PARTICULATE MATTER EMISSION FROM A HEAVY DUTY DIESEL ENGINE WITH THREE BINARY BLENDS

by

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Low temperature combustion using gasoline-like fuels has the potential to provide high efficiencies and extremely low NO_x and soot emissions. In this study, different volume percentages (30%~70%) of iso-octane, toluene, and n-butanol are blended with n-heptane separately. These blends with different composition ratios are tested on a modified single-cylinder research engine. Also, simulations are performed using a homogeneous reactor method to know the fuel-chemical effects on particulate matter emissions. Thirdly, a composition ratio of 70% is selected to perform further experiments based on the results from the initial composition ratio experiments with a focus on the particle size distributions. It was found that if the test fuel can provide sufficient ignition delay to allow fuel to premix with air fully, the soot emissions will be low and particle size is small even if the test fuel contains a lot of aromatic compounds.

Key words: *Diesel engine, particle size distribution, iso-octane,
              toluene, n-butanol*

Introduction

The past two decades have witnessed a drastic reduction in regulated limits for pollutant emissions from Diesel engines. Therefore, Diesel engine manufacturers are seeking to optimize engine-operating strategies to minimize pollutants without sacrificing fuel economy. Low temperature combustion (LTC), based on the compression ignition of a premixed or partially premixed dilute charge, can provide equal or higher efficiencies than that of a Diesel engine. At the meantime, LTC can produce NO_x and particulate matter (PM) emissions well below mandated standards, eliminating the need for expensive after-treatment systems. The formation kinetics of soot are slowed by reducing combustion temperatures. Therefore, excessive exhaust gas recirculation (EGR) can lead to very low soot formation. Unfortunately, combustion efficiency is poor and excessive unburned hydrocarbons and CO emissions are produced when high EGR rates are used well before soot formation is substantially inhibited [1]. The fuel used is also expected to have a significant role in engine optimization, so rather than using extremely high EGR rates most LTC strategies address PM emissions through using high-octane or low-cetane fuels to increase the proportion of premixed combustion [2, 3]. Since soot forms in the fuel-rich regions, increasing mixing prior to combustion can reduce or eliminate the fuel-rich mixtures thus reducing PM emissions.

Aromatic hydrocarbons are important as additive or components of transportation fuels due to their resistance to knocking. However, aromatics are much more prone to form

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soot and are recognized as the main precursors in soot formation [4]. Toluene is a good surrogate of aromatics contained in diesel fuels, thus its chemistry has to be studied in combustion conditions. Most of the experimental work on toluene chemistry are performed in a jet stirred reactor [5]. Furthermore, the influence of applying n-heptane/toluene blend in a Diesel engine has been tested [6, 7]. All these tests use less than 30 vol.% toluene in the blend. Fundamental studies of soot formation in laminar diffusion flames have shown that a very wide range of hydrocarbon compounds can form soot [8]. However, the sooting tendency changes widely, for example, aromatic compounds form soot readily, while alcohols are resistant to soot formation [9]. It is well-known that fuel composition affects Diesel engine-out PM, oxygenated fuels have shown to decrease PM (*e. g.* [10]) and reducing aromatic-content fuels in various levels also leads to less engine-out PM (*e. g.* [11]). Will toluene lead to more soot emissions and more big size particles compared to alkanes and alcohols when more than 30 vol.% toluene is used? Because of the complex compositions of conventional diesel, n-heptane was usually used as a surrogate for the hydrocarbon fuels employed in Diesel engines. In this study, n-heptane (straight paraffin) was chosen as a base fuel, various volume percentage (30, 40, 50, 60, and 70) of iso-octane (branched alkanes), toluene (aromatics), and n-butanol (alcohol) were added to n-heptane separately. Firstly, three binary blends with different composition ratios were tested and their effects on ignition delay and soot emissions were measured. Secondly, simulations with a homogeneous reactor method were done to get knowledge about the fuel-chemical effects on particulate matter emissions. Thirdly, a composition ratio of 70% was chosen to test with a focus on particle size distribution.

