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REVIEW ARTICLE

Impacts of methanol fuel on vehicular emissions: A review

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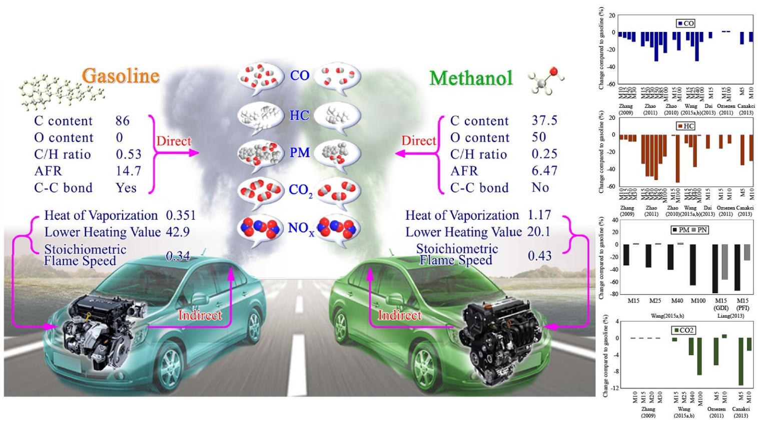
HIGHLIGHTS

- Methanol effectively reduces CO, HC, CO_2, PM, and PN emissions of gasoline vehicles.
- Elemental composition of methanol directly affects the reduction of emissions.
- Several physicochemical properties of methanol help reduce vehicle emissions.

GRAPHIC ABSTRACT



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ABSTRACT

The transport sector is a significant energy consumer and a major contributor to urban air pollution. At present, the substitution of cleaner fuel is one feasible way to deal with the growing energy demand and environmental pollution. Methanol has been recognized as a good alternative to gasoline due to its good combustion performance. In the past decades, many studies have investigated exhaust emissions using methanol-gasoline blends. However, the conclusions derived from different studies vary significantly, and the explanations for the effects of methanol blending on exhaust emissions are also inconsistent. This review summarizes the characteristics of CO, HC, NO_x, CO_2, and particulate emissions from methanol-gasoline blended fuels and pure methanol fuel. CO, HC, CO_2, particle mass (PM), and particle number (PN) emissions decrease when methanol-blended fuel is used in place of gasoline fuel. NO_x emission either decreases or increases depending on the test conditions, i.e., methanol content. Furthermore, this review synthesizes the mechanisms by which methanol-blended fuel influences pollutant emissions. This review provides insight into the pollutant emissions from methanol-blended fuel, which will aid policymakers in making energy strategy decisions that take urban air pollution, climate change, and energy security into account.

1 Introduction

Energy consumption and safety have been one of the most critical international issues since the Industrial Revolution. In 2019, global primary energy consumption increased by 1.3%, with coal still accounting for over 27% of the total energy consumption. Other major sources of energy consumption include oil (33%), natural gas (24.2%), hydroelectric (6.4%), renewable energy (5.0%), and nuclear (4.3%) (BP, 2020). And, the transportation sector accounts for a large portion of the total energy consumption. For example, it accounted for 35% of total energy consumption in International Energy Agency

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(IEA) countries in 2018 (IEA, 2021) and 28% in the USA (USEIA, 2021).

In recent decades, as atmospheric pollution and climate change have become increasingly important global environmental issues, more challenges have been posed in terms of simultaneously reducing the emissions of both atmospheric pollutants (CO, HC, NO_x, PM, etc.) and greenhouse gases (CO_2, CH_4, N_2O, etc.) (Pan et al., 2020). Automobiles are the main contributor to total pollutant emissions from road transport, a significant source of air pollution that harms human health and the environment. In 2019, CO, HC, NO_x, and PM emissions from automobiles in China exceeded 90% of total vehicle emissions. Among them, diesel vehicles accounted for 80% of NO_x emission and more than 90% of PM emission, while gasoline vehicles accounted for 80% of CO emission and more than 70% of HC emission of the total automobile emissions (MEE, 2020).

Facing such energy security and environmental issues, clean fuels' development and widespread use can be critical. Reduced emissions of greenhouse gases, PM, and other air pollutants, including CO, NO_x, and HC, is one purpose of using clean fuels (USEPA, 2010). Methanol fuel is one of the promising fuels among clean alternative fuels currently available. Methanol can be produced in a variety of ways as an alternative fuel. It can be produced from many sources, including coal, natural gas, coke oven gas, hydrogen, and biomass (Zhen and Wang, 2015). Unsurprisingly, coal plays a dominant role in primary energy consumption in countries with insufficient domestic oil production to meet energy demand. For instance, coal accounted for 57.7% of Chinese energy consumption in 2019, followed by oil (19.3%), natural gas (8.1%), and renewables (14.9%) (Hove et al., 2020). Other countries that rely heavily on coal as a primary energy source in 2019 include South Africa (70.6%), India (54.7%), Kazakhstan (53.9%), Vietnam (50.2%), Poland (44.6%), Indonesia (38.3%), and so on (BP, 2020). In these countries with an energy resource structure of "oil-poor, gas-little, coal-rich", developing and promoting coal-based methanol fuel is a realistic and feasible option for energy security and environmental protection.

Methanol has many desirable characteristics which make it an excellent vehicle fuel, such as high octane number, low stoichiometric air-fuel ratio, high heat of vaporization, high flame speed, and being liquid at standard temperature and pressure (Table 1).

Methanol fuel has a history of several decades. Since the 1970s, government agencies and automobile manufacturers in the USA, Japan, Canada, Germany, Sweden, and other countries have strongly encouraged the research of methanol vehicles to alleviate the increasing problems of energy security and environmental pollution. During the early stages of the methanol fuel research and development process, the USA was one of the countries

Table 1 Properties of gasoline and methanol

Property	Gasoline (typical)	Methanol
Chemical formula	C_5H_{12}-$C_{12}H_{26}$ (C_8H_{15})	CH_3OH
CAS Number	86290-81-5	67-56-1
Research Octane Number (RON)	95	109
Molecular weight (g/mol)	111.2	32.04
Oxygen Content (wt%)	0	50
Carbon Content (wt%)	86	37.5
Hydrogen Content (wt%)	14	12.5
Molar C/H ratio	0.53	0.25
Stoichiometric AFR (kg/kg)	14.7	6.47
Density (kg/m^3) 20 °C	760	790
Boiling Point (°C) 1 bar	27–225	64.5
(latent) Heat of Vaporization (MJ/kg) 25 °C	~0.351	1.17
Lower Heating Value (MJ/kg)	42.9	20.1
Higher Heating Value (MJ/kg)	48.0	22.9
Volumetric Energy Content (MJ/L)	31.7	15.9
Energy per Unit Mass of Air (MJ/kg)	2.92	3.09
Stoichiometric Flame Speed (m/s)	0.34	0.43
Adiabatic Flame Temperature (K)	~2275	2143
Quenching Distance (cm)	~0.2	0.18
Specific CO_2 Emissions (g/MJ)	73.95	68.44

Note: Source from Balki and Sayin, 2014; Sarathy et al., 2014; Liu et al., 2016; Sarathy et al., 2018; Verhelst et al., 2019; Balki et al., 2020.

with the most investment and the broadest range of vehicles. In California, M85 has been used for over 200 million miles with no health or safety problems (Bechtold et al., 2007). The price of crude oil declined in the 1990s as the oil supply remained consistent, while the price of natural gas used as a raw material for methanol production increased (Shen, 2010). While methanol has not become a substantial transportation fuel in the USA, its large industrial-scale use and the former availability of production methanol flex-fuel vehicle (FFV) have demonstrated that it is a viable fuel and technology exists for both vehicle application and fuel distribution (Bromberg and Cheng, 2010). In Sweden, Germany, and Norway, the shortage of oil resources led to methanol fleet trials in the 1970s and early 1980s. When the oil price dropped in 1986, interest in developing alternatives to crude oil-based fuels quickly waned, and large-scale promotion and usage of methanol fuel had to be halted due to these economic problems. The first generation of methanol-adapted vehicles performed very well in many circumstances (Mårald, 2010; Landälv, 2017; Schröder et al., 2020). Relying on the abundant coal resources in Shanxi and other provinces, China has conducted continuous research and applied coal-based methanol-gasoline blended fuel. Since the 1980s, China has successfully conducted trials and promoted M15 and M85 in several provinces. M100 has been used as taxi fuel in four cities in Shanxi since 2005 (Li et al., 2009). After entering the 21st century, although research into methanol fuel application in many countries has stagnated and many internal controversies have arisen, the research work in China is still underway.

In 2009, two national methanol-gasoline blends standards – "Fuel methanol for motor vehicles" (AQSIQ, 2009a) and "Methanol Gasoline (M85) for motor vehicles" (AQSIQ, 2009b) – were successively promulgated, ensuring the use of methanol fuel. A six-year Methanol Vehicle Pilot Project has been implemented since 2012, with positive results for all indicators, such as fuel economy, human health, and environmental impact (Zhao, 2019). Based on the favorable outcomes of the pilot project, in March 2019, eight ministries of the Chinese central government issued a promotional policy paper – "Guidance of Developing Methanol Vehicles Applications in Some Parts of China" – to develop methanol vehicles across China (Zhao et al., 2021). This policy demonstrates that the Chinese government is attempting to develop new fuel vehicles under the premise of promoting the electric drive strategy (Zhou et al., 2020). The International Organization of Motor Vehicle Manufacturers' Worldwide Fuel Charter (WWFC) Committee represents engine and vehicle manufacturers in European Automobile Manufacturers' Association (ACEA), Alliance of Automobile Manufacturers (Auto Alliance), The Truck and Engine Manufacturers Association (EMA), and Japan Automobile Manufacturers Association (JAMA) to harmonize global fuel quality criteria. In 2019, the Committee modified its charter to sanction the use of methanol-gasoline blends and permit methanol blending "where specified by applicable standards" thanks to substantial efforts by the Methanol Institute and its members (including Methanex). This update to the Charter shows growing acceptance and support for methanol among the leading automobile industry consortium and reflects increased global interest in fuel blending (Methanex, 2020).

