

Chapter 7

Study on Alternate Fuels and Their Effect on Particulate Emissions from GDI Engines



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Abstract With strict environmental legislations and to reduce related health hazards, there is immense focus on reducing particulates from gasoline direct injection engines. With increasing use of biofuels in the market, their blends with hydrocarbon fuels are also being considered as cleaner alternatives to gasoline. This chapter confers the addition of oxygenates to gasoline and their capacity to reduce sooting tendency compared to gasoline. Challenges related to optimizing combustion by appropriately choosing engine parameters such as start of ignition, duration of injection, etc. have been addressed. Optimizing combustion can reduce the particulate emissions, by sometimes increasing efficiency. Oxygenated fuels always have the advantage of higher oxidation of soot formed inside the cylinder, which further reduces particulate emissions. Towards the end of this chapter, disadvantages of using oxygenated fuel blends or alternate fuels are discussed.

Keywords Particulate matter · Legislation · Gasoline blends · Renewable fuels

7.1 Introduction

Almost a quarter of particulate emissions into the environment are caused by transportation in the world (Indicator Assessment 2015). Conventionally, world's demand for energy is mostly met from combusting fossil fuels. Fossil fuel combustion generates global warming pollutants such as CO, CO₂ and NO_x along with soot and hydrocarbons including polycyclic aromatic hydrocarbons (PAHs) that affect human health (Liu et al. 2015). Particulate matter, formed due to incomplete combustion, is a carbonaceous material. Upon inhalation, being carcinogenic, particulate matter may lead to pulmonary and respiratory diseases (Claxton 2015). Soot is also one on the contributors to global warming due to its role in regional warming and the faster melting of polar icecaps (Maione et al. 2016). It is essential to regulate soot

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formation in engines to abide by emission regulations and also towards controlling health effects (Überall et al. 2015).

Compared to diesel engines, gasoline engines emit higher number of particulates, sometimes even more than the prescribed limit (Myung et al. 2012). Moreover, this problem of soot has intensified with gasoline direct injection (GDI) engines taking over the market with their benefits compared to port fuel injection (PFI) engines (Zimmerman et al. 2016). GDI engines prevailed in the automotive industry as it offers relatively higher BSFC, volumetric efficiency and for their superior knock resistance compared to PFI. GDI engines also plays a major role in reducing CO₂ emissions from gasoline-powered engines. However, the particulate mass and the number of large particulates emitted by GDI is higher than PFI (Saliba et al. 2017). Thus, during gasoline combustion there is still a greater need to address the higher particulate emissions by reducing soot formation. Detailed discussion on health effects related to soot emissions from GDI can be found in Sharma and Agarwal (2018).

Some of the existing techniques to reduce particulate emissions include fuel reformulation, addition of metal catalysts to fuels, and methods for after-treatment of exhaust. For instance, one of the widely implemented after-treatment system, gasoline particulate filters (GPF) are used to filter out soot from entering into atmosphere from engine exhaust (Chan et al. 2014). Especially in Europe due to the upcoming stringent regulations on the number of particulates that can be emitted from gasoline engines (Johnson and Joshi 2017). GDI engines with GPF produce more ultrafine particulates compared to diesel engines fitted with diesel particulate filter (DPF) (He et al. 2012). These ultrafine particulates with sizes less than 100 nm, are found to cause adverse impacts on human health and environment (Bernstein 2004). This chapter focusses on particle number (PN) rather than particulate mass (PM) emitted from an engine as health impact of PN emissions can be strongly correlated to PN. Especially, particles as fine as ultrafine particles contribute little to the PM while being a significant part of the total PN. Finer the particulates get, deeper becomes their penetration into the lungs and therefore increased are the chances of passage into the human bloodstream (Eastwood 2008).

Moreover, these metal catalysts that are added to fuels which reduce soot, end up being more harmful to the environment and human health (Casseo et al. 2011). There are promising results from fuel blends that could lower soot emissions without the need for modifying the engine technologies (Guerrero Peña et al. 2018).

