Combustion, gaseous and particulate emission of a diesel engine fueled with n-pentanol (C5 alcohol) blended with waste cooking oil biodiesel

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Abstract

The combustion, gaseous and particulate emissions of a diesel engine fueled with biodiesel-pentanol (BP) blends were investigated under different engine loads. The results indicate that with the increased pentanol fraction, the start of combustion is delayed. All of the BP blends provide faster combustion than biodiesel and diesel fuel from CA10 to CA90. The faster combustion of BP blends leads to a higher BTE than that of biodiesel and diesel fuel in most cases. The particle mass and number concentrations are reduced by the addition of pentanol in biodiesel in most test conditions, due to the higher oxygen concentration for the fuel/air stoichiometry, longer ignition delay for fuel/air mixing, and lower viscosity for the improvement of atomization. The R-(C=O)O-R' group in biodiesel is less efficient in suppressing the soot precursor's formation than the R-OH group in pentanol. The diameter of the primary particles is reduced with the increased addition of pentanol. The particulate emission of BP10 have higher oxidation reactivity that that of BP20 and BP30. Base on this study, pentanol-biodiesel can be considered as an acceptable alternative fuel for diesel engines due to its improved combustion performance and reduced particulate emissions.

Keywords: Pentanol; Biodiesel; Particulate size distribution; Morphological analysis; Oxidative reactivity;

1. Introduction

With growing concerns about energy security and future oil supplies, the search for alternative fuels such as biodiesel, natural gas, ethanol, dimethyl ether and hydrogen for use in internal combustion engines has become a hot research topic. Due to the similar fuel properties with diesel fuel, biodiesel is considered as a potential alternative fuel for diesel engines. When biodiesel is used in a diesel engine, particulate mass concentration, hydrocarbon and carbon monoxide emissions decreased, while particle number concentration and NOx emission increased inevitably [1-3].

Alcohols, served as a fuel additive, can reduce the viscosity and surface tension, improve atomization and provide additional oxygen content to blended fuel, and thus have the potential to simultaneously reduce the emissions of both NO_x and particulate matter (PM). C1-C3 alcohols (methanol, ethanol, and propanol) have been studied in blending with diesel fuel [2, 4, 5]. However, there are lots of disadvantages for the use of lower-alcohol diesel blends, such as, lower heat value, poor miscibility, and poor stability during blending. Due to its higher carbon chain, higher cetane number, and better miscibility with diesel than that of C1-C3 alcohols, researchers have been investigating the use of butanol (C4 alcohol) in diesel blends. The addition of n-butanol to diesel fuel can significantly reduce CO and soot emissions without seriously affecting the brake specific fuel consumption and NO_x emission [6]. However, Yilmaz et al. [7] found that the CO and HC emissions were higher for butanol-biodiesel blends.

Increasing the carbon chain length of alcohol generally improves the ignition qualities of the alcohol, which makes higher alcohols (>C4) more attractive as biofuels than lower alcohols (C1-C4). N-Pentanol has 5 carbon straight-chains in the molecule. It has an even higher energy density and cetane number than butanol and the other lower alcohols. Large-scale production of pentanol can be achieved with biological pathways [8, 9]. For gasoline engine, it has been confirmed that pentanol is not an ideal alternative fuel for displacing gasoline in SI engine because of its low octane number or high water solubility[10].

Given its proper fuel properties, pentanol could be considered as a good alternative fuel for diesel engines and it provides several advantages over the C1-C4 alcohols, such as higher energy density, higher cetane number, higher heating value, better miscibility, and blend stability with diesel fuel. Moreover, for diesel engine or other modern engine concepts (HCCI), low-temperature reactivity is needed for auto- ignition. Due to the higher energy density and similar alkane-like negative temperature coefficient (NTC) behavior [11], pentanol may be served as a proper fuel for the blending use in diesel engine or HCCI. After adding pentanol to diesel fuel, the heat release could be smoothed, while the NOx and soot emissions were lower than that of diesel fuel [12]. A 25% pentanol/75% diesel fuel blend was recommended for application [13]. According to the fundamental combustion research on iso-pentanol in HCCI engines, pentanol requires a lower intake temperature than gasoline and ethanol, indicating that pentanol has a higher HCCI reactivity [14]. Wei et al. [15] found that n-pentanol addition in diesel fuel could significantly reduce the PM emissions and slightly increase the NOx emissions. However, few studies have examined the use of pentanol as a fuel additive in biodiesel in compression ignition engines or, in particular, focused on the effect of the higher alcohol (C5) on particulate emissions.

Therefore, a better understanding of the combustion, gaseous and particulate emissions of the higher alcohols (C5) is needed to evaluate the potential use of pentanol. The aim of this study is to investigate the effects of the combustion, gaseous and particulate emissions of a diesel engine fueled with biodiesel-pentanol (BP) blends under different engine loads, with particular focus on the particulate mass and number concentrations, particle size distribution, primary particle size and oxidation characteristics of the PM.

2. Experimental Setup and Measurements

A naturally aspirated, water-cooled, four-cylinder, direct-injection diesel engine was used in this study. The experimental system is shown in Figure 1. The engine specifications are similar to those in Zhu et al. [2]. The test conditions of engine are selected at 1800 r/min and 28, 84, 140, 196 and 224 Nm of engine loads, corresponding to brake mean effective pressures of 0.08, 0.24, 0.41, 0.57, and 0.65 MPa, respectively. Euro V diesel fuel (DE), biodiesel (B100), and biodiesel-pentantol (BP) blends were used. The blended fuels contained 10%, 20%, and 30% by volume of n-pentanol, and were identified as BP10, BP20, and BP30. The waste cooking oil methyl ester was supplied by Dynamic Progress Ltd. The major properties of the fuels and the instruments used in this study are given in Table 1 and Table 2, respectively. The experimental methods and procedures are similar with previous studies [2, 16]. The standard errors were 1.7% and 2.5% for the particle mass concentration and particle total number concentration, respectively. Student's T-test was used to analyze whether the differences between the results obtained from different fuels were statistically significant at the 95% confidence level.

3. Results and discussion

3.1 Combustion, thermal efficiency and fuel consumption

The combustion process, as reflected by the in-cylinder pressure and the heat release rate are compared with different fuels in Figures 2 and 3 for the engine loads of 0.24 MPa and 0.65 MPa, respectively. All the results of combustion are averaged over 400 cycles. The in-cylinder pressure and the heat release rate of the biodiesel-pentanol (BP) blends increase with the increase of pentanol fraction and occur further away from the top dead center (TDC), compared to biodiesel, indicating that there is a longer ignition delay, which can be attributed to the lower cetane number of pentanol. Moreover, due to the lower viscosity of pentanol, the addition of pentanol improved the atomization performance of biodiesel and enhanced the fuel/air mixing, resulting in increases in the premixed heat release rate and the maximum pressure.

Figure 4 shows the effects of pentanol on burning rate in cylinder. It can be observed that, CA10 is delayed for the BP blends, which is consistent with the longer ignition delay period due to the lower cetane number. While from CA10 to CA90, all of the BP blends have faster combustion than biodiesel and diesel fuel. Due to the alcohol combustion chemistry, at low temperature, pentanol reacts with O_2 at the alpha position leading to the formation of the aldehyde and HO_2 radical, and then resulting in an overall lower reactivity of pentanol. While at high temperature, the OH group in pentanol could weaken the C-C bonds between the alpha and beta position, leading to an easier breaking of the C-C bonds, resulting in a faster decomposition and oxidation of pentanol at high temperature [17]. The higher reactivity of pentanol at high temperature can also be explained by the formation of major intermediate species, such as ethylene, propene, 2-butene, and iso-pentanal.