Several review articles have previously summarized the results of studies using methanol as a fuel (Zhen and Wang, 2015; Ghadikolaei, 2016; Yusri et al., 2017; Awad et al., 2018; Verhelst et al., 2019; Göktaş et al., 2021). The influence of methanol on engines performances (brake torque, brake power, and so on) and combustion characteristics (combustion efficiency, anti-knocking, rate of heat release, brake mean effective pressure, and so on) has proven that methanol can be a good fuel component. However, researchers did not focus on a detailed and comprehensive investigation into the mechanism of methanol on regulated and CO_2 emission. The results of previous studies on emission using methanol-gasoline blended fuels vary differently; that is, whether it increases or decreases in comparison to gasoline and how much it drops if it decreases. Moreover, different mechanisms were proposed when explaining a similar phenomenon in different papers.

This review comprehensively summarizes the results of previous research on the vehicular emissions characteristics using methanol-blended fuels. Furthermore, we synthesize the mechanisms of methanol on these emissions and give a series of conclusions. This review will assist policymakers in considering the usage of methanol as gasoline vehicle fuel.

2 Characteristics of vehicular pollutant emission using methanol fuels

Since 2000, several studies on the emission characteristics of methanol as an alternative fuel for gasoline have been conducted mainly via Engine Bench Tests (EBTs) and Chassis Dynamometer Tests (CDTs). The EBTs enabled in-depth investigation of the methanol's effect on exhaust emissions under different engine operating conditions. In the majority of EBTs studies, the effects of methanol on pollutant emission were highly consistent, regardless of engine speed and load (Section 2.1). The CDTs are beneficial to investigate vehicular emission characteristics and evaluate the emission factors (EFs) using methanol fuel under actual driving conditions. Most CDTs studies showed that the EFs of vehicles fueled by methanol meet China 6a and Euro 6 standards (Section 2.2). Results from previous studies via both EBTs and CDTs have demonstrated that methanol content significantly impacts vehicular emissions.

2.1 Engine bench tests

Since the 1950s, the laboratory engine bench test has examined engine exhaust pollutants in the USA, Sweden, Germany, and other countries. The on-road vehicle driving state can be simulated on the engine bench. The test cycle for engine bench mainly includes: European steady-state cycle (ESC), European transient cycle (ETC), World harmonized transient cycle (WHTC), World harmonized steady-state cycle (WHSC), World harmonized not-to-exceed (WNTE) (Maricq, 2007; MEE, 2018; Gang et al., 2019). In this review, methanol-gasoline blended fuels are divided mainly into three categories: low-ratio blended fuels (30% or less), medium-ratio blended fuels (30%–85%), and high-ratio blended fuels (85% or more, including pure methanol).

Table 2 shows the results of previous studies on exhaust emission characteristics using methanol-blended fuels via EBTs.

Most EBTs results showed that the methanol-blended fuels reduced CO, HC, PM, and PN emissions compared to gasoline (Table 2). Elfasakhany (2015) studied regulated emissions of single-cylinder SI engines fueled with methanol-gasoline blended fuels below M10. In his research, using blended fuels resulted in lower CO and HC emissions than using gasoline in all engine conditions. He explained the reasons as follows: Firstly, oxygen in methanol enhances the combustion of blended fuels and reduces the amount of CO and HC emissions. Secondly, the leaning effect of the blended fuels due to their lower stoichiometric air/fuel ratio (AFR) promotes complete combustion and reduces CO and HC emissions. Thirdly, because the blended fuels have a relatively low boiling point, they are entirely vaporized and burned when injected into the combustion chamber. Fourthly, the relatively high latent heat of vaporization of blended fuels lowers the intake manifold temperatures, allowing the engine to intake more air for the process of fuel combustion. CO_2 emission increased when compared to gasoline because sufficient oxygen will enhance the CO-CO_2 and HC-CO_2 processes. According to Nuthan Prasad et al. (2020), CO and HC emissions from medium-ratio blended fuel (M50) were lower than gasoline at all test engine conditions due to oxygen-enriched methanol blends. Geng and Yao (2015) studied the PM and PN emissions in a four-cylinder PFI engine using low-ratio blended fuel (M15) and medium-ratio blended fuel (M45). Compared to gasoline, both PM and PN emissions decreased in M15 but increased in M45. The reasons for the decrease of particulate emission in M15 are as follows: Firstly, compared with gasoline, the high-temperature oxidation of methanol produces fewer unsaturated micro-molecules (e.g., C_2H_2), the precursor of the polycyclic aromatic hydrocarbons (PAHs). Secondly, methanol is hard to form carbonaceous particles during the combustion process. Thirdly, methanol has a lower molecular weight, a higher oxygen content, a faster flame speed, and no aromatic content than gasoline fuel. However, M45 emits more particulates than gasoline. The following are the reasons given in the study: Firstly, the exhaust tempe rature is lower. Secondly, blended fuels will combust incompletely due to the high latent heat of vaporization of methanol, increasing HC emission, especially when the cylinder temperature is relatively low in the high proportion of methanol. This increased HC emission could also form more amounts of particulates. But, in a study on the PN emission characteristics according to the ratio of methanol (M0 to M100) under the two conditions of M–G (PFI-methanol DI-gasoline) and G–M (PFI-gasoline DI-methanol) of a Dual-Fuel SI engine, blending of methanol always reduced PN emission, and the higher the content of methanol, the greater the reduction effect, up to more than 95% (Liu et al., 2015a; Liu et al., 2015b).

In studies on methanol-blended fuels, the methanol content is the most important parameter. Figure 1 shows the effects of different methanol content levels in methanol-blended fuels on various exhaust emissions. Wei et al. (2008) studied the regulated emission characteristics of blended fuels with varying methanol contents (M10, M20, M85) in a three-cylinder PFI engine. Their result showed that CO emissions were all reduced in low- and high-ratio blended fuels compared to gasoline, because methanol-blended fuels contain less carbon and more oxygen than gasoline. The oxygen enrichment coming from methanol causes a "pre-mixed oxygen effect" (Wu et al., 2004) to promote complete combustion. Compared to gasoline, NO_x emission was not significantly different in the low-ratio blended fuel but decreased by 60% to 80% in the high-ratio blended fuel (M85). The effects of the methanol blending on NO_x emission were discussed in terms of negative and positive aspects. On the one hand, the methanol blending increased the combustion temperature due to the fast flame propagation speed, which increased the NO_x emission. On the other hand, the high latent heat and the large gas heat capacity of the triatomic molecules – for the same amount of heat release, methanol has 49% more triatomic molecules in the combustion products than gasoline – reduced the peak combustion temperature, which reduced the NO_x emission. Because of these factors, the NO_x emission from low-ratio blended fuels (M10, M20) was approximately equal to those from gasoline, but NO_x emission from M85 was reduced (60%–80%) under the overall operating condition. Compared to gasoline, HC emission decreased in M10, increased in M20, and decreased by 20% in M85. When fueled with methanol-gasoline blends, the combustion process is accelerated, lowering the exhaust temperature. As a result, the post oxidation of HC was weakened, and

Table 2 Emission characteristics using methanol-blended fuels via EBTs

Test fuel	Engine type	Test condition	Effect on emission					References
			CO	HC	NO_x	CO_2	PM/PN	
M3, M7, M10	single-cylinder SI engine	1.3–1.6 kW, 2600–3450 r/min	↓	↓		↑		Elfasakhany, 2015
M3, M7, M10	single-cylinder SI engine	2600, 3400 r/min	↓	↓		↑		Elfasakhany2017
M10	single-cylinder SI engine	load 0–100%	↓	↓	↓	↓		Kak et al., 2015
M3–M15	four-cylinder SI engine		↓	↓	↑			Mallikarjun and Mamilla, 2009
M5, M10, M15	single-cylinder SI engine	500–1500 r/min	↓	↓	■	↓		Mishra et al., 2020
M10, M20	four-cylinder MPFI SI engine	10–60 Nm, 1500–3500 r/min	↓	Lower BMEP;↑ Higher BMEP;↓	NO↓		PN↓	Agarwal et al., 2014
M20	four-cylinder SI engine	1000–6000 r/min	↓	↓	↑	↑		Masum et al., 2014
M10–M30	three-cylinder PFI engine	2500 r/min, 10–30 Nm	↓	↓	=			Liu et al., 2007
M10, M20, M30	single-cylinder SI engine	0–1.4 kW	↓	↓	NO↑	↑		Farkade and Pathre, 2012
M15, M30, M50	four-cylinder SI engine	1000–4000 r/min	↓	↓		↑		Rifal and Sinaga, 2016
M50	single-cylinder SI engine	1200–1800 r/min	↓	↓	↓	↓		Nuthan Prasad et al., 2020
M10, M30, M60	single-cylinder SI engine	1200 r/min	↑	M10↑; M30↑; M60↓	↓			Li et al., 2017
M10, M20, M85	three-cylinder PFI engine	3000 r/min	↓	M10↓; M20↑; M85↓	M20=; M85↓			Wei et al., 2008
M15, M30	single-cylinder Dual-Fuel engine	M–G	↓	↑	↓		PM↓; PN↓	Kalwar et al., 2020
M15, M45	four-cylinder PFI engine	2000 r/min					PN, PM; M15↓; M45↑	Geng and Yao, 2015
M0–M100	four-cylinder Dual-Fuel SI engine	M–G, G–M					PN↓	Liu et al., 2015a; Liu et al., 2015b
M100	four-cylinder SI engine	1500–6000 r/min	↓		↓			Pourkhesalian et al., 2010
M100	single-cylinder SI engine	1500–3500 r/min	↓	↑	↓	↓		Çelik et al., 2011
M100	four-cylinder flex-fuel PFI engine	1500–4500 r/min	↓		↓	↓		Vancoillie et al., 2013
M100	four-cylinder HCCI engine	1200, 2400 r/min	↑					Maurya and Agarwal, 2014
M100	single-cylinder SI engine	1600–3600 r/min, Low power	↓	↓	↓	↑		Balki et al., 2014
M100	single-cylinder SI engine	2400 r/min, Low power	↓	↓	↓	↑		Balki and Sayin, 2014
M100	single-cylinder SI engine	1600–3600 r/min	BSCO↓	BSHC↓	BSNO↓	$BSCO_2$↑		Balki et al., 2016
M100	single-cylinder PFI engine	1500–4500 r/min, 10, 20 Nm	=		↓	↓		Turner et al., 2018
M57, M100	four-cylinder PFI engine	1500–3500 r/min, 40 Nm			↓			Turner et al., 2018
M56	single-cylinder DI engine	1500 r/min					Light load;PN ↑High load;PN ↓	Turner et al., 2018