Materials and test methods

In this section, firstly the test engine is introduced. Secondly, an emission testing equipment (engine exhaust particle sizer) and its dilution systems are talked about. Thirdly, the properties of test fuels are given. Fourth, the data acquisition process is discussed and the related parameters used in this paper are defined. Lastly, the experimental conditions and simulation method are introduced, respectively.
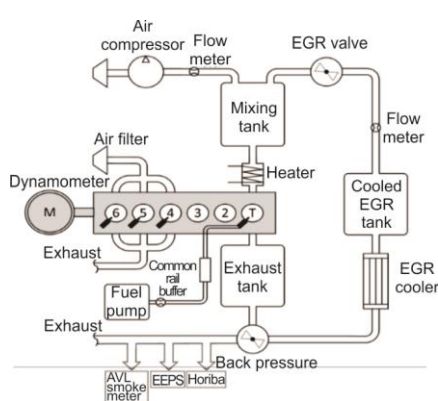

Figure 1. Schematic diagram of experimental set-up

Test engine set-up

The experimental engine is based on a DAF XE355c heavy-duty engine modified to operate with one test cylinder and three propelling cylinders. Figure 1 shows the test bench and tab. 1 gives the engine specifications. The test cylinder was isolated and has separate intake and exhaust systems, an independent EGR system, and fuel injection system. The second and third cylinders are not fired during measurements, while cylinder four to six are controlled by the stock engine control unit and are responsible for maintaining the rotational engine speed. Because brake torque measurements are not reliable for a single cylinder conversion from a six-cylinder engine, as used for this study, all loads and thermodynamic efficiencies presented in this article are indicated values. An Atlas Copco air compressor is used to boost intake charge and an electric heater is installed to heat up the intake charge. The injector used was a production one with the nozzle specification shown in tab. 2.

In this study, the EGR ratio is experimentally determined from the ratio of the CO_2 volume concentration in the intake gas to the CO_2 volume concentration in the exhaust gas, as expressed by eq. (1):

$$EGR = \frac{CO_{2_{inlet}}}{CO_{2_{outlet}}} \qquad (1)$$

Soot emissions are measured with an AVL 415S smoke meter, with a measurement range from 0 to 10 filter smoke number. The HC, CO, exhaust CO_2, NO_x, and intake CO_2 are measured using a Horiba Mexa 7100 DEGR exhaust analysis system. Each analyzer is calibrated with an appropriate calibration gas before every set of measurements.

Table 1. Geometrical properties of the DAF XE355c

Stroke [mm]	Bore [mm]	Connecting rod [mm]	Compression ratio [–]
158	130	266.7	15.8
Intake valve close [°CA]	Intake valve open [°CA]	Exhaust valve open [°CA]	Exhaust valve close [°CA]
−153	344	128	−346

Table 2. Injector specifications

Included spray angle [°]	Orifices [–]	Orifices diameter [mm]
139	7	0.18

Engine exhaust particle sizer and dilution systems

The engine exhaust particle sizer (TSI EEPS 3090) is used in the experiments to obtain detailed information about particle concentration and size distribution. The EEPS is a fast-response, high-resolution instrument that measures very low particle number concentrations in the diluted exhaust. It offers the fastest time resolution available, 10 times per second, which makes it well suited by engine tests. It measures the size distribution and number concentration of engine exhaust particle emissions in the range from 5.6 to 560 nm.

The thermo-dilution unit contains a rotating disk thermo-diluter (model 379020A) and a thermal conditioner air supply (model 379030). This thermo-dilution separates sampling, dilution, and conditioning of the aerosol into following steps:
− undiluted exhaust emissions are sampled at a rate of approximately 1.0 L per minute, and a portion of the raw exhaust is captured by each cavity of the rotating disk and transported to the measurement channel where it is mixed with particle-free dilution air; the thermo-diluter also features variable dilution ratio over the range from 15:1 to 3000:1 and selectable heated dilution temperature up to 150 °C,
− the thermal conditioner model 379030 generates the primary dilution air for the diluter with a calibrated and controlled flow of 1.5 L per minute; it uses a heated evaporation tube, which can be heated up to 400 °C to eliminate nano-droplets that may have formed during the dilution process; since the sampled aerosol is below the dew point after primary dilution, thus no re-condensation takes place in the cooling down zone, and
− the primary diluted measuring gas (1.5 L per minute) from the model 379020A is diluted with secondary dilution air (flow is adjustable from 0 to 15 L per minute) generated in the model 379030; therefore, a maximum flow of 16.5 L per minute can be achieved; the EEPS spectrometer operates at 10 L per minute; fig. 2 shows the sample air path (diluted) for the combined 379020A-30 system.

In this study, real-time sampling rate and a sampling period of EEPS were set at 1 Hz and 1 minute, respectively. During all the experiments primary dilution (with rotating disk diluter) temperature was set at 150 °C with a dilution factor of 50 and the secondary dilution temperature was set at 350 °C (evaporation tube) with a dilution factor of 6.7, thus the overall dilution ratio is 335. In this study, the EEPS spectrometer data was processed with the soot

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Figure 2. Sample air path (diluted) for the combined 379020A-30 system

matrix which matches more closely to the data from a TSI scanning mobility particle sizer (SMPS) spectrometer. Software version 3.2.5 and firmware version 3.1.2 were used when using EEPS.