The faster combustion process also leads to the higher brake thermal efficiency (BTE) of the BP blends, as can be seen in Figure 5. According to the heat release rate analysis, the heat release of the BP blends is more centered, resulting in higher positive work and hence higher BTE than that of biodiesel and the diesel fuel [2]. Moreover, the increase in BTE could also be due to the improvement in the combustion process on account of the increased oxygen content in the BP blends. The BTE increases with the increase of the engine load. However, the BTEs of BP20 and BP30 decreased at 0.65 MPa, which indicates that the combustion is deteriorated at this engine load, which could contribute to a longer combustion period, as seen in Figure 4. The studies of lower alcohol (C1-C4) addition had been already conducted [2, 18-21], in which the efficiency was improved by alcohol addition, especially for 10% blend ratio. However, large percentage of alcohol adding in diesel fuel or biodiesel, especially for lower alcohol, leads to vapor lock for fuel system, resulting in unstable of cycle fuel injection quantity and combustion deteriorate. The variation of brake specific fuel consumption (BSFC) is also given in Figure 6. The BSFC of pentanol-biodiesel blends are higher than that of biodiesel and diesel fuel, which is more obvious at low and high engine load.

3.2 Gaseous emissions

As seen in Figure 7, HC and CO emissions of BP blends are higher than that of diesel and biodiesel, especially at low engine load from 0.08MPa to 0.24MPa. It is expected that n-pentanol/biodiesel blends have lower cetane number compared with biodiesel, which results in more time for the fuel to vaporize, leading to a broader lean outer flame zone

[15], which results in higher HC emissions with the addition of n-pentanol. For NOx emission, BP10 and BP20 give lower NOx emissions than that of biodiesel. While the NOx emission of BP30 is higher than other fuels. Lower cetane number, higher oxygen content and higher latent heat of vaporization are these key factors to improve or suppress the formation of NOx. These factors interact each other leads to the results of NOx emission of BP blends.

3.3 Particulate mass and number concentration

Figure 8 shows the variation of particulate mass concentrations of different fuels. In general, the particulate mass concentration increases with the engine load. The particulate mass concentrations are reduced for the BP blends at medium and high engine loads. The possible reason is that the BP blends have higher oxygen content but lower carbon content than those of biodiesel, which improves the combustion process. According to the above discussion, the fuel-air mixing process is more adequate for the BP blends, which could suppress the formation of primary soot particles in the fuel rich zone. Moreover, the higher concentrations of the small radicals (H, OH, O) produced by pentanol promotes the oxidation reaction from carbon atom to CO and CO_2, which could reduce the chances of soot precursor formation from carbon atom.

However, at low engine loads of 0.08 MPa and 0.24 MPa, the BP blends have higher particulate mass concentrations than that of biodiesel. The effect of the carbon chain length was often more important than the direct chemical effect of the oxygen function [22], indicating that although pentanol has 18% oxygen content, the high fraction of carbon-carbon bonds has a higher contribution to the PM mass formation tendency at the low engine loads of 0.08 MPa and 0.24 MPa.

The effects of the BP blends on the particle size distributions are shown in Figures 9 and 10 for two of the five engine loads, 0.24MPa and 0.65MPa, respectively, while the variations in the total particle number concentration are shown in Figure 11. For each test fuel, the distribution curve shifts upwards and toward a larger size as the engine load increases, leading to increases in both the total number concentration and geometric mean diameter (GMD) of the particles. After adding pentanol to the biodiesel, the particle size distribution curves become lower and flatter, resulting in a lower total number concentration.

The decrease of particle number concentration after adding pentanol to biodiesel can be explained from several perspectives. Firstly, because of the lower cetane number and lower viscosity of pentanol, there is a longer period for the fuel/air mixing, leading to a more thorough mixture. Secondly, pentanol with a single imbedded oxygen atom is more effective in the reduction of soot precursors than the two oxygen atoms in biodiesel as R-(C=O)O-R'. Because the $CH_3O(CO)$ radical is decomposed primarily to CH_3+CO2, rather than CO+CH_3O, leading to the bonding of the two oxygen atoms to a single carbon atom, and then these two oxygen atoms are less efficient in removing carbon atoms from the reactive medium [23]. Thirdly, the addition of pentanol can increase the concentrations of OH by the reaction of methyl radical with HO_2 radical ($CH_3+HO_2=CH_3O+OH$), which could promote the oxidation of soot precursors.

Diesel particles can be classified as nucleation mode (D_{pm}<50nm) and accumulation mode (D_{pm}>50nm) particles. Nano-particles with smaller diameter are more dangerous because these particles can be easily breathed into the lung, and even transferred to the blood [24]. Due to the harmful effects of smaller nano-particles, the variation in the number concentration of the nucleation mode particles (D_{pm}<50nm) is also shown in Figure 12. The number concentration of nucleation mode biodiesel particles is higher than that of diesel fuel. BP blends could reduce the number concentration of nucleation mode particles, especially at high engine loads.

3.4 Size of primary particle

Typical TEM images for the biodiesel-pentanol blends at the engine load of 0.57 MPa are shown in Figure 13. The primary particle diameter is analyzed using statistics based on the TEM images with the Image Pro software, and the mean diameters (Dp_{mean}) are calculated. The typical primary particle diameter distributions for BP10, BP20, and BP30 are shown in Figure 14, which varies in the narrow range from10 to 60 nm. Based on the figure, it can be concluded that the diameter of primary particle can be reduced by increasing the amount of pentanol added. The Dp_{mean} is 27.50 nm, 25.75 nm, and 21.23 nm for BP10, BP20, and BP30, respectively.

There are two stages for the formation of particles, the in-cylinder stage and the exhaust dilution stage, which involved many processes such as soot precursor formation, nucleation, growth, oxidation, and aggregation. Those processes are strongly affected by the combustion condition inside the engine cylinder. The oxygen atoms in pentanol molecules can improve combustion of the fuel/air mixture in the local fuel rich zone, suppress the formation of soot nuclei, and accelerate the surface oxidation of the primary particles. Moreover, the higher in-cylinder temperature of BP blends improves the oxidation of primary particles, due to higher premixed combustion. On the other hand, the diesel soot oxidation in the cylinder is governed mainly by the content of aliphatic C–H, the oxygenated groups [25], and the higher initial active sites (ASA) [26] in the particle surface functional groups. In the case of pentanol addition, the PM samples have a higher intensity of aliphatic groups and higher OH oxygenated groups [27], which could improve the oxidation of the primary particles in-cylinder and reduce their size.