Notes: ↓: Reduction compared to gasoline; ↑: Increase compared to gasoline; ■: Different results under different conditions, which means there is no rule; =: No significant changes compared to gasoline; M-G**:** PFI-methanol DI-gasoline; G–M**:** PFI-gasoline DI-methanol; BSCO: brake specific carbon monoxide; BSHC: brake specific unburned hydrocarbon; BSNO: brake specific nitrogen oxides; $BSCO_2$: brake specific carbon dioxide.

the HC emission increased. However, HC emission was reduced because M85 fuel does not contain heavy hydrocarbons that boil at high temperatures (Nakata et al., 2006). So, the HC emission produced by M85 was significantly lower than that produced by gasoline. Therefore, researchers concluded that the mixture of methanol reduces CO emissions. And for HC and NO_x emissions, factors that increase and decrease the emission act simultaneously. Li et al. (2017) studied M10, M30, and M60 in a single-cylinder SI engine in related work.

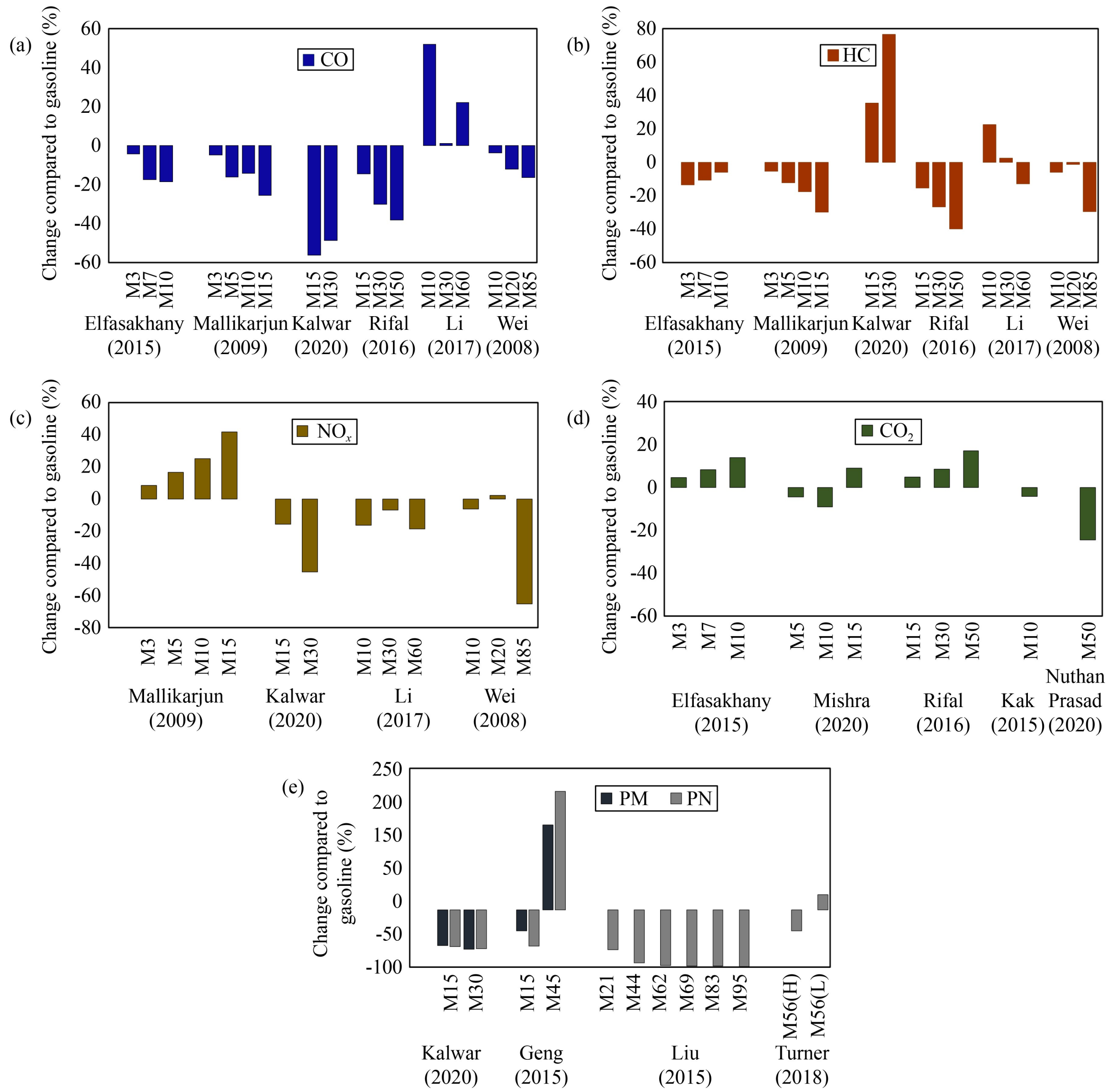

Fig. 1 The effect of methanol content on exhaust emission using methanol-gasoline blends (via EBTs). (a) CO. (b) HC. (c) NO_x. (d) CO_2. (e) PM and PN. Change compared to gasoline; A positive value indicates an increase in emissions compared to gasoline, and a negative value indicates a decrease.

The results showed that methanol-blended fuels increased CO emission compared to gasoline, contrary to most other researchers' results. Researchers explained that alcohol-containing fuels produced more triatomic products, which lowered the combustion temperature (Zhang et al., 2015) and further slowed down the oxidation process of CO emission. In addition, the shorter combustion duration of alcohol-gasoline blends may lead to insufficient oxygenation of CO, thus increasing CO emission. For HC emission, compared to gasoline, it decreased in M10 and M30 but increased in M60. In their study, the addition of methanol has both an increase and a decrease effect. The lower AFR of methanol led to more fuel injection, resulting in more fuel leaking into the crevice volumes or being absorbed and deposited in the oil layer, thereby increasing the HC emission. The oxygen content of alcohol fuel was beneficial to improving combustion quality, which resulted in a decrease in HC emission. NO_x emission was reduced because the combustion temperature of methanol-gasoline blended fuels was lowered due to insufficient fuel combustion. Rifal and Sinaga (2016) investigated the CO, HC, and CO_2 emission characteristics of M15, M30, and M50 of the four-cylinder SI engine. The higher the methanol content, the lower CO and HC emissions, and the higher the CO_2 emission. The decrease in the CO and HC emissions was explained by the higher oxygen content and lower carbon content of blended fuel. Mallikarjun and Mamilla (2009) studied the emission characteristics of CO, HC, and NO_x in a four-cylinder SI engine by adding methanol in various percentages at low ratios in gasoline (M3–M15). The CO, HC, and NO_x

emissions are correlated with the mixing ratio of methanol. As the blending ratio of methanol increased, the CO and HC emissions decreased while NO_x emission increased. Due to the presence of oxygen in methanol, sufficient oxygen was available for oxidation, and the combustion efficiency was improved, resulting in a reduction in CO and HC emissions. The oxygen content of methanol increased NO_x emission. Another reason for the increase in NO_x formation is that methanol has a higher peak cylinder pressure and temperature when compared to gasoline. Liu et al. (2007) conducted a study on exhaust emission characteristics of low-ratio blended fuels (M10–M30). They found that CO and HC emissions decreased in all methanol-ratio tested, while there was no significant difference in NO_x emission compared to gasoline.