Fuel specifications

Primary reference fuel (PRF) is a commonly used surrogate for gasoline and is a mixture of iso-octane and n-heptane. It is easy to blend a PRF which has a specific research octane number (RON) or motor octane number (MON). However, PRF have the same RON and MON values and therefore cannot provide a realistic representation of the octane sensitivity exhibited by the practical fuels [12]. When the end-gas temperature rises above the standard MON test condition, the octane sensitivity is a measure of the deterioration in the anti-knock property of a fuel [13]. The addition of high-octane compounds such as toluene and n-butanol to n-heptane provides the opportunity to formulate blends with non-zero sensitivities. Firstly, different volume percentages (30%~70%) of iso-octane, toluene, and n-butanol are blended with n-heptane, respectively. The number following PRF represents the volume percent of iso-octane. The toluene-containing blend is called TRF and the number following TRF represents the volume percent of toluene. The n-butanol-containing blend referred to as BRF and the number following BRF represents the volume percent of n-butanol. The properties of the fuel blends are estimated by a calculation based on the mole fraction of each constituent in the mixture, as listed in tab. 3. A lubricant was added to each test fuel (1 mL lubricant to 1 L fuel) to reduce friction and prevent wear of the injector.

Table 3. The properties of the fuels for the first set of experiments

Fuel name	LHV [MJkg^{-1}]	Density [kgm^{-3}]	AF stoich [−]	H/C ratio [−]	O/C ratio [−]
PRF30	44.52	683	15.07	2.27	0
PRF40	44.51	684.1	15.07	2.27	0
PRF50	44.50	685.2	15.06	2.27	0
PRF60	44.48	686.3	15.06	2.26	0
PRF70	44.47	687.4	15.05	2.26	0
TRF30	43.08	749.7	14.5	1.86	0
TRF40	42.65	769.8	14.32	1.74	0
TRF50	42.25	788.65	14.15	1.62	0
TRF60	41.88	806.3	13.99	1.51	0
TRF70	41.53	822.9	13.84	1.41	0
BRF30	39.87	733	13.75	2.35	0.071
BRF40	38.61	747.2	13.33	2.37	0.095
BRF50	37.47	760.1	12.93	2.39	0.120
BRF60	36.44	771.8	12.55	2.41	0.145
BRF70	35.49	782.51	12.17	2.43	0.170

Data acquisition and definition of related parameters

The pressure transducer signals were digitized and recorded at 0.1 CAD increments for fifty consecutive cycles using an SMETEC Combi crank angle (CA) resolved system. An in-house data acquisition system is used to measure slow changing parameters, such as intake air-flow and intake temperature. Slow changing parameters are measured during a period of 40 seconds at a frequency of 20 Hz.

The CA of 50% burn point (CA50) was used to monitor the combustion phasing. CA50 was determined from the cumulative apparent heat-release rate, computed from the

cylinder pressure data. The average of 50 consecutive individual-cycle CA50 values was used in this paper. The start of combustion (CA10) was defined as the CA where 10% of the heat was released. Ignition delay (ID) in this paper was defined as the CA difference between the start of injection (SOI) and CA10: ID = CA10 − SOI (CAD). The gross indicated mean effective pressure (IMEPg) which excludes the gas exchange stroke is used to present engine load and calculate the indicated emissions.

Engine operation

Prior to starting the experiments, the engine was fully preheated until the lubrication oil and coolant fluid temperature reach 85 ℃. An auxiliary heater mounted close to the engine provided precise control of the intake temperature. All data were taken at an engine speed of 1200 rpm. In the first set of experiments, PRF, TRF and BRF were tested at three engine loads, which are low load (5 bar IMEPg), medium load (10 bar IMEPg), and high load (15 bar IMEPg), respectively. The experimental conditions with fuels of different blending ratios are summarized in tab. 4. The dependency of soot levels on fuel composition can be caused by fuel chemical property effects, such as different molecular structure, or by the mixing degree of fuel/air mixture caused by different ID. Secondly, PRF70, TRF70, and BRF70 were tested with a focus on particle size distribution. The test conditions are listed in tab. 5.