Fuel reformulation facilitates increase in the amount of oxidizer inside the combustion chamber by blending oxygenated and conventional fuels that leads to reduction in soot formation. Quite a lot of oxygenated compounds such as methanol (Wang et al. 2015), ethanol (Masum et al. 2013), butanol (Gu et al. 2012), furans (Tanaka et al. 2015), dimethyl carbonate (Schifter et al. 2016), ethers (Cataluña et al. 2008) and ketones (Elfasakhany 2016) have been found to decrease soot emission when blended with gasoline (Guerrero Peña et al. 2018). The reduction in sooting tendency of such blended fuels comes from the fact that the oxygen atoms present in these fuels present favorable chemical or dilution effects (Lemaire et al. 2010, 2015), or makes soot particles more susceptible towards oxidation (Gogoi et al. 2015). Such high soot oxidation can also help with the rapid regeneration of GPFs at low temperatures.

This chapter describes the characteristics of PN emitted from GDI engines from the view point of impact of fuel composition, in terms of oxygenates blended with gasoline making it an alternative to the existing engine technology. Many studies have tested individually on the application and possible benefits of such alternate fuels. This study provides a comprehensive report on recent progress in reducing PN from GDI research using alternate fuels including gasoline blended with renewable fuels.

7.2 PN from GDI Engines and Alternate Fuels

Particulates are usually formed from GDI engines during fuel combustion in the combustion chamber. The ones that exit the engine can also originate from nucleation of supersaturated vapors in the exhaust gas after-treatment systems (Heywood 1988). Formation of fuel films on the combustion chamber walls contribute significantly to the PN formation since fuel deposited this way form fuel-rich areas. From these locally rich areas, soot can easily be formed and subsequently emitted. Many studies recently looked at improving mixture quality that promote soot reduction. For available engine technology fuel reformulation and alternate fuels provide an easily adaptable alternative.

A variety of fuel properties affect PN formation in an engine including aromatic content and oxygenate content, enthalpy of vaporization and boiling points of individual components. Soot formation is highly affected by the existence of aromatic fuel rings as combustion researchers believe that gaseous polycyclic aromatic hydrocarbons (PAHs) act as precursors for soot. As expected, it was found in research that higher amount of aromatics in the fuel would lead to a higher level of PN (Raza et al. 2018).

The impact of fuel volatility on PN emissions comes from the fact that slower evaporation of a fuel due to its lower vapor pressure produce a relatively poorer mixture formation leading to higher PN. Pool fires that occur due to higher impingement on walls and piston that cause significantly higher PN making their occurrence a case of extremity. However, higher volatility in a fuel can also cause higher PN with flash evaporation leading to a mixture with locally rich zones causing incomplete combustion.

Of all the available fuel blends, E5 (5% of ethanol and 95% of gasoline by volume) is now common in Europe (European Committee for Standardization 2008); blends of E10 are ubiquitous, and E15 is on its way to the market for newer vehicles in the USA (U.S.C. §7546). Introducing ethanol into gasoline fuel blends tends to increase the enthalpy of vaporization for the fuel. Higher enthalpies of vaporization in a way, increase the charge cooling effect for the fuel and improve volumetric efficiency. However, charge cooling and lower boiling point of ethanol that promote less production of soot can sometimes play the opposite role and have competing effect which could lead to the formation of soot (Leach et al. 2018).

In general, addition of ethanol helps with soot reduction. However, some studies have found that under certain conditions, ethanol blended with gasoline can increase particulate emissions, while other studies have shown a reduction in PN emissions with increasing levels of ethanol. Thus, there is still a need for clear distinction of points where presence of oxygen in fuel can help reduce soot formation. After several attempts made to link the fuel composition and particulate emissions, the Honda Particle Matter Index (PMI) has shown a very good correlation with emissions for gasoline engines (Wittmann and Menger 2017). Researchers are still searching for such an index to provide a robust prediction to PN emission from a gasoline fuel blend. However, PMI is still used for comparative purposes (Bock et al. 2019).

