends and slightly higher than pure diesel. As compared to pure diesel fuel, the increased trend of CO emissions displayed at low and medium loads, while high and full loads obtained the opposite trend, was agreed with the study by Bui et al. (Bui *et al*., 2020).

There are some inherent problems with methanol and ethanol such as the miscibility with diesel fuel. This has led the researchers to look for a potential alternative additive like propanol. However, propanol is used as a tertiary solvent instead of mixing with diesel fuel (Rajesh Kumar and Saravanan, 2016). In terms of emission characteristics, the presence of oxygen in propanol promotes combustion and reduces CO concentrations in the exhaust gas with CI engines operating at high loads (Balamurugan and Nalini, 2014a). Research by Yilmaz et al. (Yilmaz *et al.*, 2016) revealed that increasing levels of propanol in diesel fuel helped reduce NOx emissions. This was also consistent with the test results of blends including diesel, diesel-propanol, and diesel-butanol carried out by Balamurugan et al. (Balamurugan and Nalini, 2014b). With a contribution of 8%vol. propanol and 8%vol. butanol, NOx emissions were recorded the reduction of 19.7% and 14.3% compared to pure conventional fuels, respectively (Figure 5).

The main reason is that higher latent heat causes an increase in the cooling effect which reduces the maximum burning temperature in the cylinder, thereby contributing to reducing NOx emissions. An unexpected challenge for a propanol additive is HC emissions. The higher the propanol additive blend ratio caused more HC emissions. For example, a study by Balamurugan et al. (Balamurugan and Nalini, 2014b) revealed that the HC emissions of modified fuels with 8% n-propanol and 8% n-butanol increased by 29.8% and 23.8% compared to diesel respectively, that depicted in Figure 5. This is explained through the mechanism of cooling the combustion mixture in the cylinder, so it pulls the burning temperature down, resulting in poor mixing quality. In addition, the higher viscosity and higher cetane number may result in longer ignition delays leading to increased HC emissions (Deep *et al.*, 2014)(Nayak *et al.*, 2022).