Several researchers compared the exhaust emission characteristics of pure methanol (M100) as fuel versus gasoline (Fig. 2). Many studies have shown that M100 reduces CO, HC, and NO_x emissions when compared to gasoline. Pourkhesalian et al. (2010) studied the CO and $BSNO_x$ emission characteristics of M100 at different engine speeds of the four-cylinder SI engine and found an evident reduction effect. The decrease in CO emission was attributed to the low stoichiometric AFR and carbon/hydrogen ratio (C/H ratio) of methanol. The $BSNO_x$ emission was reduced by 53% because the methanol has a lower heating value, a faster flame speed, a lower spark advance, and a lower combustion temperature. Çelik et al. (2011) studied the CO, HC, NO_x, and CO_2 emission characteristics of M100 under different engine speeds and CR conditions of a single-cylinder SI engine. Compared to gasoline, CO emission was significantly reduced due to the presence of oxygen in methanol. NO_x emission was also reduced by relatively low combustion temperature due to higher latent heat of vaporization of methanol, and CO_2 emission is reduced due to lower C/H ratio and carbon content of methanol. However, HC emission increased significantly compared to gasoline, which was

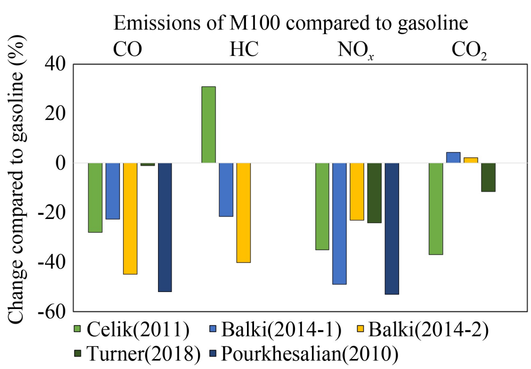

Fig. 2 Emission characteristics of M100 compared to gasoline. Change compared to gasoline; A positive value indicates an increase in emissions compared to gasoline, and a negative value indicates a decrease.

explained by the fact that pure methanol reduces the cylinder temperature as the heat of vaporization of methanol was higher (about 3.0 times) than that of gasoline, resulting in a misfire and partial burn in the regions near the combustion chamber wall. According to Vancoillie et al. (2013), using M100 reduced CO, NO_x, and CO_2 emissions compared to gasoline. Because of the oxygenation nature of methanol, complete combustion occurred, resulting in the decline of CO emission compared to gasoline. The decrease of NO_x emission was due to the combustion temperature of methanol being 2%–7% lower than that of gasoline. Also, methanol's CO_2 formation per unit energy was lower than gasoline, and the brake heat efficiency was higher, so CO_2 emission decreased by over 10%. In other studies, CO, HC, and NO_x emissions using M100 decreased significantly while CO_2 emission slightly increased. The decrease in CO and HC emissions was attributed to methanol's oxygen content, carbon content, and laminar flame speed. The decrease in the NO_x emission was driven by the lower adiabatic flame temperature and higher heat of vaporization of methanol than gasoline, which caused the reduction of the cylinder's peak temperature. The CO_2 emission was slightly increased compared to gasoline because of the oxygen content of methanol, and the increase was less than 5% (Balki and Sayin, 2014; Balki et al., 2014; Balki et al., 2016). Turner et al. (2018) studied CO, NO_x, and CO_2 emission characteristics of M100 in a single-cylinder PFI engine, NO_x emission characteristics of M57 and M100 in a four-cylinder PFI engine, and PN emission characteristics of M56 in a single-cylinder DI engine. In their study, no significant changes in CO emission were observed. The lower peak combustion temperature of methanol led to a reduction in NO_x emission. Methanol improved the performance of the combustion system, has a lower CO_2 formation per unit energy, and increasing the brake thermal efficiency (BTE), resulting in CO_2 emission decreased by over 10% compared to gasoline. At a light load, the PN emission using M56 increased compared to gasoline because the blending of methanol increased the vapor pressure, resulting in flash-boiling of the fuel on injection and inferior mixture preparation. At a high load, the PN emission using M56 was smaller than that of gasoline. That might be because less flash boiling occurs at higher cylinder pressure. Therefore, the factor for reducing the concentration of intermediate species, which were essential to the formation of precursors to soot by oxygen in the fuel (Litzinger et al., 2000), was effective.

Increased engine load led to an increase in temperature, which affects the exhaust emission characteristics. Agarwal et al. (2014) studied the CO, HC, NO, and PN emission characteristics of low-ratio blended fuels (M10, M20) at different load conditions in a four-cylinder MPFI SI engine. CO, NO, and PN emissions were lower in all loads when blended fuels were used instead of gasoline.

HC emission increased at low loads and decreased at high loads. Researchers explained that the mixture is too lean to burn and sustain flames at low loads. Also, due to methanol's relatively high latent heat of vaporization, the in-cylinder temperature gets lowered, increasing the HC emission than gasoline. Still, this tendency might reverse at high loads. Farkade and Pathre (2012) found that CO and HC emissions from methanol-blended fuels (M30 or less) were lower than gasoline under all load conditions, while NO_x and CO_2 emissions increased. However, Kak et al. (2015) observed that using methanol-blended fuel (M10) reduced the emissions of CO, HC, NO_x, and CO_2 compared to gasoline under all load conditions.

With increasing engine speed, the cylinder temperature is rising, affecting the exhaust emission characteristics. The increase and decrease in emissions from methanol-blended fuels varied with engine speed. Still, there was no reversal phenomenon (that is, according to the engine speed, the emissions increase and then decrease or decrease and then increase compared to gasoline). Masum et al. (2014) studied exhaust emissions from M20 at different engine speeds of 1000 to 6000 r/min. The exhaust gas temperature (EGT) is a significant indicator of cylinder temperature. When the engine speed was less than 3000 r/min, the EGT was lower with methanol-blended fuel than with gasoline, but the effect was reversed at engine speeds above 4000 r/min. However, CO and HC emissions were reduced in all engine speeds while NO_x and CO_2 emissions increased compared to gasoline. The benefits of HC emission reduction effects were greater at low engine speeds (less than 3000 r/min), but the increase of CO_2 emission was similar at all engine speeds. In addition to the oxygen content, the methanol-blended fuel needs to be consumed more than gasoline to obtain the same engine power, increasing carbon flow rates (Melo et al., 2012), another reason for the increased CO_2 emission. Rifal and Sinaga (2016) found that methanol-blended fuels (M50 or less) reduced CO and HC emissions while increasing CO_2 emission at all engine speeds (1000 to 4000 r/min) when compared to gasoline. In Elfasakhany's (2017) study, the emission characteristics of low-ratio blended fuels (M10 or below) at 2600 r/min and 3400 r/min engine speeds showed a downtrend in CO and HC emissions compared to gasoline, with the better reduction effects at higher engine speeds. But it showed the uptrend of CO_2 emissions, and the increase in CO_2 emission compared to gasoline weakened at higher engine speeds. Meanwhile, according to Mishra et al. (2020), methanol-blended fuel (M10) showed a reasonable reduction in CO, HC, and CO_2 emissions at all engine speeds tested (500 to 1500 r/min), and NO_x emission was lower than that using gasoline in some cases.

The GDI engine has several advantages over the PFI engine, such as higher power output, greater fuel economy, and lower CO_2 emission, contributing to its more reliable and preferred technology. However, the relatively high PM emission of the GDI engine remains a major disadvantage. This disadvantage can be solved by using methanol-blended fuel in the GDI engine. Kalwar et al. (2020) studied the CO, HC, NO, PM, and PN emission characteristics of low-ratio blended fuels (M15, M30) in a single-cylinder Dual-Fuel GDI engine (PFI alcohol + DI gasoline). Compared to gasoline, CO emissions using two blended fuels were reduced due to the relatively low stoichiometric AFR. And NO_x emissions were reduced because of the decrease in cylinder temperature. The higher the methanol content, the greater the effect of decreasing the cylinder temperature. CO emission using M30 increased more than M15, and NO_x emission decreased more than that of M15. HC emission using the blended fuel slightly increased compared to gasoline. It was suggested that the homogeneous alcohol-air mixture during compression stroke might have enhanced the trapping of unburnt fuel in the crevices, leading to higher HC emission. PM and PN emissions using blended fuel decreased compared to gasoline. The reasons are as follows: Firstly, the presence of alcohol resulted in relatively higher flame speed, resulting in less soot formation due to less time available during combustion. Secondly, fuel-bound oxygen of alcohol also contributed to PM reduction by improving soot oxidation. Their study suggested dual-fuel operation could be a potential solution for GDI engines to reduce PM emission.

As described above, the results of several exhaust emission characteristics using methanol-blended fuels studied via EBTs are different. There are various explanations for the mechanism, which are summarized in Section 3.

2.2 Chassis dynamometer tests

The Chassis dynamometer mainly includes a road simulation system, power absorption system, inertial simulation system, dynamometer system, and emission measurement system. The test cycle used in CDTs has a more accurate simulation effect than EBTs. The CDTs were initially used for vehicle power, fuel economy, and vehicle assembly inspections. With the increase of vehicle exhaust pollution, CDTs began to be applied to vehicle emission tests (Durbin et al., 2002; Huai et al., 2004; Beddows and Harrison, 2008). The test cycle for chassis dynamometer mainly includes Federal test procedure (FTP), New European driving cycle (NEDC), Worldwide harmonized light vehicles test cycle (WLTC), China automotive test cycle (CATC) (Gong et al., 2017).

Table 3 shows the results of the research on regulated and CO_2 emission characteristics via CDTs.

The majority of CDTs results indicated that the methanol-blended fuels (including M100) reduced CO, HC, CO_2, PM, and PN emissions compared to gasoline,