3.5 Particle oxidation characteristics

Thermogravimetric analysis (TGA) is used to investigate the oxidative reactivity of the particulate matter samples of different fuels at the engine load of 0.57 MPa. Meanwhile, each initial sample mass is kept around 2mg to ensure the reliability of TGA results. Figure 15 shows the mass loss curves with the temperature increase. As seen in the figure, all mass loss curves are composed of two periods: from 200 to 300^{o}C and from 400 to 700^{o}C. During the formation of particulate matter emission, some small particles are formed by the nucleation of semi-volatile substances, combining with condensation or absorption of semi-volatile substance on the surface or between carbon layers of existing particles. Thus, the first stage of mass loss curve is mainly caused by the remove of the low volatile and semi-volatile substances, while the second stage is caused by the oxidation of non-volatile substances or dry soot.

According to the result in the first stage, biodiesel have higher semi-volatile fraction than that of diesel fuel. After addition of pentanol, it can be seen that the semi-volatile fraction condensed on particulates of BP blends decrease with pentanol blend ratio. The semi-volatile fraction of particulate samples increase in this order: DE100<BP30<BP20<BP10<biodiesel. When the temperature rises to 400°C, the oxidation reactivity of non-volatile substance varies for each fuel. According to the figure, the ignition temperature of particulate samples increase in this order: biodiesel<BP10<BP20<BP30<DE100. The lower ignition temperature of particulate means that this particulate is much easier to be oxidized. With increase of pentanol in BP blends, the curve of mass loss moves towards to that of diesel fuel, indicating that the particulate of BP blends could be oxidized with higher temperature than that of

biodiesel. The possible reason is that the higher concentration of semi-volatile condensed or absorbed on the surface or between carbon layers of particles contributes to disorder and loose nano-structure of particles, which could increase the specific surface area for reacting with oxygen, leading to the higher oxidative reactivity.

4. *Conclusion*

In this study, the combustion, gaseous and particulate emissions of a diesel engine fueled with BP blends are examined, with particular focus on their effects on particulate mass and number concentrations, particle size distribution, and morphological analysis of the PM. The conclusions can be summarized as follows:

1). With increase of pentanol fraction, the start of combustion and the crank angle for maximum heat release move further away from TDC. While the in-cylinder pressure and heat release rate of BP blends increase continuously with pentanol.

2). All of the BP blends have faster combustion than biodiesel and diesel fuel in most test modes. The differences of pentanol effect in CA90 and CA50 are attributed to the different reactivity of pentanol at high and low temperature. The faster combustion process in the premixed combustion stage can increase the BTE of the BP blends, especially for BP10. The BSFC of pentanol-biodiesel blends are higher than that of biodiesel and diesel fuel.

3). HC and CO emissions of BP blends are higher than that of diesel and biodiesel, especially at low engine load from 0.08MPa to 0.24MPa. BP10 and BP20 give lower NOx emissions than that of biodiesel. While the NOx emission of BP30 is higher than other fuels.

3). The particle mass and number concentrations can be reduced by the addition of pentanol in biodiesel, especially particles in the nucleation mode. The R-(C=O)O-R' group in biodiesel is less efficient in suppressing the soot precursor's formation than the R-OH group in pentanol.

4). The diameter of the primary particle is reduced as the amount of pentanol added increases, due to the higher in-cylinder temperature and higher intensity of aliphatic groups, leading to the oxidation of the primary soot particles.

5). The semi-volatile fraction of particulate samples increase in this order: DE100<BP30<BP20<BP10<biodiesel, leading to the higher particles oxidative reactivity of BP10 than that of BP20 and BP30.

Base on this study, BP blend is considered as an acceptable alternative fuel for diesel engines due to its improved combustion performance and reduced particulate emissions. The addition of 10% pentanol to biodiesel is recommended as a suitable replacement ratio for diesel engines.

Acknowledgements

The authors would like to thank the National Science Foundation of China (Project No. 51306117; 51436005), the Chinese Ministry of Science and Technology (MOST) (Project No. 2012BAD47B04), the Foundation of Shanghai Jiao Tong University and the Research Grants Council of the Hong Kong SAR (Project No. G-YBF7) for supporting this research.

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and nanostructure of diesel particulate matter. Fuel 2015; 161: 18-25.

Figure captions

Fig.1 Experimental setup

Fig.2 In-cylinder pressure and heat release rate at 0.24MPa

Fig.3 In-cylinder pressure and heat release rate at 0.65MPa

Fig.4 CA10, CA50 and CA90

Fig.5 Brake thermal efficiency

Fig.6 Brake specific fuel consumption

Fig.7 Gaseous emissions

Fig.8 Particulate mass concentration

Fig.9 Particle size distribution at 0.24MPa

Fig.10 Particle size distribution at 0.65MPa

Fig.11 Total number concentration

Fig.12 Particle number concentration of $PM_{<50nm}$

Fig.13 TEM images of BP blends

Fig.14 Primary particle diameter distribution of BP blends

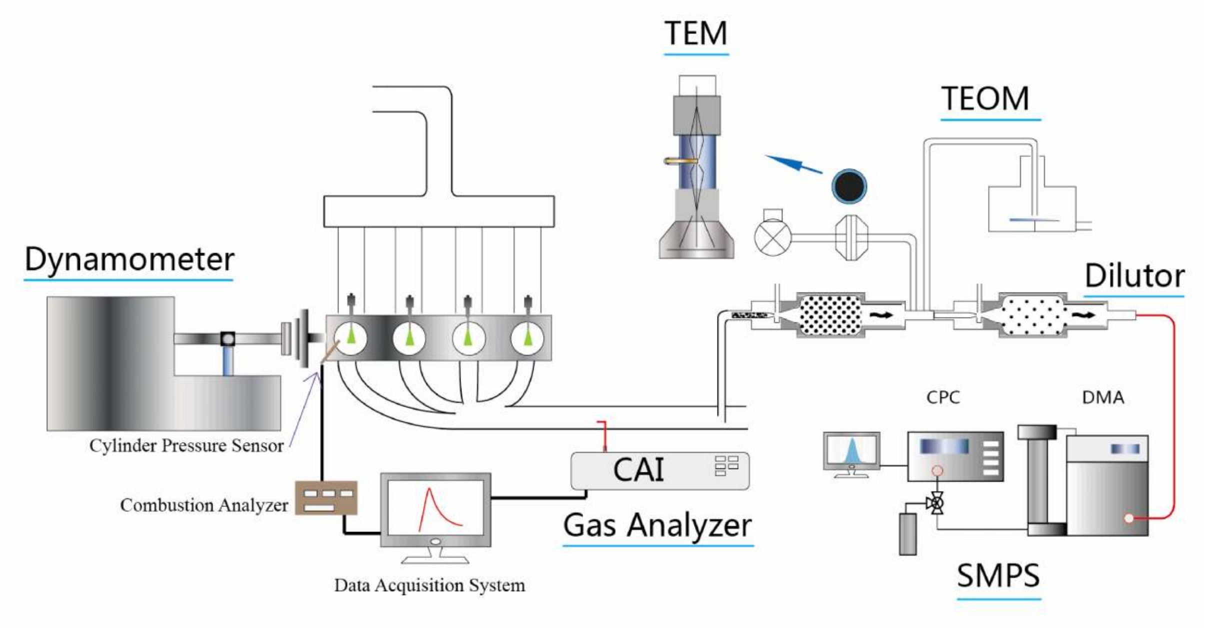
Fig.15 Mass loss curves of different particle samples