7.3 Impact on PN

Alternate fuels are oxygenated fuel blends, having a great potential to reduce well-to-wheel CO₂ emissions from vehicles. Ethanol is the most commonly added oxygenate component to gasoline; other oxygenates include alcohols such as methanol, butanol and ethers such as methyl tert-butyl ether (MTBE), ethyl tert-butyl ether (ETBE) formed out of etherification of respective alcohols. E5 with 5% (v/v) ethanol blended in gasoline is everywhere in Europe, with E10 and E15 prevailing in the USA; E85 and E100 are also available, although requires vehicles to acquire special adaptation.

Compared to pure gasoline, oxygenated fuels are found to have higher vapour pressures, significantly higher heat of vaporization, and significantly lower LHVs. These characteristics most likely affect the amounts of fuel injected into the cylinder. Thus, also affecting the spray evaporation once after the fuel has been injected. These affected phenomena in turn result in higher or lower PN depending on engine operating conditions. Due to the presence of the –OH bond in almost all oxygenates, it increases the reactivity towards oxidation of soot precursors, which reduce PN emissions.

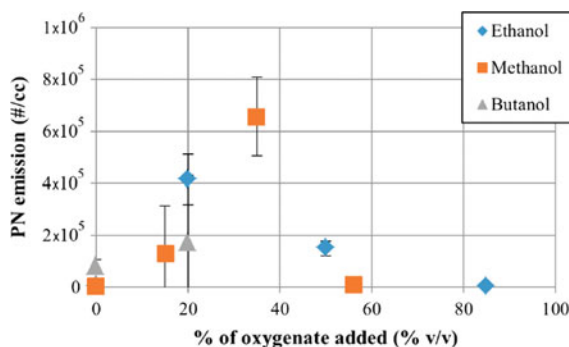
What makes alcohols such great oxygenates that are commonly available these days? Some factors include:

- (a) they can be produced from renewable feedstocks (Bae and Kim 2017),
- (b) have low water solubility, posing less impact to our water supply,
- (c) can be blended with gasoline and burned directly as a neat fuel, and
- (d) reduces carbon monoxide emissions forcing leaner combustion than pure gasoline (Fig. 7.1).

7.3.1 *Effect of Ethanol on PN*

Since, ethanol is produced from renewable sources, most of the times recycled, ethanol utilization doesn't contribute to possible global warming and pollution

Fig. 7.1 PN emissions from a single cylinder optical engine for fuel blends with pure components having varying levels of oxygenates (Leach et al. 2013)



(Wyman and Hinman 1990). Hence, making ethanol a sustainable drop-in fuel for spark ignition engines.

Using ethanol to reduce PN has already been established in PFI engines reduces PN. However, studies show mixed behavior in GDI engines, i.e., adding ethanol can both increase and decrease. As in the case of GDI, quality of combustion and engine operating conditions are deemed important for PN. Liquid fuel impingement on cylinder walls, valves and piston is one of the major causes of particulate formation, and it is rather prevalent at cold start and transients. With ethanol blends, spray formation is relatively challenging which may lead to higher PN compared to gasoline. However, in case where impingement can be compensated by time and energy for relatively better mixture formation, PN reduces.

Once, the level of blended ethanol increases there is a decrease in PN emissions reported. The presence and dominance of the chemically bonded oxygen and much diluted aromatics, there is a reduction in formation of PAHs, soot precursors which in turn reduces soot. However, recently Burke et al. (2017) found that high levels of ethanol can stratify the aromatics within the evaporated mixture and lead to higher PN although certain amount of dilution of aromatics in the fuel with blended ethanol.

Some investigations with ethanol blended with gasoline include Storey et al. (2010) who used E10 and E20 and measured reduced PN from GDI engine over FTP75 and US06 cycle. Zhang et al. (2014) studied the impact E10 and E20 on PN and found that the PN before the catalyst (TWCs) decreased from a GDI engine, while some others reported contrary results. In general, ethanol reduces PN emissions at low and part load conditions. However, in some cases, liquid fuel impingement arises, then ethanol generates more PN emissions as compared to gasoline. This increase in PN may be attributed to the higher heat of vaporization of ethanol.