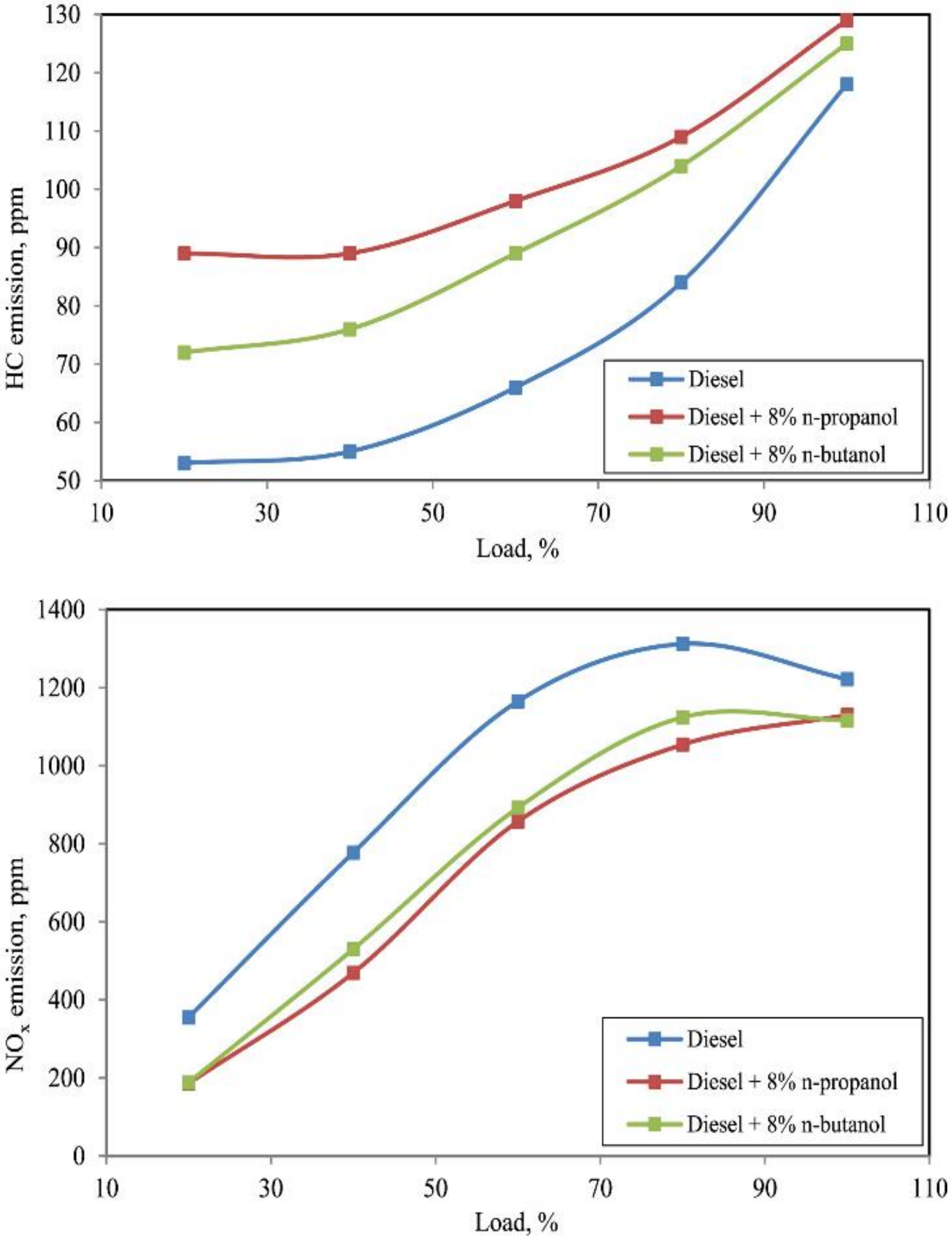

Fig. 5. The change of HC and NOx emission with blends of diesel-propanol and diesel-butanol (Balamurugan and Nalini, 2014b)

Furthermore, in the study by Senthil Kumar, Thirumalini, and Praveen (Senthil Kumar *et al.*, 2019), the addition of n-butanol to B20 (waste cooking oil-based biodiesel) observed a reduction of 19% and 28% in NOx and CO_2 emissions, respectively. The employ of n-butanol blends reduced smoke emissions by 4.7% but increased HC emissions by 22%. The effect of isomers of butanol (1 -butanol, 2-butanol, and isobutanol) on the exhaust emissions and performance characteristics was analyzed by Fushimi et al. (Fushimi *et al.*, 2013). They obtained that increase in 1 -butanol content in diesel results in lower smoke emission, but low load conditions showed an increase of HC and CO, despite the longer ignition delay (Hoang, 2019). The stability of the operating conditions of the engine was found out except full load, especially with blending 50% by mass of 1 - butanol. Other results observed that the optimal blending ratio for 1-butanol in fuel blend was 40% which is the best among all butanol isomers. At low loads, HC and CO increased for all variants of blends. In addition, Zhou et al. (Zhou *et al.*, 2014) examined the combustion characteristics of the blend of 80% diesel and 20% vol. ABE consist of acetone-30%, butanol-60%, and ethanol-10% without water. Good combustion characteristics of ABE-diesel blend were reported because the presence of oxygen content is high which helps in oxidizing soot. Soot lift off the length and longer ignition delay allow more air entrainment upstream of the spraying lead to better mixing of fuel-air. Further found observed that ABE20 presents soot luminosity which is approximate zero with combustion efficiency enhancement as compared to pure diesel fuel at the condition of the low ambient oxygen content of 11% and 800 K of ambient temperature. ABE additive is also investigated in the study of Trindade et al. (Trindade and Santos, 2017) because its combustion characteristics are higher than that of n-butanol. The emission characteristics of n-butanol in comparison to pure diesel fuel were conducted by Zheng et al. (Zheng *et al.*, 2015). Negligible change in performance characteristics of the engine was detected in this research but the formation of soot and NOx declined. He et al. (He *et al.*, 2013) showed a slight decrement in formatting NOx emissions in most cases by increasing n-butanol content in the fuel. In addition, in a study by Xie et al. (Xie *et al.*, 2016), it is noticed that n-butanol can reduce CO_2 emissions in HCCI engines.

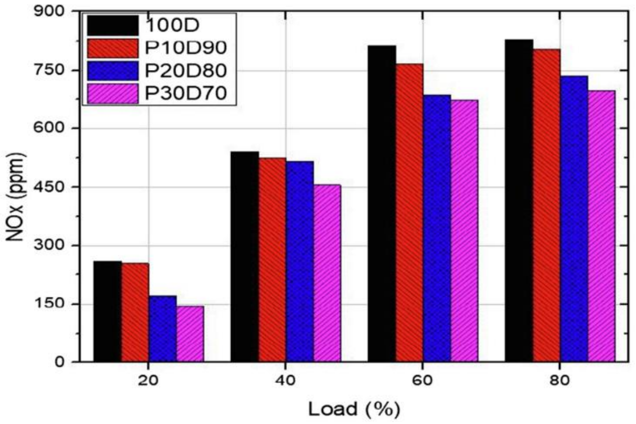

Fig. 6. The change of NOx from a CRDI engine with modified blends according to various loads (Santhosh *et al.*, 2020)