Table 4. Experiments with fuels of various mixing ratios

Parameters	Load		
	Low	Medium	High
IMEPg [bar]	5	10	15
Fuel pressure [bar]	800	1200	1600
CA50 [CAD aTDC]	5	10	10
EGR [%]	30	30	30
Intake pressure [bar]	1.5	2.0	2.5
Exhaust pressure [bar]	1.8	2.3	2.8
Intake temperature [K]	313	313	313
Engine speed [rpm]	1200	1200	1200

The 0-D simulation

In order to better know the fuel-chemical effects on PM, simulations are performed using a homogeneous reactor method. The mechanism used in this research composes of 272 species and 1,615 reactions. N-heptane, iso-octane and toluene are simulated with this mechanism, unfortunately, n-butanol is not included in this mechanism. Homogeneous reactor simulation is a common method to assess the sooting tendency of fuels (in fact is the approach to generate the so-called φ-T map). In this study the constant temperature simulation is used, which can provide fundamental insights regarding the sooting tendency for different fuels. The simulations are done with homogeneous mixtures at various equivalence ratios (from 0.1 to 7) and different initial temperatures (from 700 to 2,800 K). The soot nucleation process is quantified using the nucleation rate, which is calculated with eq. (2) using the concentrations of soot precursors naphthalene (A2) and naphthyl radical (A2*):

Table 5. Experiments of particle size distribution

Parameters	Load		
	Low	Medium	High
IMEPg [bar]	5	10	15
Fuel pressure [bar]	800	1200	1600
CA50 [CAD aTDC]	CA50 is changed from −4 to 12 CAD aTDC if possible		
EGR [%]	30	30	30
Intake pressure [bar]	1.5	2.0	2.5
Exhaust pressure [bar]	1.8	2.3	2.8
Intake temperature [K]	313	313	313
Engine speed [rpm]	1200	1200	1200

$$\omega_{\text{nucleation}} = 20\,k_{s1}\,\text{A2}\,\text{A2}^* + 20\,k_{s2}\,\text{A2}\,\text{A2}^* \qquad (2)$$

The unit of the elements in square bracket is molcm^{-3}. The kinetic parameters k_{s1} and k_{s2} are expressed in eqs. (3) and (4):

$$k_{s1} = 1e^{13}T^{0.5}e^{\frac{-15000}{RT}} \tag{3}$$

$$k_{s2} = 2e^{15}T^{-1.5} \tag{4}$$

where T is the temperature [K] and R is the molar gas constant [Jmol^{-1}K^{-1}].

Results and discussion

This section discusses the main results of this research. The first subsection discusses the experimental results with various fuel blends. The second subsection introduces the corresponding simulation results. Finally, the third subsection gives the particulate size distribution analysis.

Experiments on fuels of various mixing ratios

The results regarding the fuel composition ratio investigation are presented in figs. 3-5. Figure 3 shows clear trends that the indicated specific PM (ISPM) decreases as the volume percentage of iso-octane, butanol, and toluene increases. This is caused by the increased ID resulting from the decreased cetane number, as shown in fig. 4. Although ID for TRF and BRF are very similar, BRF produces less soot. This fact stimulates the research of the influence of the AF stoich on the soot emissions for equalized ID and composition ratio. The TRF, in general, produces higher ISPM than PRF and BRF at low composition ratios (30%~50%). However, the increased ID of TRF with higher composition ratio (60% and 70%) begins to strongly affect the soot formation process even though higher amounts of aromatic compounds are present. Longer ID enhances the mixing of fuel and air, thereby reducing the fuel-rich regions and resulting in less soot emissions.

Figure 5 shows the heat release rate (RoHR) comparison of PRF30, TRF30, BRF30 and PRF70, TRF70, BRF70 at medium load. The premixed combustion increases as the blend ratio increases, thus the physical effects on the soot processes must be considered in addition to the fuel-caused chemical effects. The combustion and soot process that occur later during the diffusion combustion will also be affected by the ignition quality of the fuel since the quasi-steady lift-off length is affected by the fuel ignition quality [14]. To meet the emission legislations regarding soot emissions as best as possible, PRF70, TRF70, and BRF70 were selected for further investigation. Also to investigate whether TRF70 will lead to more big-size particles when it has a longer ID to premix with air.
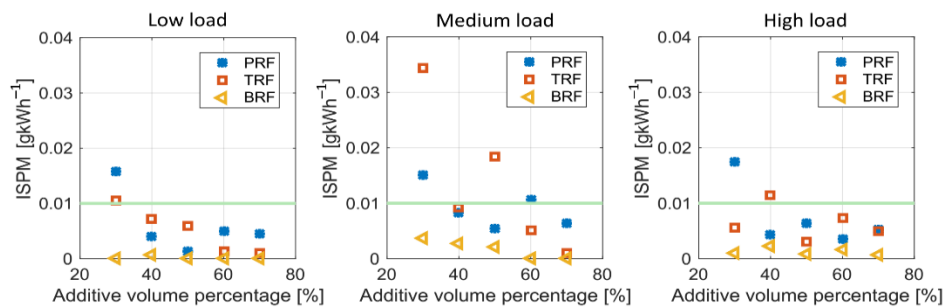