Table 1 Properties of diesel, biodiesel and n-pentanol

Fuel properties	Diesel fuel	Biodiesel	n-Pentanol
Cetane number	52	51	20
Lower heating value(MJ/kg)	42.5	37.5	34.65
Density(kg/m3) at 20℃	827	871	814
Viscosity (mPa s) at 40℃	2.86	4.6	2.88
Heat of evaporation(kj/kg)	250-290	300	308
Oxygen content(%wt)	0	10.8	18.1
Sulfur content(mg/kg)	<10	<10	0

Table 2 Experimental instruments

Test Item	Instrument
Cylinder pressure	Kistler piezoelectric sensor (Type 6056A)
Heat release rate	Combustion analyzer (DEWE-ORION-0816-100x)
Particle mass concentration	Tapered element oscillating microbalance (1105)
Particle size distribution	Scanning mobility particle sizer (TSI Model 3071A).
CO2 concentration	Non-dispersive infrared analyzer (CAI 300)
Image of primary particle	Transmission electron microscope (Tecnai G2 20 S-TWIN)
Image processing software	Image-Pro Plus 6.0 (Media Cybernetics)
Thermogravimetric analysis	Netzch STA 449 TGA/DSC with Al_2O_3 crucible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TEOM
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DMA
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Combustion Analyzer
Gas Analyzer
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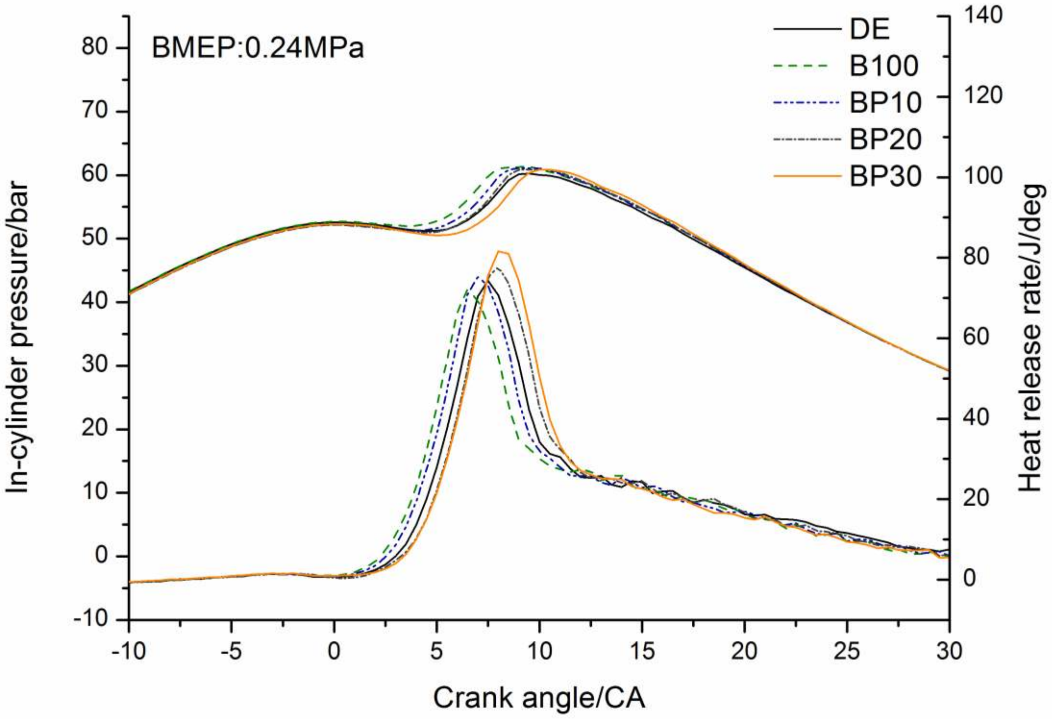
BMEP:0.24MPa
DE
B100
BP10
BP20
BP30
In-cylinder pressure/bar
Heat release rate/J/deg
Crank angle/CA
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0
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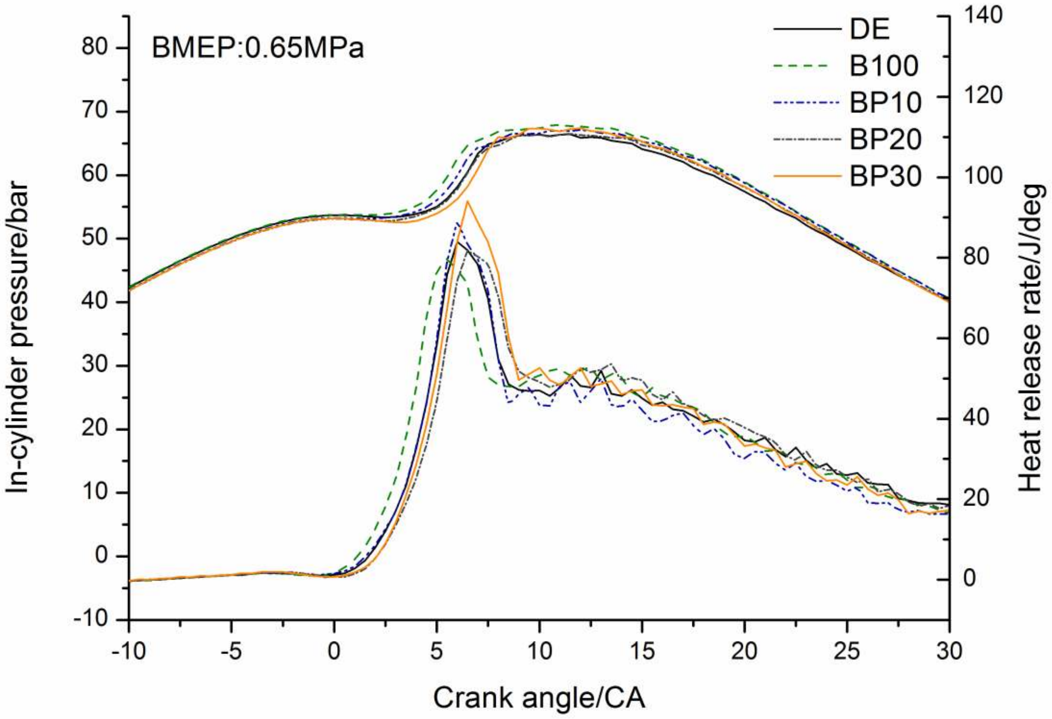
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B100
BP10
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In-cylinder pressure/bar
Heat release rate/J/deg
Crank angle/CA
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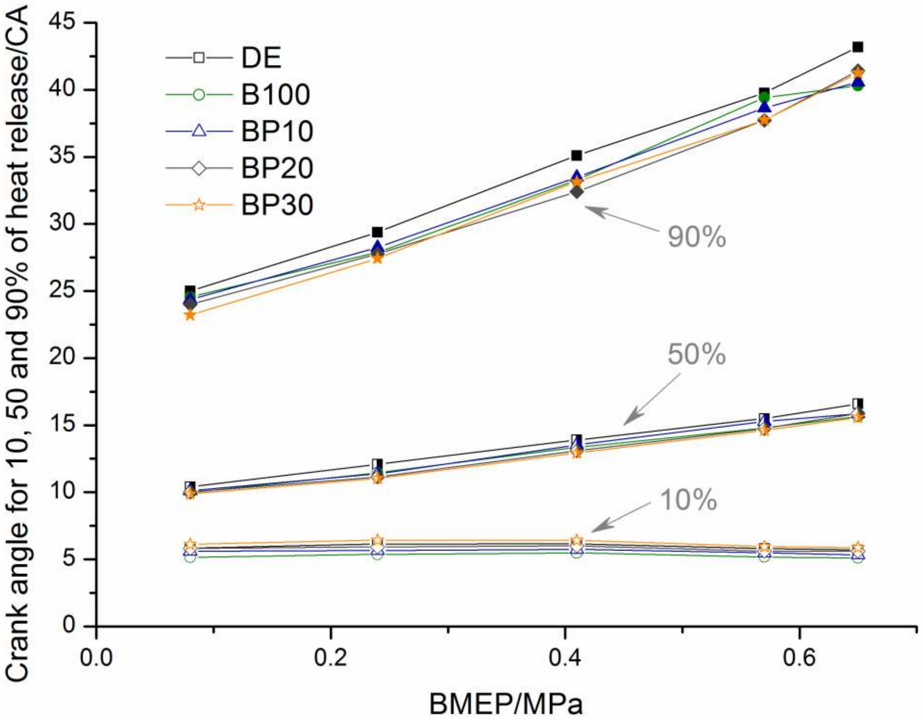
Crank angle for 10, 50 and 90% of heat release/CA
BMEP/MPa
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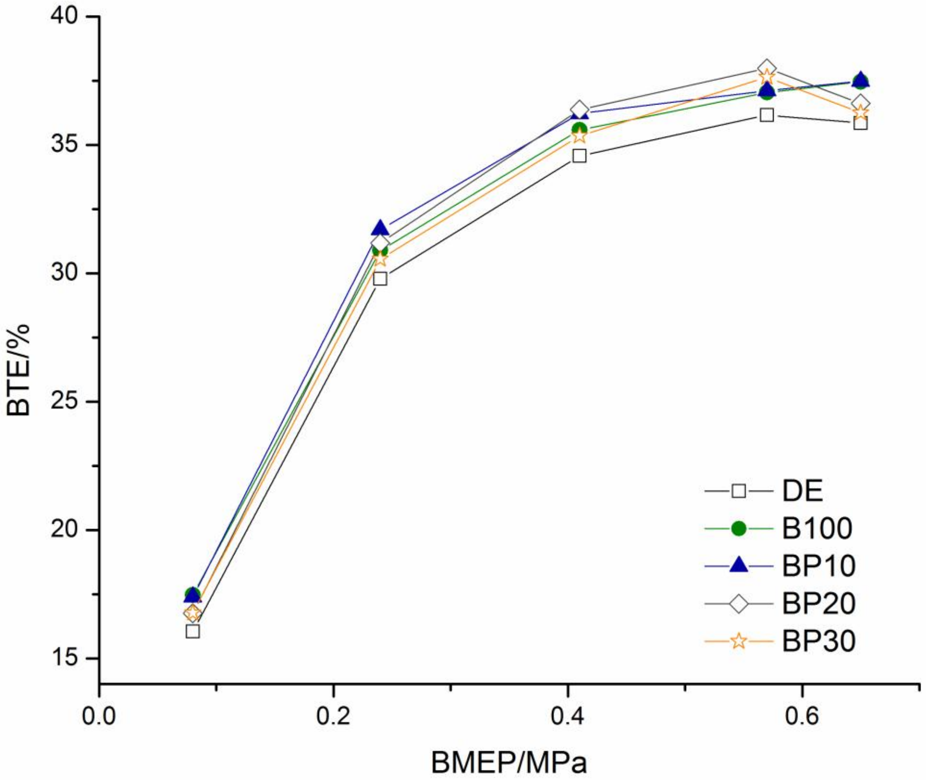
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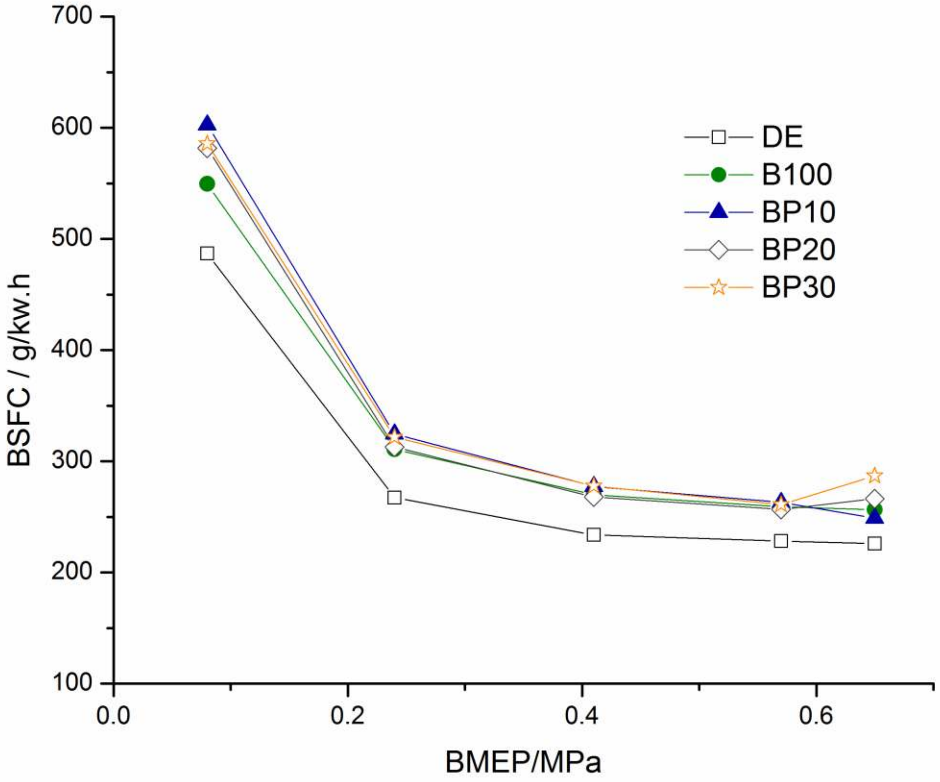