Table 3 Emission characteristics of methanol-blended fuels via CDTs

Test fuel	Vehicle	Test condition	Effect on emission: CO	HC	NO_x	CO_2	PM/PN	References
M10, M15, M20, M30	Euro IV car	NEDC	↓	↓	↓	=		Zhang et al., 2009
M15	Euro IV passenger car	NEDC25±2 °C	↓7%, 0.567^{EF}	↓16%, 0.072	↑85%, 0.133			Dai et al., 2013
M15, M25, M40 (production)	GDI engine car	NEDC22.0 ±0.5 , 24%±2%	↓9.4%–33.2% (0.28–0.21)	↓9.7%–36.9% (0.06–0.04)	M15↓M40= (0.012–0.017)	↓0.8%–4.1%	PM↓ 33.2%–40.2% PN↑	Wang et al., 2015b
M15, M20, M30, M50, M85, M100	Four passenger cars	NEDC25±2 °C	↓11%–34% (0.55–1.05^{EF})	↓10%–49% (0.05–0.15)	↑53%–474% (0.02–0.58)			Zhao et al., 2011
M15, M100	Two passenger cars	NEDC25±2 °C	M15↓9% M100↓21%	M15↓1% M100↓55%	M15↑175% M100↑233%			Zhao et al., 2010
M15	GDI engine car and PFI engine car	Cold start NEDC25±2 °C 45.0%±2%					GDI:PM↓78% (0.003)PN↓56%(2×10^{12} /km) PFI:PM↓74% (0.001)PN↓25% (7.5×10^{11} /km)	Liang et al., 2013
M5, M10	1.4i SI engine car	Vehicle speed: 40–100 km/h	↑	↓	40 km/h:↑ 100 km/h:↓	=		Ozsezen and Canakci, 2011
M5, M10	1.4i SI engine car	Vehicle speed:80, 100 km/h	80 km/h:↓14% 100 km/h: M5↓6% M10↑3%	↓10%–35%	80 km/h:↓9% 100 km/h: M5↓M10↑2.8%	↓M5:11.3% M10:3%		Canakci et al., 2013
M100	China V Dual-Fuel passenger car	NEDC22.0±0.2 °C 45.0%±0.3%	↓11.2% (0.35)	= (0.040)	= (0.011)	↓8.8% (160)	↓65.5% (1.25)	Wang et al., 2015a
M100	Methanol-fueled China Vcar	NEDC25–27 °C 30%–40%	(0.33)	(0.17)	(0.14)			Wang et al., 2016
M100	Six China IV methanol taxis		0.326–0.911	0.051–0.0799	0.0226–0.0386		PN0.65–2.71 $\times10^{11}$ /km	Su et al., 2020
M15	Three motorcycles	UDC20–30 °C	↓63%–84% 0.13–0.21	↓11%–34.5% 0.12–0.19	↑76.9%–107.7%0.21–0.27			Li et al., 2015
M30	Motorcycle	5000–8500 r/min	↓	↓		↓		Sugita et al., 2019
M85	Motorcycle	Low speed: 0–60 km/h High speed: 61–120 km/h	Low speed:↑ High speed:↓	Low speed:↑ High speed:=	↑			Agarwal et al., 2020

Notes: ↓: Reduction compared to gasoline; ↑: Increase compared to gasoline; =: No significant changes compared to gasoline; EF Emission factor identified in the literature (unit: g/km), and number in parentheses means emission factor inferred from the figure in the literature; percentage values are changes in emissions compared to gasoline.

but it is difficult to conclude that NO_x emission increases or decreases (Table 3). Up to now, most of the studies on exhaust emission characteristics of methanol-blended fuels via CDTs have been conducted on NEDC. Zhang et al. (2009) studied exhaust emission characteristics of low-ratio blended fuels (below M30) with EURO IV vehicle. Compared to gasoline, CO, HC, and NO_x emissions from methanol-blended fuels decreased. Additionally, there was no discernible difference in CO_2 emissions between methanol-blended fuel and gasoline. EFs of CO, HC, and NO_x using M10, M20, and M30 were generally lower than those using gasoline in ECE and EUDC cycle. Dai et al. (2013) studied the CO, HC, and NO_x emission characteristics of low-ratio blended fuel (M15) with Euro IV passenger cars and evaluated EFs. Compared to gasoline, CO emission decreased by 7% and HC emission by 16% due to promoted complete combustion of methanol with higher oxygen content and lower carbon content. NO_x emission increased by 85% with more oxygen and higher combustion temperature. The EFs of M15 were 0.567 g/km for CO, 0.072 g/km for HC, and 0.133 g/km for NO_x. Wang et al. (2015a) studied the emission characteristics of CO, HC, NO_x, CO_2, and PM of M100 with a China V dual-fuel passenger car compared to gasoline mode and evaluated EFs. In both modes of gasoline and methanol, the test vehicle was ignited with gasoline. Compared to the gasoline mode, the M100 mode reduced CO emissions by 11.2%, CO_2 emissions by 8.8%, PM emissions by 65.5%. For HC and NO_x emissions, there was no evident difference between the two modes. According to researchers, practically all the HC emissions from a China V certificated model were emitted during the cold-start phase because of poor vaporization of liquid fuel and hence incomplete combustions, implying that HC emission from methanol fuel is not significantly different from gasoline. Two reasons for the decrease in CO_2 emissions using M100 are as follows: Firstly, methanol produces less CO_2 per unit energy than gasoline. Secondly, the relatively higher latent heat of vaporization of methanol was beneficial to cooling intake charge down, and faster flame speed promoted the anti-knock quality of methanol fuel. These features enabled advanced spark timing, which prolonged constant-volume combustion duration in the chambers improved the engine's thermal efficiency. The higher engine efficiency contributed to tailpipe CO_2 reduction. PM emission from M100 decreased because methanol did not form precursors of PAHs, which are important intermediates for forming particulate matter, due to the absence of C-C bonds in its chemical structure. The EFs of M100 were approximately 0.35 g/km for CO, 0.040 g/km for HC, 0.011 g/km for NO_x, 160 g/km for CO_2, and 12.5 mg/km for PM, respectively.

According to the results of the study via CDTs, the effects of methanol contents on reducing CO, HC, and PM emissions are generally evident (Fig. 3). Zhao et al. (2011) studied the CO, HC, and NO_x emission characteristics of blended fuels in the whole range of low-ratio (M15, M20, and M30), medium-ratio (M50), and high-ratio (M85 and M100) blended fuels using four-passenger cars. CO and HC emissions decreased compared to gasoline in all the methanol content bands, and the higher the methanol content for the same vehicle, the more effective the reduction effect. On the contrary, NO_x emissions increased compared to gasoline. As the methanol content increased, the NO_x emissions increased. From the exhaust emission contributions of blended fuels from each stage of the entire NEDC test cycle (ECE and EUDC), the majority of CO emission was produced during the first ECE (also known as UDC) cycle with gasoline. The contributions to total CO emissions from the first ECE cycle were consistently over 90% in methanol-gasoline fueled cars, which was greater than that in gasoline-fueled cars. HC emissions followed a similar pattern to CO, although there was no discernible difference between gasoline-fueled cars and methanol-gasoline fueled. Vehicles fueled with methanol-gasoline blended fuels emitted low NO_x in the ECE cycles because the temperature of the engine cylinder was not high enough to produce NO_x, but reached a maximum value in the EUDC cycle, which is 60%–70% of total NO_x emission. The EFs were 0.55–1.05 g/km for CO, 0.05–0.15 g/km for HC, and 0.02–0.58 g/km for NO_x.

Another study examined the CO, HC, and NO_x emission characteristics of the M15 and M100 using two passenger cars. Due to the "pre-mixed oxygen effect" caused by the lower carbon content and the presence of oxygen in methanol, methanol blending promoted complete reaction, resulting in an apparent reduction in CO and HC emissions. In particular, in the absence of heavy HCs in methanol, HC emissions were reduced by 55% in M100. NO_x emissions using methanol-blended fuels increased significantly due to the fast flame propagation speed and the enhanced combustion temperature. On the other hand, the high latent heat and the large gas heat capacity of triatomic molecules limit the peak combustion temperature, reducing NO_x emissions, but it did not play a leading role in the study. The decrease of CO and HC emissions and the increase of NO_x emission were observed when the methanol content was increased from 15% to 100% (Zhao et al., 2010).

Methanol vehicles powered by pure methanol (M100) showed good regulated and CO_2 emission characteristics. In Wang et al. (2016) study, the EFs of M100 were evaluated as 0.33 g/km for CO, 0.17 g/km for HC, and 0.14 g/km for NO_x, respectively. Su et al. (2020) evaluated the EFs of M100 using six high-mileage, China IV methanol taxis, and found that they were 0.326–0.911 g/km (Average: 0.655±0.211 g/km) for CO, 0.051–0.0799 g/km (Average 0.066±0.010 g/km) for HC, 0.0226–0.0386 g/km (Average 0.0227±0.0062 g/km) for NO_x, 0.65–2.71× 10^{11} /km (Average 1.44±0.73× 10^{11} /km) for PN.

As mentioned above, GDI engines have become the preferred technology. Wang et al. (2015b) examined the CO, HC, and NO_x emission characteristics of M15, M25,