Figure 3. The ISPM at three different loads for different composition ratios of PRF, TRF, and BRF blends
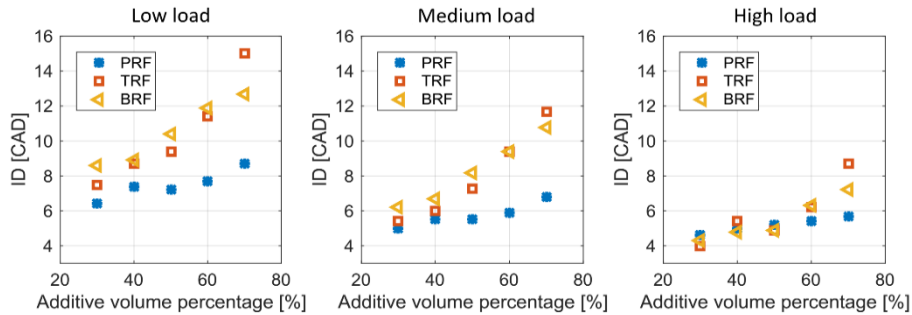
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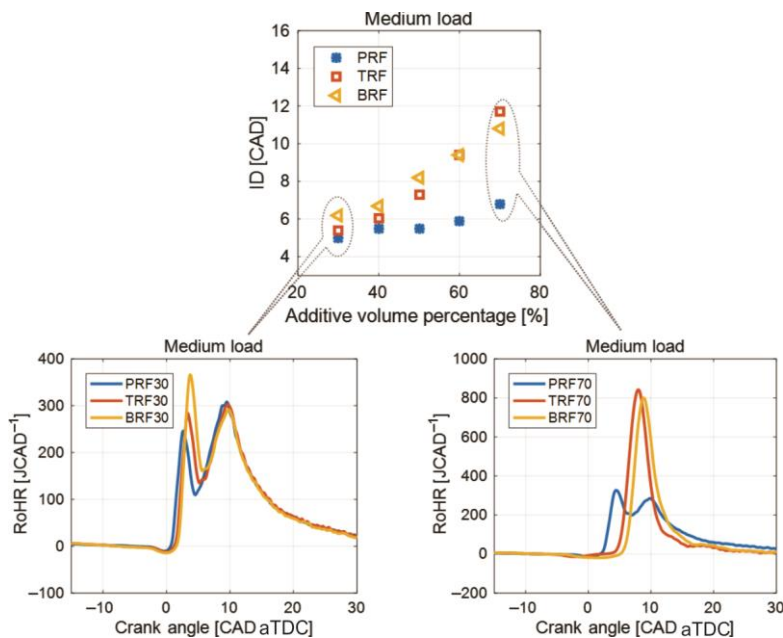**Figure 4. The ID plotted with fuel composition ratios at different loads**

**Figure 5. Comparison of the RoHR as a function of CA for
PRF30, TRF30, BRF30, and PRF70, TRF70, and BRF70 at medium load**

The 0-D simulation

Based on the results presented in previous section, only the nucleation rates of three neat fuels (n-heptane, iso-octane, and toluene, respectively) and two blends (PRF70 and TRF70) are shown and discussed. Since n-butanol is not included in the mechanism used in this study, unfortunately, no results of neat n-butanol and BRF70 will be presented. Toluene has a significantly higher soot nucleation rate compared to n-heptane and iso-octane, and its soot formation region is wider than n-heptane and iso-octane, as shown in fig. 6. The soot formation region of toluene reaches lower equivalence ratios, resulting in more soot formation with leaner mixtures compared to n-heptane and iso-octane. The soot nucleation rate of iso-octane is only marginally higher than n-heptane, which could originate from its extra one C-atom and two hydrogen atoms. Obviously, since NO_x is mainly characterized by the thermal mechanism (Zeldovich), the NO_x regions do not show any differences for all three fuels, they