BSFC / g/kw.h
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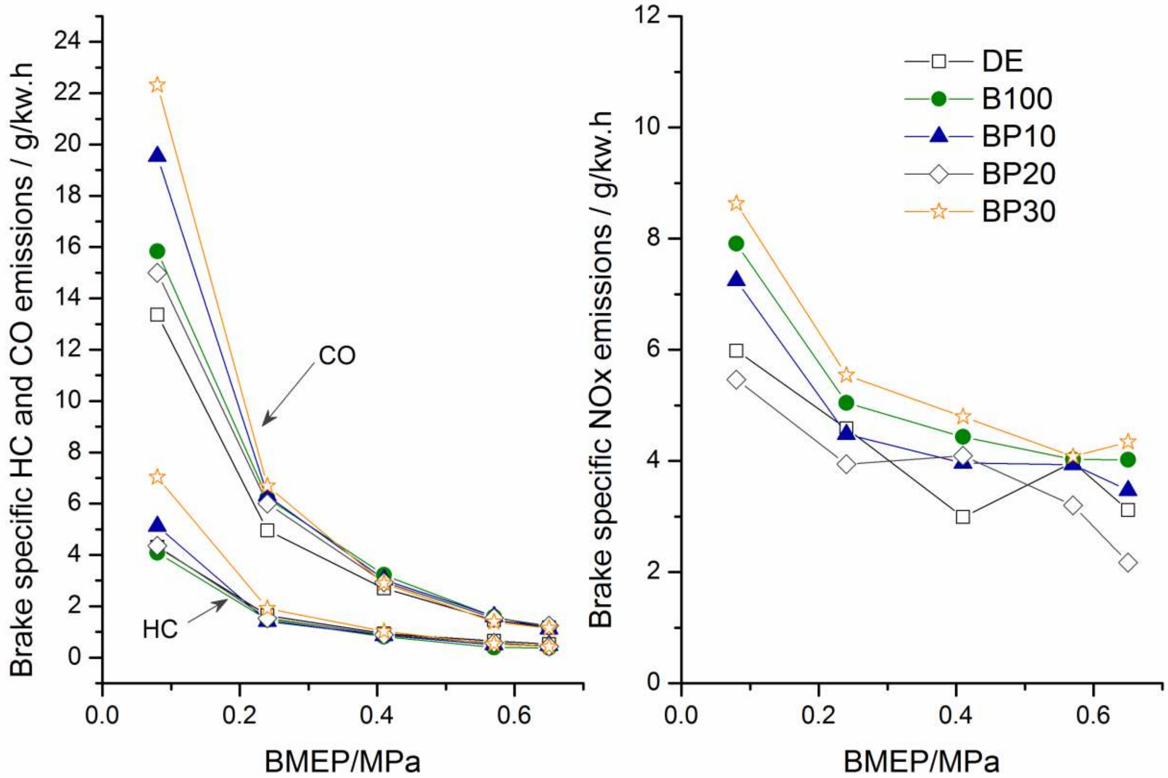
Brake specific HC and CO emissions / g/kw.h
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CO
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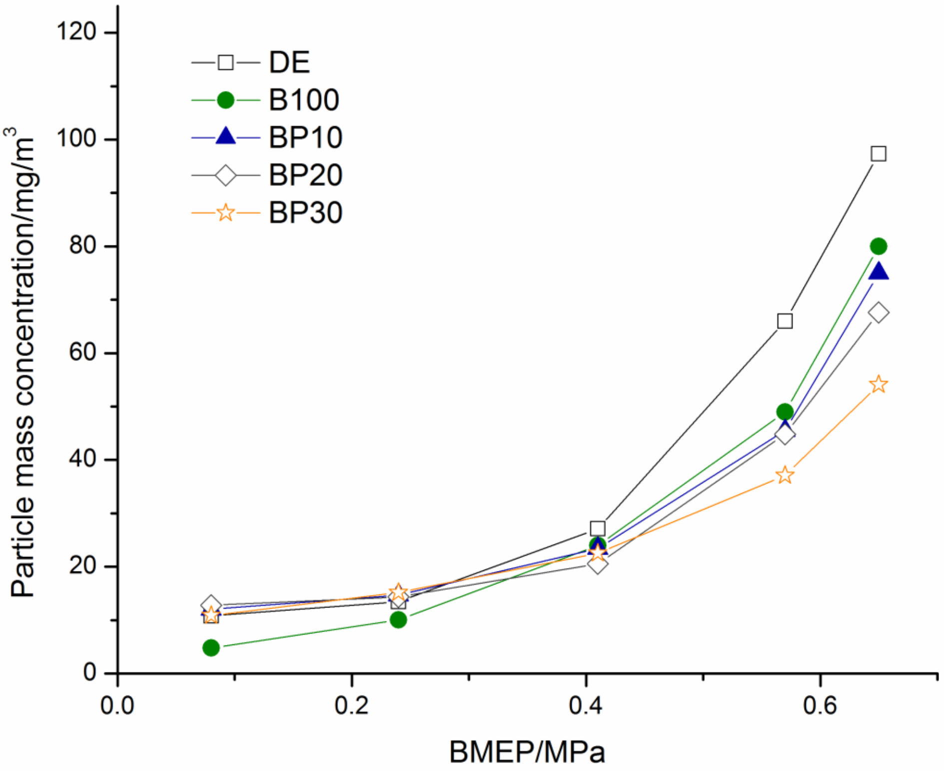