Pentanol has been showing off as a potential candidate for blending with diesel fuel (Ganesana *et al.*, 2022). Its outstanding features are blending stability, higher cetane number, and energy density than that of the reviewed alcohol-based additives. In terms of CO emissions, the increase in pentanol additives in diesel fuel has contributed to reducing CO emissions. The main reason for the increase in CO emissions is its higher latency heat that prompted the cooling mechanism for the air-fuel mixture in the cylinder (Saravanan, 2015). The results of Saravanan have revealed that NOx emissions tend to decrease with increasing pentanol additive in diesel fuel due to the decrease in-cylinder temperature caused by factors such as high vapor latency heat and low cetane number (Anh and Anh, 2019). However, under high load conditions, the cylinder temperature and NOx concentration increase. This can be explained as the role of oxygen in pentanol exhibited dominance with decreasing cetane number prolonging ignition delay which promotes the growth of fuel droplets during the premix combustion (Wei *et al.*, 2014). An experimental study evaluating the control effect of NOx emissions of pentanol-diesel blends on a common rail direct injection CRDI engine was investigated by Santhosh et al. (Santhosh *et al.*, 2020). The presence of 1-pentanol in the modified fuels has played an important role in the reduction of NOx emissions. P30D70 blend has recorded the lowest NOx emissions at rated load. Apparently, the synergistic effect of the higher latent heat of vaporization and lower cetane number caused a cooling effect in the combustion chamber, resulting in a decrease in the combustion temperature. As a result, NOx formation gradually decreased with increasing pentanol contribution. Figure 6 has shown that at 60% load, NOx emissions for P30D70 are 16.7% lower compared to diesel fuel. In summary, pentanol is a potentially sustainable alternative fuel to reduce harmful emissions, especially NOx. Studies have made recommendations for mixing up to 30% by volume of pentanol with diesel to supply CI engines with no engine modifications and additives (Biswajeet *et al.*, 2021).

4. Challenges and prospects

There are several challenges in the production of bio-alcohol fuels that many countries may be facing because the decarbonization efficiency is not as expected. The excessive use of fossil fuels in the production process as well as the use of edible biomass sources has reduced the attractiveness of the first generation of biofuels. Therefore, to meet the sustainable development strategies of energy and circular economy (Atabani *et al.*, 2021), second and third-generation biofuels have become promising candidates for green policy in industry and transportation (Dong *et al.*, 2021)(Xuan and Viet, 2021). The production of high alcohols and furan additives is more sustainable because of the greater involvement of renewable energy sources as well as lignocellulosic biomass feedstock (Huynh *et al.*, 2021). However, a comprehensive solution to maintaining commitments to combating global climate change would be sustainable energy policies along with a viable renewable energy transition strategy (Balasubramanian *et al.*, 2021b)(Thomas *et al.*, 2021). In particular, encouraging the development of electric vehicles is a solution that meets both medium and long-term goals (Tran *et al.*, 2020)(Subramanian *et al.*, 2021).

In the future, the combination of lower and higher alcohol additives to form tertiary or quaternary blends could be an interesting priority for further research to synergize their benefits in improving combustion characteristics as well as reducing emissions (Nayak *et al.*, 2021). Besides, solutions that combine organic additives and carbon and metal nanoparticles can be a promising proposal to promote the synergistic effect of additives in improving combustion quality and environmental aspects (Atarod *et al.*, 2020). Finally, although additive technology will achieve further breakthroughs in the future, it will be difficult to commercialize if the stability and durability issues of the engine are not thoroughly studied *(Tham et al.*, 2019)(Pham and Hoang, 2019b).

5. Conclusions

This work focuses on evaluating the significant effects of the chemical and physical properties of the five common alcohol additives on the emission characteristics of diesel engines. Changes in emission characteristics such as CO, NOx, and HC have been reported mainly in the presence of additives such as methanol, ethanol, propanol, butanol, and pentanol in diesel fuel. The impact of alcohol additives on the polluting emissions from diesel engines is considered and concluded as follows:

- More additive of methanol in diesel and biodiesel fuel has significantly improved the flame transmission rate, moreover, high latent heat stimulates a strong extinguishing effect, thus reducing NOx. Meanwhile, the use of ethanol mixed with diesel fuel can increase CO and HC emissions, due to the extremely high cooling effect of ethanol.
- All typical alcohol additives with high available oxygen content available enrich possibly the fuel blend with oxygen. Blend of diesel fuel with appropriate proportion of additives could result in emissions reduction because of proper characteristics of the combustion process.
- Two different trends of NOx emission related to alcohol additives are a controversial topic among researchers. Lower alcohol additives (methanol and ethanol) appear to be difficult to control for NOx increases, while higher alcohol additives have been found a positive effect on NOx reductions compared to unmodified fuels. The number of researches showed the growth in NOx emission which is explained by the reason of temperature increment in the combustion chamber. On the other hand, the decline of NOx was obtained by several researchers, its cause is possibly variation of different parameters which are exhaust gas recirculation, the number of additives, compression ratio, etc.
- It cannot affirm that all these additives can drop the concentration of emissions, due to the different trends are obtained in certain testing results. An optimal dosage for each fuel additive needs to be specified to curb the emission levels.

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