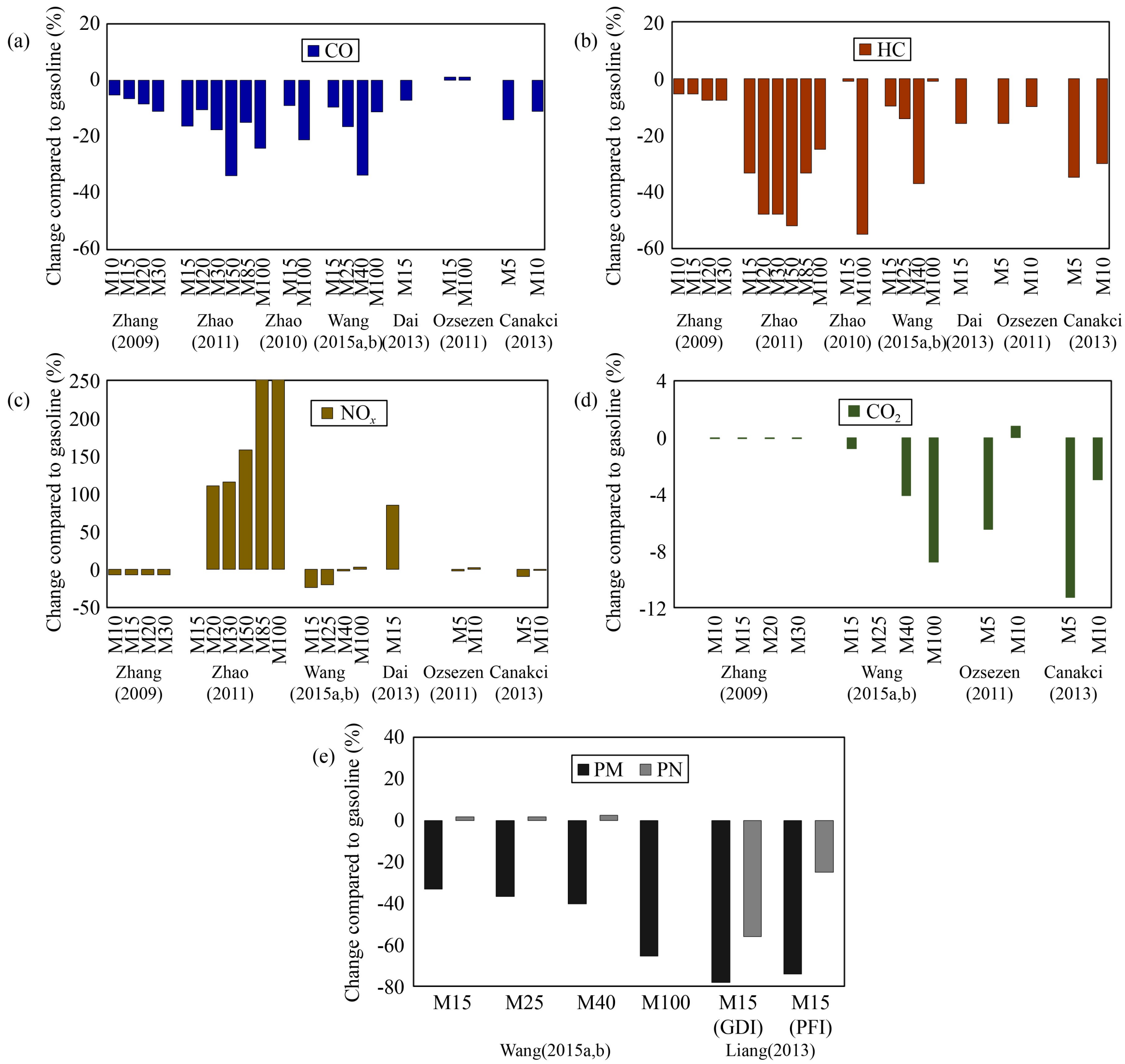

Fig. 3 The effect of methanol content on exhaust emission using methanol-gasoline blends (via CDTs). (a) CO. (b) HC. (c) NO_x. (d) CO_2. (e) PM and PN. Change compared to gasoline: A positive value indicates an increase in emissions compared to gasoline, and a negative value indicates a decrease.

and M40 (commercial product) passenger cars equipped with a GDI engine and evaluated EFs. The results indicated that CO, HC, and NO_x emissions using methanol-blended fuels were all reduced compared to gasoline (Fig. 3). The reasons for reducing CO and HC emissions were explained by the lower stoichiometric AFR, molar C/H ratio of methanol, and the absence of C-C bond in the methanol molecule. For NO_x emission, the study described the effects of decreasing emission and increasing emission acted simultaneously. The effect of decreasing emission is due to the relatively low adiabatic flame temperature and the relatively high vaporization heat value of methanol. The effect of increasing emission is due to driving the in-cylinder air-fuel mixtures to a slightly lean side in blended fuel. In the study, commercial products were especially used as methanol-blended fuels, and they contained various additives. Researchers explained that additives in blended fuels shared a tiny percentage of their mass or volume. But their function in the processes of air-fuel mixing and burning, particularly during the warm-up phase of TWC, would have vital impacts on NO_x and HC concentration at the tailpipe. This suggests that additives in methanol-blended fuels may affect the regulated emission characteristics. The EFs of blended fuels were 0.28–0.21 g/km for CO, 0.06–0.04 g/km for HC, and 0.012–0.017 g/km for NO_x. Liang et al. (2013) studied the emission characteristics of PM and PN of M15 over the cold start NEDC test cycle with a GDI engine passenger car and a PFI engine passenger car and evaluated EFs. Like gasoline, PM and PN emissions from GDI engine vehicles were higher than those from PFI engine vehicles when using methanol-blended fuel. Compared to gasoline, methanol-blended fuel had a pronounced reduction in PM and PN emissions (PM: 74%–78%, PN: 25%–56%). The study indicated that promoted complete combustion proceeded due to the "pre-mixed oxygen effect" caused by the higher oxygen content of methanol. Methanol did not have aromatic components that were prone to soot production. The EFs were 3 mg/km for PM and 2×10^{12}/km for PN in GDI engine vehicles, and 1 mg/km for PM and 7.5×10^{11}/km for PN in PFI engine vehicles.

Some researchers have investigated the exhaust emission characteristics of methanol-blended fuels at different speeds. Compared to gasoline, CO and NO_x emissions using methanol-blended fuels (M5, M10) slightly increased or slightly decreased depending on the test conditions (methanol content and vehicle speed), and some test conditions showed apparent reduction effects (14% decrease for CO and 9% for NO_x). Overall, the study found a decreasing tendency in NO_x emission when methanol blends were used in place of pure gasoline at all vehicle speeds. Compared to gasoline, HC emissions from methanol-blended fuels decreased at all vehicle speeds by up to 35%. CO_2 emissions also decreased in most conditions, reducing using M5 (11.3%) higher than M10 (3%). CO emissions could be reduced by the leaning effect due to the oxygen content of methanol, and HC emissions decreased by enhancing the combustion effect by the oxygen content of methanol. HC emissions using M10 rather increased than those using M5 due to the quenching effect caused by the high heat of vaporization of methanol. Due to the higher latent heat of vaporization and lower heating value of methanol, the combustion temperature was dropped, resulting in reduced NO_x emission. Additionally, M10 had a larger oxygen content than M5, which increased its NO_x emission. CO_2 emissions decreased due to the molar C/H ratio and lower carbon content (Ozsezen and Canakci, 2011; Canakci et al., 2013).

The results of studies on motorcycles fueled with methanol-blended fuel via CDTs have also been reported. The blending of methanol reduced motorcycles' CO, HC, and CO_2 emissions (Table 3). Li et al. (2015) studied the emission characteristics of CO, HC, and NO_x of motorcycles fueled with M15. Compared to gasoline, M15's CO emissions decreased by 63%–84%, HC emissions decreased by 11%–34.5%, and NO_x emissions increased by 76.9%–107.7%. The study showed that the higher oxygen content of methanol-blended fuel made combustion more complete, which reduced CO and HC emissions. In contrast, promoted complete combustion and higher combustion temperature caused by the faster flame propagation rate of methanol increased NO_x emission. The EFs of M15 in motorcycles were 0.13–0.21 g/km (standard limit value 2.0) for CO, 0.12–0.19 g/km (standard limit value 0.8) for HC, and 0.21–0.27 g/km (standard limit value 0.15) for NO_x. Sugita et al. (2019) investigated the exhaust emission characteristics of M30 for motorcycles, and CO, HC, and CO_2 emissions decreased compared to gasoline. Agarwal et al. (2020) studied the regulated emission characteristics of M85 at different speeds of a motorcycle. Compared to gasoline, NO_x emissions increased at low and high speeds, while CO and HC emissions increased at low speeds and decreased at high speeds. The study suggested that at low vehicle speeds (0–60 km/h), the in-cylinder temperature

Table 4 Mechanism of methanol on CO emission

Impacts of methanol	Mechanism	Class	References
Methanol reduces CO emission	Methanol has a lower carbon content than gasoline. The carbon in the fuel is directly converted into CO during the combustion process, so the use of methanol-blended fuel reduces the formation and emission of CO.	Direct	Wei et al., 2008; Zhao et al., 2010; Dai et al., 2013; Balki et al., 2014; Rifal and Sinaga, 2016
	Methanol has a lower C/H ratio than gasoline.	Direct	Pourkhesalian et al., 2010; Wang et al., 2015b
	The oxygen enrichment from methanol leads to a "pre-mixed oxygen effect" that promotes complete combustion.	Direct	Liu et al., 2007; Wei et al., 2008; Mallikarjun and Mamilla, 2009; Zhao et al., 2010; Çelik et al., 2011; Zhao et al., 2011; Farkade and Pathre, 2012; Dai et al., 2013; Vancoillie et al., 2013; Agarwal et al., 2014; Balki and Sayin, 2014; Elfasakhany, 2015; Li et al., 2015; Rifal and Sinaga, 2016; Elfasakhany, 2017
	The lower stoichiometric AFR of methanol leads to the leaning effect of the methanol-blended fuel, and this promotes complete combustion.	Direct	Qi et al., 2005; Pourkhesalian et al., 2010; Canakci et al., 2013; Masum et al., 2014; Elfasakhany, 2015; Wang et al., 2015b; Elfasakhany, 2017; Kalwar et al., 2020
	Methanol has no C-C bond in its structure, which could help complete the combustion of the methanol-blended fuel.	Direct	Wang et al., 2015b
	The lower boiling point of methanol makes the methanol-blended fuel completely vaporize, allowing for complete combustion.	Indirect	Elfasakhany, 2015; 2017
	The higher heat of vaporization of methanol-blended fuel leads to lower intake manifold temperatures, and more air access occurs during fuel combustion.	Indirect	Elfasakhany, 2015; 2017
Methanol increases CO emission	Methanol-blended fuel produces more triatomic products, which lowers the combustion temperature and slows down CO oxidation.	Indirect	Li et al., 2017
	The shorter combustion process of methanol-blended fuel might result in insufficient oxygenation of CO.	Indirect	Li et al., 2017

of the engine was relatively low. At the same time, the latent heat of vaporization of the M85 was relatively higher, which may cause incomplete vaporization and consequently increase the CO and HC emissions. Also, at high vehicle speeds (61–120 km/h), the in-cylinder temperature of the engine was high enough for methanol in the fuel to vaporize so that there was no difference in HC emission. Because the presence of oxygen in methanol can promote more effective oxidization, CO emission from M85 was further reduced compared to gasoline.