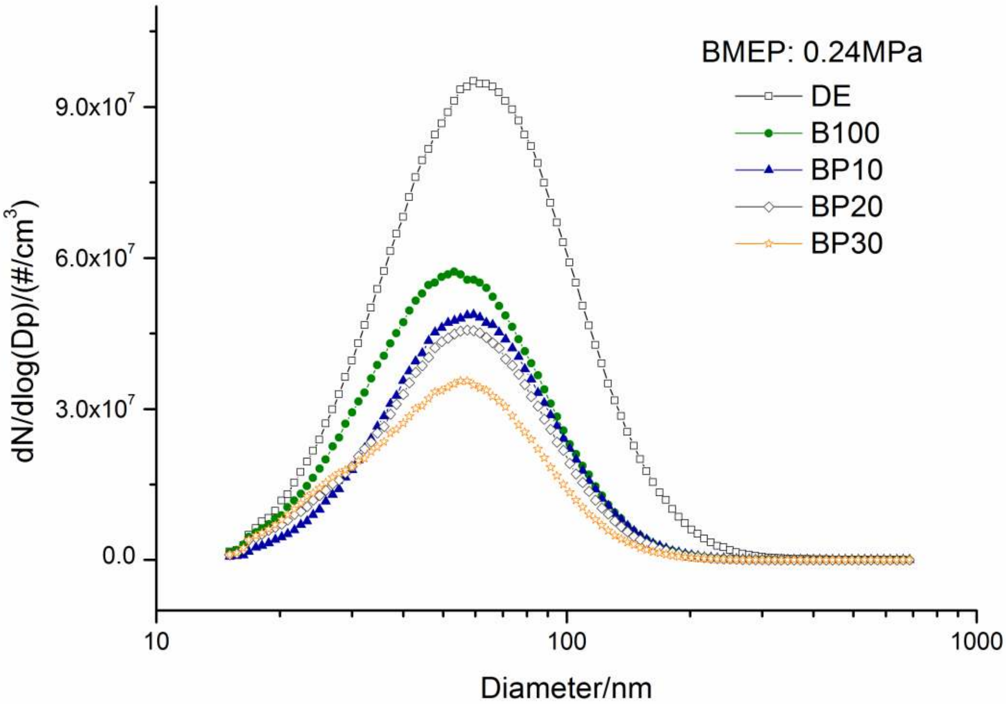
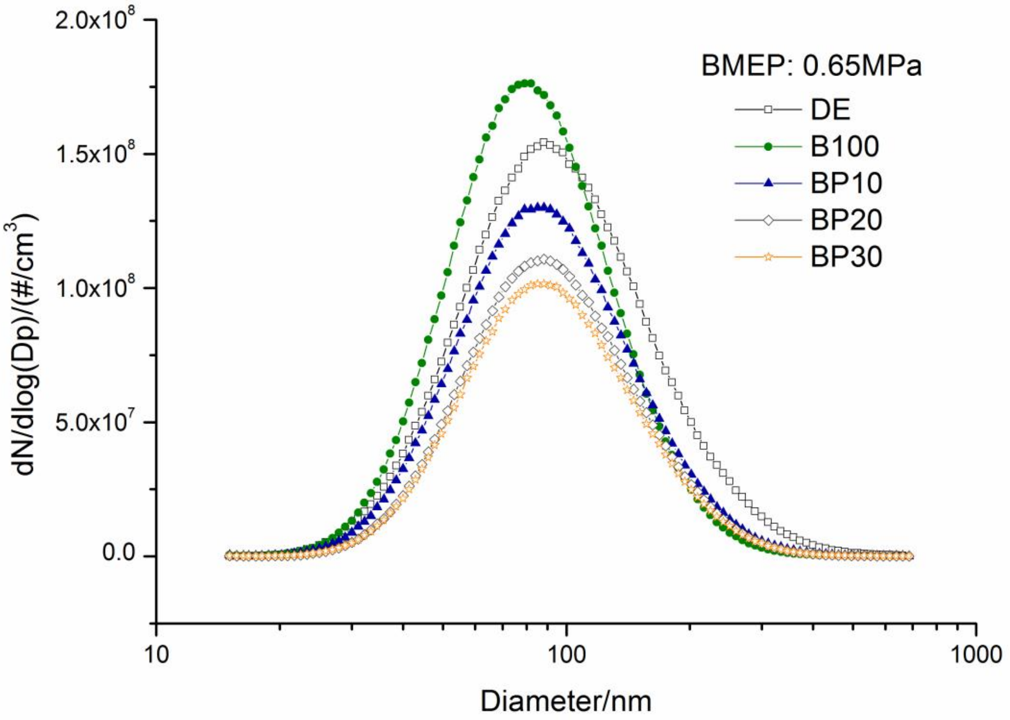
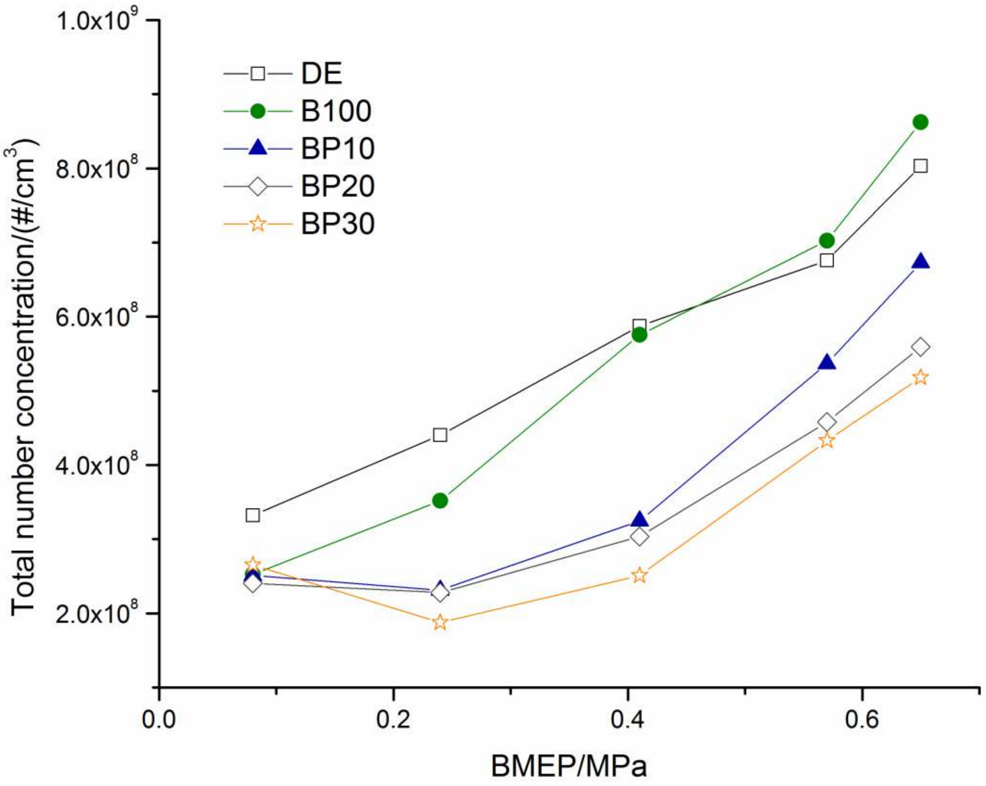
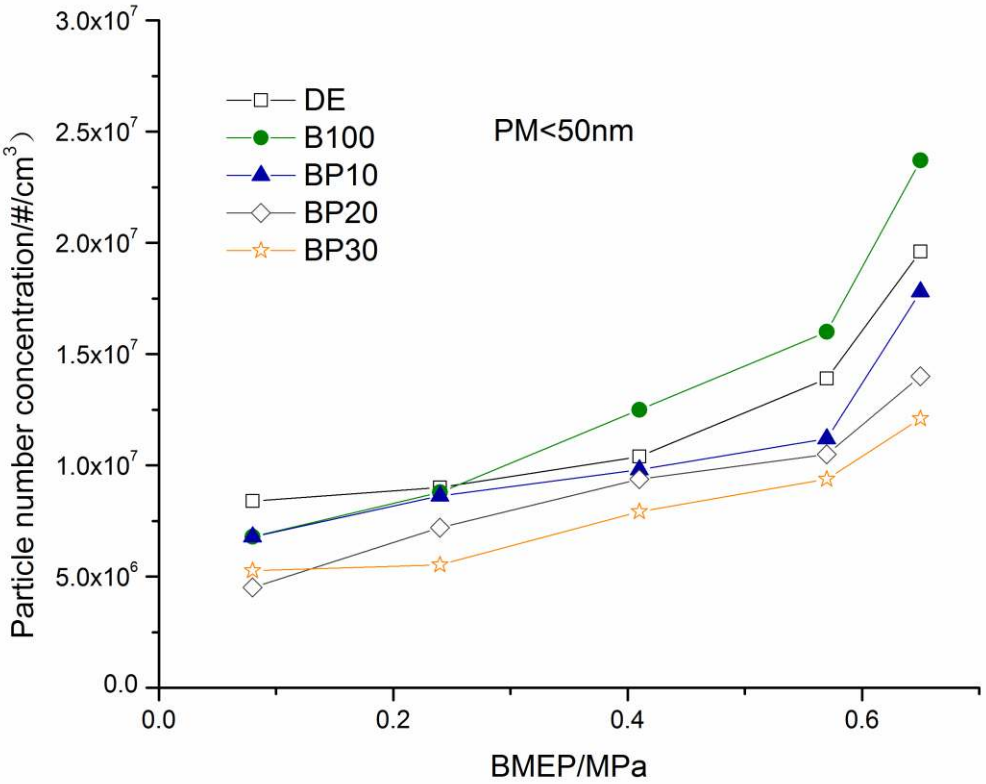
0.6
BMEP/MPa
Brake specific NOx emissions / g/kw.h
0
2
4
6
8
10
12
DE
B100
BP10
BP20
BP30
0.0
0.2
0.4
0.6
BMEP/MPa