are presented in figs. 6 and 7 for completeness. The TRF70 has a much higher soot nucleation rate than PRF70 since it contains 70 volume percentage of toluene, as shown in fig. 7. Therefore, it is expected in the experiments TRF70 produces higher soot emissions than PRF70 if TRF70 undergoes a similar mixing process as PRF70. However, the results of the experiments indicate that TRF70 is able to produce less soot than PRF70 (fig. 3) due to its lower local equivalence ratio resulting from the prolonged ID, fig. 4. Hence, it becomes interesting to investigate the particle size distributions of PRF70 and TRF70 to know whether the degree of mixing or chemical effects has a greater impact on particle size. This will be discussed further in the following section.
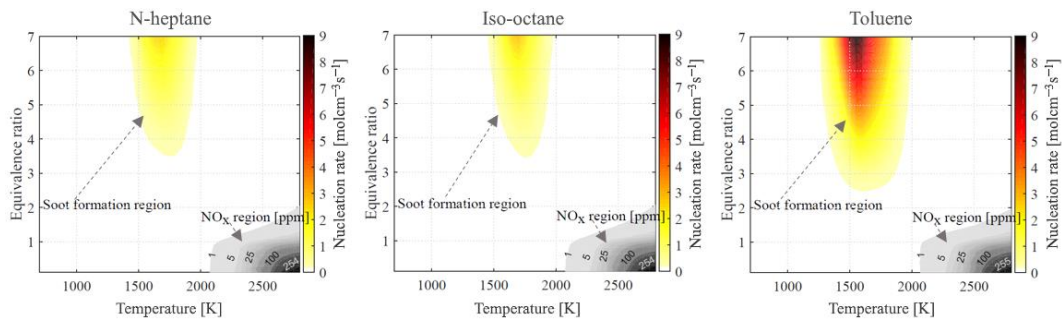

Figure 6. The soot nucleation rate and NO$_x$ region of n-heptane, iso-octane and toluene
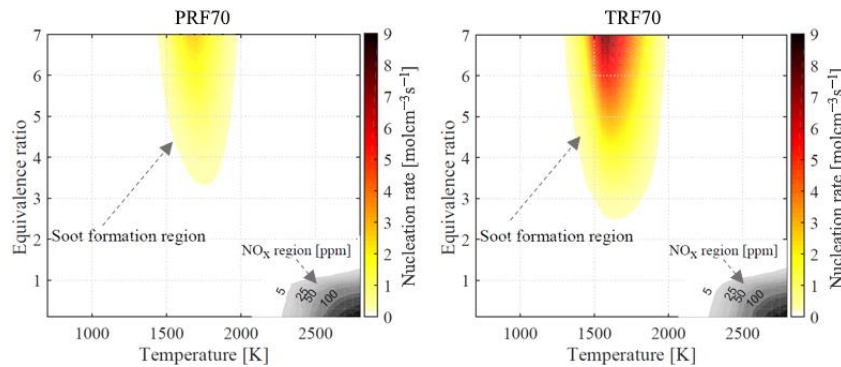(*for color image see journal web site*)

Figure 7. The soot nucleation rate and NO$_x$ region of PRF70 and TRF70
(*for color image see journal web site*)

Experiments on particle size distribution

The results based on changing CA50 are discussed in this section. Figure 8 presents the ID of PRF70, TRF70 and BRF70 at different loads. The TRF70 has the longest ID, followed by BRF70, and PRF70 has the shortest ID regardless of the operating load. The ID is strongly influenced by the engine operating conditions especially at low load where in-cylinder temperature and pressure are relatively low compared to medium and high loads. It is noteworthy that at low load for TRF70 controlling combustion phasing through adjusting the SOI is difficult when TRF70 is injected earlier into the cylinder, as shown in fig. 9. Given that TRF70 provides long ID for fuel to premix with air before ignition when SOI is before −19 CAD aTDC, the fuel chemistry plays an important role under this HCCI-like combustion
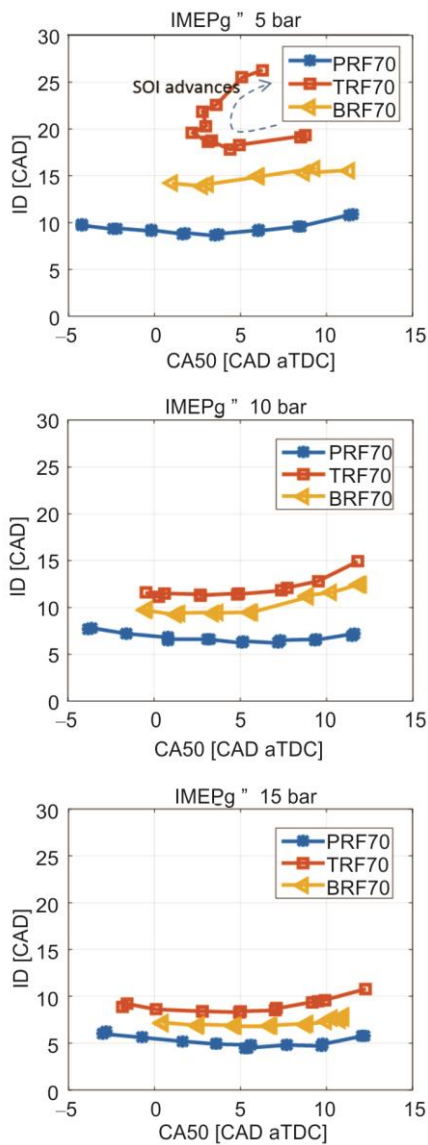
Figure 8. The ID of PRF70, TRF70 and BRF70 for different combustion phaings at various loads