As via EBTs, in studies via CDTs, there exist various mechanisms that methanol affects vehicular emissions, and these mechanisms are summarized in Section 3.

3 How methanol affects emission

In this section, the mechanisms of each pollutant concluded by researchers are synthesized and interpreted.

3.1 CO

CO is a byproduct of incomplete fuel combustion in the engine combustion chamber and represents the loss of chemical energy that is not fully utilized in the engine (Canakci et al., 2009). CO emissions are increased in conditions such as locally rich fuel, insufficient oxidant (i.e., air), low combustion temperature, and interruption of combustion cycle time (Liu et al., 2007; Li et al., 2017).

Table 4 summarizes the mechanisms of methanol on the increase and decrease of CO emissions.

Almost all the research showed that mechanisms of reducing CO emissions were applied when methanol was used as a fuel component of gasoline engines. Compared to gasoline, methanol can ensure a more complete combustion of the fuel. This is due to methanol's elemental composition characteristics (e.g., lower carbon content, presence of oxygen, lower C/H ratio, etc.), structural characteristics (e.g., non-presence of C-C bond), and physicochemical characteristics (e.g., lower boiling point, etc.). Therefore, it is considered in this paper that a decrease in CO emission is natural when methanol is used as an alternative to gasoline.

3.2 HC

HC emission is an unburned or incomplete combustion product of fuel in the combustion chamber. Like CO, it represents the loss of chemical energy that is not fully utilized in the engine (Canakci et al., 2013). HC is emitted in such conditions as misfires, regulated valve leakage, liquid fuel effects (especially during cold start and warm-up), fuel or fuel/air mixture protected from the combustion process in crevices, oil films and deposits, lack of complete air supply, low cylinder compression, lean mixture and lower in-cylinder temperature (Kaiser et al., 1991; Cheng et al., 1993; Kaiser et al., 1994; Stanglmaier et al., 1999; Farkade and Pathre, 2012; Kak et al., 2015; Kalwar et al., 2020).

Table 5 summarizes the mechanisms of methanol on the increase and decrease of HC emissions.

Since methanol is free of unsaturated hydrocarbons, it reduces aromatics, while increases carbonyls (especially formaldehyde). In most studies, total HC emissions were reduced when methanol was used as a fuel component of gasoline engines, anyway. HC is a byproduct of incomplete combustion of fuel. Methanol blending can promote complete combustion of fuel. As with CO, it is considered in this paper that a decrease in HC emission is natural when methanol is used as an alternative to gasoline.

3.3 NO_x

NO_x in the regulated emissions, a mixture of NO and NO_2, is formed by the oxidation of nitrogen from the air in the combustion chamber. The formation of NO_x is strongly dependent on the combustion temperature and in-cylinder temperature, the oxygen concentration, and residence time in the combustion chamber (Ajav et al., 1998; Ozsezen et al., 2011; Masum et al., 2013; Maurya and Agarwal, 2014; Nuthan Prasad et al., 2020).

Table 6 summarizes the mechanisms of methanol on the increase and decrease of NO_x emissions.

When methanol is used as a fuel component, the combustion temperature and in-cylinder temperature are reduced due to various physicochemical properties such as lower adiabatic flame temperature and higher latent heat of vaporization of methanol, and higher triatomic molecular content in the combustion products. So that NO_x emissions can be reduced. On the one hand, NO_x emissions may increase as a result of methanol's characteristics such as oxygen and the faster flame propagation speed. This paper admits that research results differ between researchers as the enhancement and attenuation of these effects were complicated according to the methanol content.

3.4 CO_2

CO_2 is produced by the complete combustion of fuel. The formation is affected by the C/H ratio of fuel (Farkade and Pathre, 2012; Masum et al., 2014).

Table 7 summarizes the mechanisms of methanol on the increase and decrease of CO_2 emissions.

In CDTs studies, all showed decreased results or no apparent difference with gasoline. But in EBTs studies, the CO_2 emissions from methanol-blended fuels increased or decreased. In some EBTs, the gradients in engine speed are generally more gradual than in CDTs, so that the CO_2 emissions may be highly evaluated. Since methanol produces less CO_2 formation per unit energy

Table 5 Mechanism of methanol on HC emission

Impacts of methanol	Mechanism	Class	References
Methanol reduces HC emission	Methanol has no heavy HC.	Direct	Wei et al., 2008; Zhao et al., 2010
	The oxygen in methanol provides sufficient oxygen, and this improves combustion efficiency.	Direct	Mallikarjun and Mamilla, 2009; Zhao et al., 2010; Zhao et al., 2011; Farkade and Pathre, 2012; Canakci et al., 2013; Dai et al., 2013; Balki and Sayin, 2014; Masum et al., 2014; Elfasakhany, 2015; Li et al., 2015; Rifal and Sinaga, 2016; Elfasakhany, 2017; Li et al., 2017
	Methanol has a lower carbon content than gasoline, which makes the combustion reaction more complete.	Direct	Zhao et al., 2010; Dai et al., 2013; Balki et al., 2014; Rifal and Sinaga, 2016
	Methanol has a lower C/H ratio than gasoline	Direct	Wang et al., 2015b
	The lower stoichiometric AFR of methanol leads to the leaning effect of the methanol-blended fuel, and this promotes complete combustion.	Direct	Elfasakhany, 2015; Wang et al., 2015b; Elfasakhany, 2017
	The higher heat of vaporization of methanol-blended fuel leads to lower intake manifold temperatures, and more air access occurs during fuel combustion.	Indirect	Elfasakhany, 2015; 2017
	The lower boiling point of methanol makes the methanol-blended fuel completely vaporize, allowing for complete combustion.	Indirect	Elfasakhany, 2015; 2017
Methanol increases HC emission	The lower AFR of methanol leads to more fuel injection, which results in more fuel entering the crevice volumes or being absorbed in the oil layers.	Direct	Li et al., 2017
	The combustion process of methanol-blended fuel is advanced, decreasing the exhaust temperature, which weakens the oxidation of HC.	Indirect	Wei et al., 2008
	The higher heat of vaporization of methanol-blended fuel causes lower in-cylinder temperature, which may lead to misfire and partial combustion.	Indirect	Çelik et al., 2011; Agarwal et al., 2014

Table 6 Mechanism of methanol on NO_x emission

Impacts of methanol	Mechanism	Class	References
Methanol reduces NO_x emission	Methanol-blended fuel produces more triatomic products, which decrease the peak combustion temperature, reducing NO_x emission.	Indirect	Wei et al., 2008; Zhao et al., 2010
	The higher heat of vaporization of methanol-blended fuel leads to the lower temperatures of the combustible mixture and the lower peak combustion temperatures in the cylinder, reducing NO_x emission.	Indirect	Çelik et al., 2011; Canakci et al., 2013; Agarwal et al., 2014; Balki et al., 2014; Wang et al., 2015b; Kalwar et al., 2020; Su et al., 2020
	The lower heating value and faster flame speed of methanol-blended fuel lead to lower spark advance, decreasing combustion temperature and reducing NO_x emission.	Indirect	Pourkhesalian et al., 2010; Canakci et al., 2013; Balki and Sayin, 2014
	The lower adiabatic flame temperature of methanol would help lower NO_x emission using methanol-blended fuel.	Indirect	Wang et al., 2015b; Su et al., 2020
Methanol increases NO_x emission	The oxygen in methanol may supply additional oxygen for NO_x production.	Direct	Mallikarjun and Mamilla, 2009; Farkade and Pathre, 2012; Canakci et al., 2013; Dai et al., 2013; Masum et al., 2014
	The in-cylinder air-fuel mixture may become slightly lean when using methanol-blended fuel, resulting in more NO_x production.	Indirect	Wang et al., 2015b
	The faster flame propagation speed of methanol-blended fuel results in the increase of combustion temperature, and this may increase NO_x production.	Indirect	Wei et al., 2008; Zhao et al., 2010; Zhao et al., 2011; Dai et al., 2013; Li et al., 2015

than gasoline, the addition of methanol theoretically reduces the total CO_2 emission. Besides, it is considered in this paper that due to the high oxygen content of methanol-blended fuel, promoting complete combustion will not serve as a basic factor to increasing its emission but will rather serve as a basis for reducing actual CO_2 emission per unit work.

3.5 PM and PN

Particulate emissions from vehicles include emissions from the tailpipe (i.e., regulated emissions) and emissions due to wear of vehicle parts such as brake, tire, and clutch (i.e., non-regulated emissions) (Pant and Harrison, 2013). Here, when methanol was used as a fuel component for

Table 7 Mechanism of methanol on CO_2 emission

Impacts of methanol	Mechanism	Class	References
Methanol reduces CO_2 emission	The lower CO_2 formation per unit energy of methanol reduces CO_2 emission from methanol-blended fuel.	Direct	Vancoillie et al., 2013; Wang et al., 2015a; Turner et al., 2018
	The lower C/H ratio and carbon content of methanol reduce CO_2 emission from methanol-blended fuel.	Direct	Çelik et al., 2011; Canakci et al., 2013
	The higher BTE of methanol-blended fuel reduces CO_2 emission.	Indirect	Vancoillie et al., 2013; Turner et al., 2018
	The higher heat of vaporization and faster flame speed of methanol-blended fuel benefits the engine's thermal efficiency, which reduces CO_2 emission.	Indirect	Wang et al., 2015a
Methanol increases CO_2 emission	The oxygen in methanol enhances the combustion efficiency of methanol-blended fuel, which increases CO_2 production.	Direct	Farkade and Pathre, 2012; Balki et al., 2014; Masum et al., 2014
	The amount of methanol-blended fuel consumed per unit energy is higher, which results in a higher carbon flow rate.	Indirect	Masum et al., 2014