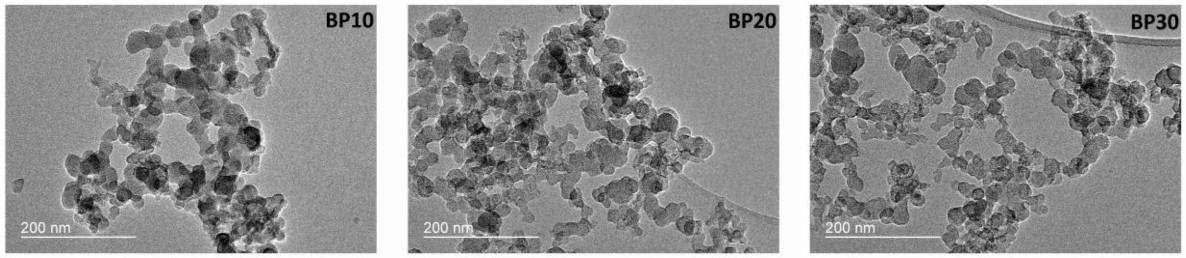

Particle mass concentration/mg/m3
BMEP/MPa
DE
B100
BP10
BP20
BP30
0
20
40
60
80
100
120
0.0
0.2
0.4
0.6

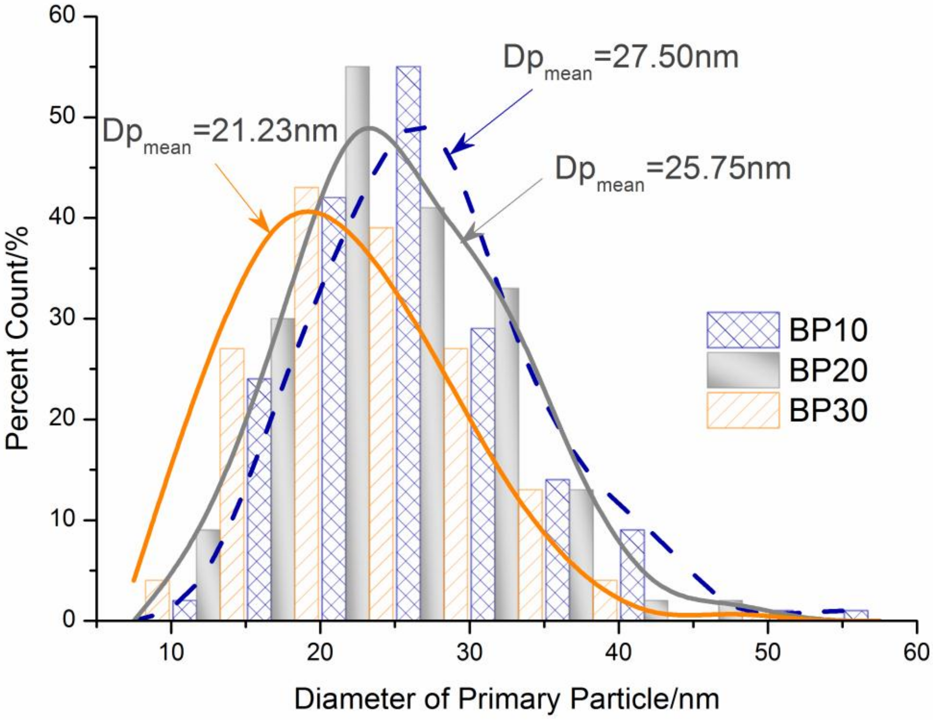

BMEP: 0.24MPa
DE
B100
BP10
BP20
BP30
dN/dlog(Dp)/(#/cm³)
9.0x10⁷
6.0x10⁷
3.0x10⁷
0.0
10
100
1000
Diameter/nm

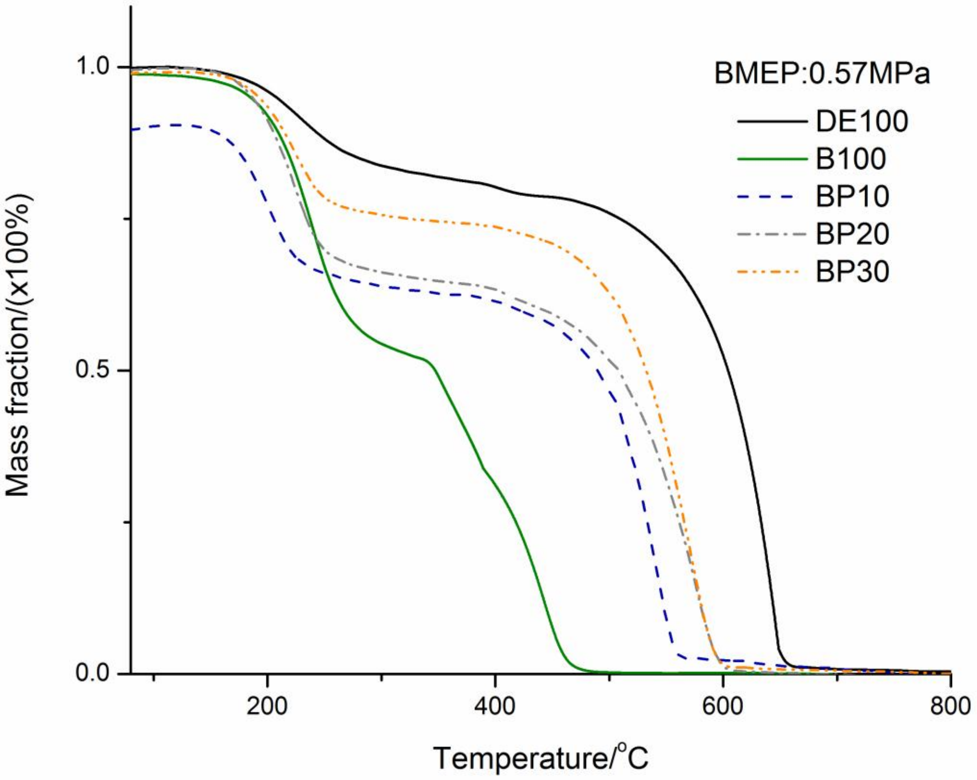

BMEP: 0.65MPa
DE
B100
BP10
BP20
BP30
dN/dlog(Dp)/(#/cm³)
2.0x10⁸
1.5x10⁸
1.0x10⁸
5.0x10⁷
0.0
10
100
1000
Diameter/nm


1.0x10^9
8.0x10^8
6.0x10^8
4.0x10^8
2.0x10^8
Total number concentration/(#/cm^3)
0.0
0.2
0.4
0.6
BMEP/MPa
DE
B100
BP10
BP20
BP30


PM<50nm
DE
B100
BP10
BP20
BP30
Particle number concentration/(#/cm^3)
0.0
5.0x10^6
1.0x10^7
1.5x10^7
2.0x10^7
2.5x10^7
3.0x10^7
0.0
0.2
0.4
0.6
BMEP/MPa


BP10
200 nm
BP20
200 nm
BP30
200 nm


Percent Count/%
0
10
20
30
40
50
60
Diameter of Primary Particle/nm
10
20
30
40
50
60
Dp_{mean}=27.50nm
Dp_{mean}=21.23nm
Dp_{mean}=25.75nm
BP10
BP20
BP30


BMEP:0.57MPa
DE100
B100
BP10
BP20
BP30
Mass fraction/(x100%)
1.0
0.5
0.0
200
400
600
800
Temperature/^{o}C