mode where ignition timing is not controlled by injection timing but by chemical kinetics. When TRF70 and air have a sufficient mixing time, advancing SOI from −20 to −23 CAD aTDC causes overleaning and leads to lower combustion speed and a retarded combustion phasing. This is consistent with the findings of [15] that burn duration increases with ID for mainly premixed combustion. If mixing time is long, the mixture becomes leaner and will gradually approach the global λ. As the most reactive scalar is on the rich side of stoichiometry, ID will become longer.

In this paper, particles smaller than 20 nm are referred to as nucleation mode while particles larger than 20 nm are typical of accumulation mode. As shown in fig. 10, TRF70 produces less soot emissions at low load compared to PRF70 and more soot emissions at higher loads. The soot emissions of BRF70 are the lowest and increase with increasing load. The soot emissions of TRF70 at low load is chosen in the controllable area and the soot emissions of BRF70 at low load is zero. At 5 bar IMEPg, TRF70 is dominated by premixed combustion where the very long ID has a decisive effect on soot formation and produces most particles in the nucleation mode, as shown in fig. 11. The TRF70 has the longest ID independent of operating loads and combustion phasings. However, more mixing-controlled combustion appears with the increase in load and PM emissions shift to the accumulation mode, fig. 11. Hence, it is concluded that the fuel structure has a bigger impact on PM emissions than ID when the mixing-controlled combustion starts to influence the whole combustion process. Although BRF70 provides shorter ID than TRF70, it contains oxygen and therefore has a lower AFstoich. The BRF70 also has a much lower sooting tendency, so these two factors result in lower soot emissions and less particles in the accumulation mode, fig. 11.

Next, the effect of changing combustion phasing on PM emissions of PRF70, TRF70, and BRF70 is discussed. Figure 12 depicts the particle size distributions of these three blends at two different combustion phasings at low load. The accumulation mode is composed primarily of carbonaceous agglomerates and adsorbed material. They are formed in locally fuel-rich regions of the flame [16]. At low load, PRF70 has the shortest ID. Therefore, it is more likely that fuel-rich regions will be formed and more particles will be produced in the accumulation mode. Moreover, the particulate number concentration of PRF70 increases with the retarded

CA50 due to the decreasing combustion temperature. For TRF70, when CA50 is controlled in the vicinity of 3 CAD aTDC, it generates most of the particles in the nucleation mode, and the amount of particles generated in the accumulation mode is small. The change of CA50 does not have a noticeable impact on the I of TRF70, fig. 8. However, the decrease in combustion temperature reduces the oxidation of soot and results in the formation of larger particles.
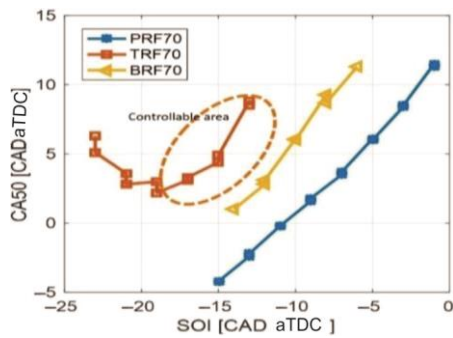

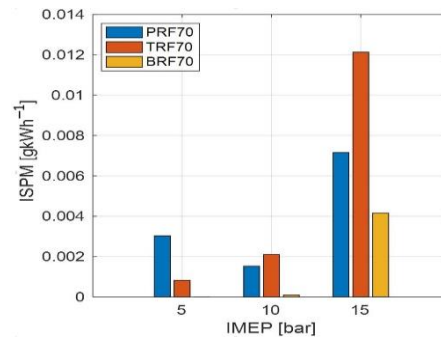**Figure 9. The SOI *vs.* CA50 at low load (5 bar IMEPg)**