7.3.2 Effect of Methanol on PN

Sometimes, methanol can be used instead of ethanol as a blend-in fuel with its lower costs of production compared to ethanol. Using methanol is not as common as

ethanol, however in China one can find M100 (100% methanol) and is widely used in their transportation. Like ethanol, methanol has a lower volumetric energy density and higher RON compared to gasoline.

Following its predecessor, methanol, like ethanol, when blended with gasoline has similar effect on PN. One of such effect from methanol is that it promotes fuel evaporation by reducing the final boiling point of the fuel that helps in reducing PN. Also, there is increased heat of vaporization and high volatility which may cause poorer mixtures leading to higher PN. In a study, Qin et al (2014) investigated the impact of gasoline, 100% methanol, and other combinations of methanol-gasoline blends and found that with increase of methanol concentration in gasoline, PN emissions decreased significantly. As expected, neat methanol produced even fewer PN compared to gasoline. Also supported by another study that reported compared to gasoline, methanol blends (M15, M25, and M40) reduced PN (Turner et al. 2013). However, studies like Mohd Murad et al. (2016) reported that M15 does not consistently lower PN from gasoline engines. There is a recent interest in mixed blends of gasoline, ethanol and methanol.

7.3.3 *Effect of Butanol on PN*

Compared to ethanol and methanol, butanol is a higher alcohol containing four-carbon (C4) molecules (Jin et al. 2011). Butanol has higher energy density among the mentioned alcohols, smaller latent heat of vaporization and is comparatively less corrosive. Both *n*-butanol and isobutanol isomers are commonly used as blend-ins for gasoline. Butanol, having higher energy density among other benefits compared to other alcohol blend-ins is now being considered as the next major oxygenate to blend-in with gasoline. Having higher energy density shows promise towards minimal increase in fuel consumption as opposed to its competitors. *N*-butanol is also produced from a fermentation process. From the recent research on the effect of butanol on PN, the trend reported is similar to ethanol which showed a modest increase in PN with butanol levels (Jin et al. 2011; Tao et al. 2014).

7.4 Conclusions

Particulate matter emissions from GDI engines comprise of complex mix of volatile and solid components containing soot, organic carbon and hydrocarbons. Nucleation mode (<50 nm) particles have often been considered as volatiles, but recently studies have reported to find solid particles in nucleation mode as well. Accumulation mode (50–200 nm) particles include black carbon, carbonaceous soot particles with a rudimentary carbon structure and adsorbed volatiles.

In general, homogeneous operation in GDI engines have fully vaporized fuel-air mixture and give low levels of PN compared to other charge compositions. Any

diffusion flames caused by liquid fuel remains at ignition, particularly on combustion chamber surfaces, leads to PN, also known as pool fire. Many injection strategies have been developed and investigated addressing such liquid fuel impingement. This phenomenon is also prevalent at cold starts with cold conditions inside the cylinder.

Type of fuel is very essential to engine-out PN emissions. In general, effect of fuel is often masked by other dominating engine operating parameters. It has been made possible to compare the effect of fuel while running at stoichiometric operation. Aromatics, the most commonly PN attributed fuel property typically promote formations of PAHs, soot precursors which in turn increases PN. Some other fuel properties do influence PN but are dependent on engine design and load point.

Adding oxygenated to a fuel dilutes the aromatic content of the fuel, theoretically leading to lower PN compared to the fuel in its pure form. Since aromatics is not the only influencing property of a fuel towards PN, there is a mixed trend that is reported for oxygenated blends. Although, lower levels of oxygenates in the blends show promise for reducing PN at lower loads, they fail to reduce PN at higher loads. Coming to higher levels of oxygenates, overall the PN emitted from the GDI engines have reduced almost to 0 (i.e., not detectable by measurement instrument) in some cases.

PN from GDI engines still stands as one of the complex physiochemical phenomena with many parameters and variables affecting the engine out performance. However, with increased levels of understanding now available of PN formation in GDI engines from research it is possible to take measures to reduce PN and its formation. Certainly, this chapter has only focused on engine-out PN emissions, many aftertreatment devices, such as gasoline particulate filters are available, and are only delivering better performance with time.

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