Table 8 Mechanism of methanol on PM emission

Impacts of methanol	Mechanism	Class	References
Methanol reduces PM emission	Methanol has no C-C bond in its structure, which significantly prevents the formation of hydrocarbon fragments during the combustion process, hence reducing PM level.	Direct	Wang et al., 2015a
	Methanol has no aromatic components that cause soot formation.	Direct	Liang et al., 2013
	The oxygen in methanol improves soot oxidation, which contributes to PM reduction.	Direct	Kalwar et al., 2020
	The faster flame speed of methanol-blended fuel leads to less time available for combustion, which reduces soot formation.	Indirect	Kalwar et al., 2020
Methanol increases PM emission	The higher heat of vaporization of methanol-blended fuel results in the decrease of the cylinder temperature, which increases HC production, leading to more particulate formation.	Indirect	Geng and Yao, 2015

Table 9 Mechanism of methanol on PN emission

Impacts of methanol	Mechanism	Class	References
Methanol reduces PN emission	Compared with gasoline, high-temperature oxidation of methanol forms fewer unsaturated micro-molecules (e.g., C_2H_2).	Direct	Geng and Yao, 2015; Su et al., 2020
	The oxygen in methanol reduces the concentration of intermediate species that are important precursors of soot.	Direct	Turner et al., 2018
	Methanol has a lower molecular weight, high oxygen content, and no aromatic content prone to soot formation.	Direct	Liang et al., 2013; Geng and Yao, 2015
	The light molecular weight of methanol benefits in allowing combustion products to remain in the gas phase instead of becoming droplets through condensation after emission, which greatly reduces PN.	Direct	Su et al., 2020
Methanol increases PN emission	Methanol-blended fuels with methanol content above a certain value lowers the exhaust temperature, which results in increase PN emission.	Indirect	Geng and Yao, 2015
	The higher heat of vaporization of methanol-blended fuel results in the decrease of the cylinder temperature, which increases HC production, leading to more particulate formation.	Indirect	Geng and Yao, 2015
	The addition of methanol increases the vapor pressure, resulting in flash-boiling of the fuel on injection and leading to inferior mixture preparation, which increases the PN emission.	Indirect	Turner et al., 2018

gasoline engines, the mechanisms that affect regulated PM emissions have been summarized.

Vehicle regulated particulate matter is microscopic solid or liquid matter produced by incomplete combustion of hydrocarbon fuel and volatilization of lubricant in the combustion process (Vouitsis et al., 2009; Muñoz-Boado and Caldona, 2017; Qian et al., 2019). Organic compounds are the most important chemical composition of vehicular PM emissions (Kleeman et al., 2000; Hao et al., 2019). Two characteristic values generally characterize particulate emissions: particle mass (PM; mg/km) emission and particle number (PN; #/km) emission.

Tables 8 and 9 summarized the mechanisms of methanol on the increase and decrease of particulate emission.

Studies on the effects of methanol on particulate emissions show that PM and PN are generally reduced. Because methanol has no C-C bond in its chemical structure and no aromatic component, soot precursors can not be formed. Methanol also can ensure complete combustion conditions. Thus, in this paper, it is considered that

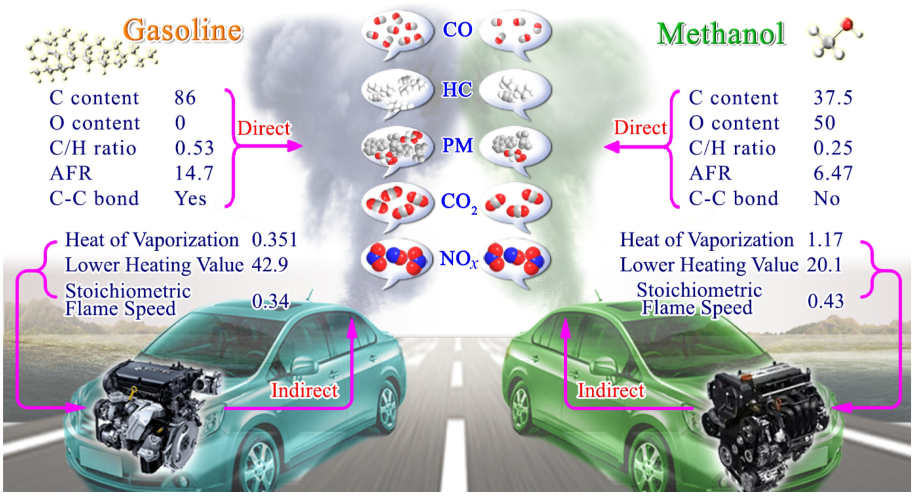

Fig. 4 Summary of the impacts of methanol fuel on vehicular emissions.

it is natural for particulate emissions to decrease when methanol is used as an alternative to gasoline.

Figure 4 summarizes the impacts of methanol blending on vehicular emissions described in Tables 4–9.

Methanol blending generally helps reduce several vehicular emissions.

4 Conclusions and future perspective

This paper discussed the research findings on the vehicular emission characteristics of methanol-gasoline blended fuels separately by EBTs and CDTs. And, on this basis, the mechanisms of methanol on regulated and CO_2 emissions were summarized. When methanol is used as a gasoline engine fuel component, the final CO and HC emissions were reported to be reduced compared to gasoline in most of the EBTs and CDTs. CO_2 emissions decreased in most of the study results of CDTs. Meanwhile, PM and PN emissions generally decreased compared to gasoline. The results of many studies on the effect of methanol blending on vehicular emissions vary considerably. When methanol is blended, various characteristics of the blended fuel (e.g., carbon and oxygen content, molar C/H ratio, stoichiometric AFR, the heat of vaporization, etc.) change significantly. According to the study conditions (engine type, engine cooling conditions, working conditions, catalytic converter, etc.), these characteristics have varying degrees of impact on emissions.

It is considered that the effects of methanol addition on regulated and CO_2 emissions can be divided into two classes.

First, the mechanisms associated with the elemental composition of the methanol molecule include lower carbon content, molar C/H ratio, stoichiometric AFR, and CO_2 formation per unit energy (specific CO_2 emissions) than gasoline; and, unlike gasoline, presence of oxygen and non-presence of C-C bond in the molecule, and no heavy HC and aromatic components, etc. These can be called direct effects mechanisms. Researchers reached the same conclusion on these mechanisms affecting regulated and CO_2 emissions. The direct effects mechanisms act to reduce the emissions of CO, HC, CO_2, PM, and PN, but to increase NO_x emission.

Second, the mechanisms that affect the working conditions of the engine by physicochemical properties of methanol: thrice heat of vaporization, half the heat value, lower boiling point, faster flame propagation speed, 49% more triatomic molecules in combustion products, etc. than gasoline. These mechanisms affect combustion temperature, in-cylinder temperature, post oxidation of regulated gas, and air-fuel mixing processes. These can be called indirect effects mechanisms. The indirect effects mechanisms usually work to reduce NO_x emissions.

Integrating the mechanism of effects on regulated and CO_2 emissions caused by adding methanol, it can be concluded that there is a sufficient theoretical and experimental basis for CO, HC, CO_2, PM, and PN emissions to decrease when methanol is used as a gasoline engine fuel component. For the NO_x emission, the increase and decrease effects vary depending on the study conditions and the methanol-blending ratio, so it can be considered that the NO_x emissions can be reduced with a reasonable methanol-blending ratio. Therefore, methanol is an excellent substitute fuel for gasoline in terms of CO, HC, CO_2, PM, and PN emissions. Automobiles are the significant source of air pollution, so use of methanol as a transportation fuel would help reduce these pollutions.

To use methanol-blended fuel generally in vehicles, it is necessary to add additives to improve some properties of methanol (e.g., corrosiveness to metals, low-temperature phase separation, etc.). It is recommended to research the effect of these additives on the emissions of methanol-blended fuel. Emission studies on methanol fuel via EBTs and CDTs (NEDC) have been conducted comparatively, but other methods have no research results. Further studies via other emission research methods (e.g., Portable Emission Measurement System, WLTC of CDTs, etc.) are required to fully characterize methanol-fueled vehicle emissions.

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Abbreviations

Abbreviations	Nomenclature
ACEA	European Automobile Manufacturers' Association
AFR	air/fuel ratio
Auto Alliance	Alliance of Automobile Manufacturers
BTE	brake thermal efficiency
CDTs	chassis dynamometer tests
CH_4	methane
CO	carbon monoxide
CO_2	carbon dioxide
C/H ratio	carbon/hydrogen ratio
DI	direct injection
EBTs	engine bench tests
EFs	emission factors
EGT	exhaust gas temperature
EMA	The Truck and Engine Manufacturers Association
EUDC	Extra-Urban Driving Cycle
FFV	flex-fuel vehicle
GDI	gasoline direct injection
HC	hydrocarbons
HCCI	homogeneous charge compression ignition
IEA	International Energy Agency
JAMA	Japan Automobile Manufacturers Association
MPFI	multipoint fuel injection
M3	consisting of 97% gasoline and 3% methanol by volume blends
M7	consisting of 93% gasoline and 7% methanol by volume blends
M10	consisting of 90% gasoline and 10% methanol by volume blends
M15	consisting of 85% gasoline and 10% methanol by volume blends
M20	consis ting of 80% gasoline and 20% methanol by volume blends
M30	consisting of 70% gasoline and 30% methanol by volume blends
M45	consisting of 55% gasoline and 45% methanol by volume blends
M50	consisting of 50% gasoline and 50% methanol by volume blends
M85	consisting of 15% gasoline and 85% methanol by volume blends
M100	pure methanol
NO	nitric oxide
NO_2	nitrogen dioxide
NO_x	nitrogen oxides
N_2O	nitrous oxide
PAHs	polycyclic aromatic hydrocarbons
PFI	port-fuel injection
PM	particle mass
PN	particle number
SI	spark ignition
TWC	three-way catalytic converters
UDC	Urban Driving Cycle

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