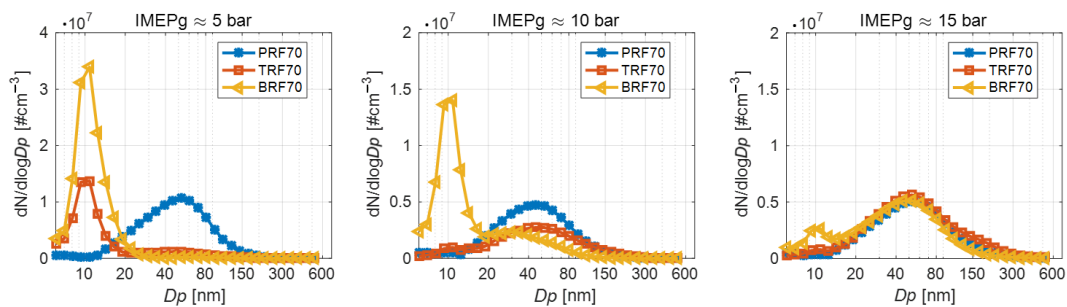**Figure 10. Soot emissions of PRF70, TRF70, and BRF70 with a constant CA50 (5 CAD aTDC) at different loads**

Figure 11. Particulate number concentrations and size distributions of PRF70, TRF70, and BRF70 with a constant CA50 (5 CAD aTDC) at different loads
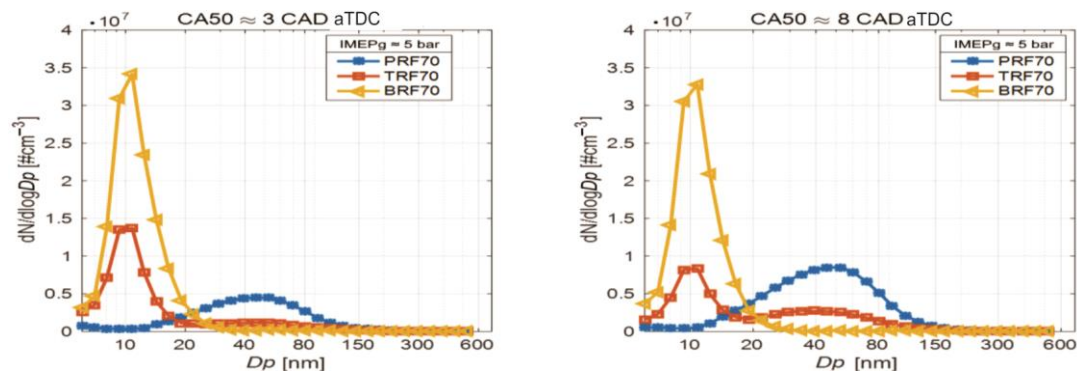

Figure 12. Particulate number concentrations and size distributions of PRF70, TRF70, and BRF70 for different combustion phasing's at low load

Conclusions

High-octane fuels have shown a great advantage in reducing soot emissions. In this paper, the effects of fuel structure and the selected engine parameter on particulate emissions regarding number concentration and size distribution are studied. Several conclusions could be drawn from this study.

Experiments on fuels of various mixing ratios.

− Increasing the volume percentage of iso-octane, toluene and n-butanol in the blends increases the ID and consequently decreases the emitted soot emissions.
− The BRF produces the lowest ISPM regardless of the composition ratios and operating loads.
− The ID is an important factor which affects the in-cylinder soot processes. As ID increases, the proportion of premixed combustion increases.
− If ID is not kept constant, the physical effects on the soot processes must be considered in addition to the fuel-caused chemical effects.

Simulation:

− In the simulation, toluene is found to have the highest soot nucleation rate.
− Iso-octane and n-heptane have approximately identical nucleation rates.
− If mixing process is the same for PRF70 and TRF70, TRF70 will produce more PM due to its higher nucleation rate.

Experiments on particle size distribution:

− The control of combustion phasing for TRF70 through adjusting injection timing at low load is difficult.
− The TRF70 produces most particles in the nucleation mode at low load due to its longest ID. At such conditions TRF70 is sufficiently mixed with air to reduce locally fuel-rich regions, thus reducing soot formation.
− Even though TRF70 still has longer ID than BRF70 and PRF70 at higher loads, the increase of mixing-controlled combustion makes the fuel-caused chemical effects have a greater influence on the formation of larger particles.

Acronyms

AD	– aerodynamic diameter	IMEP	– indicated mean effective pressure
AFStoich	– air-fuel stoichiomrtric ratio	IMEPg	– gross indicated mean effective pressure
CAD	– crank angle degree		
CA10	– crank angle where 10 percent of the heat has been released	ISPM	– indicated specific particulate matter
CA50	– crank angle where 50 percent of the heat has been released	LHV	– lower heating value
		LTC	– low-temperature combustion
CN	– cetane number	MON	– motor octane number
EEPS	– engine exhaust particle sizer	PM	– particulate matter
EGR	– exhaust gas recirculation	PPC	– partially premixed charge compression ignition
HCCI	– homogeneous charge compression ignition		
		RON	– research octane number
HD	– heavy duty	RoHR	– rate of heat release
ID	– ignition delay	SOI	– start of injection

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