An experimental investigation of wide distillation fuel based on CTL on the combustion performance and emission characteristics from a CI engine

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Abstract

Coal to liquid (CTL) has promising application prospects as alternative diesel fuel, but the direct application of coal-based synthetic diesel with high cetane number (CN) in compression ignition (CI) engines also has problems. Therefore, the CTL is blended with gasoline to adjust the physicochemical properties of the fuel, which is necessary to meet the requirements of efficient and clean combustion. From the perspective of fuel design and combustion boundary condition control, the effects of CTL/gasoline blends on the combustion performance and emission characteristics in a CI engine are investigated in this study. Meanwhile, the variation in the start of injection (SOI) along with the addition of exhaust gas recirculation (EGR) permits achieving clean combustion with CTL/gasoline blends. Experimental results present that adding gasoline to CTL forms wide

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distillation fuel (WDF), which is conducive to reducing the required mixing timescale and lengthening the chemical preparation timescale. CTL/gasoline blends bring in a higher premixed combustion ratio (PCR) and keep NO_x and soot emissions at the lowest level after introducing EGR. Simultaneously, the inhibition effects of CTL/gasoline blends on particulates' emissions are apparent with or without EGR due to prolonged ignition delay (ID) and improved quality fuel-air mixture, and the particulate mass of CG60 is significantly reduced by above 90% compared to pure CTL. In addition, the CTL/gasoline blends show refined engine characteristics for broad SOI, and the addition of gasoline to CTL is valid to alleviate the deterioration of combustion processes and emissions caused by EGR. Coupling EGR and gasoline addition is an effective way to break the trade-off relationship between NO_x and particulates' emissions for CTL.

Keywords: Coal to liquid; Wide distillation fuel; Combustion process; Particulate mode; Pollutant emissions

Highlights:

- CTL with high reactivity is not conducive to improving PCR and it is susceptible to EGR.
- The CTL/gasoline blends as WDF suppress particles' emissions and promote the shifting of particles towards smaller sizes.
- The CTL/gasoline blends are beneficial to enhance the tolerance of the combustion processes and emissions results to EGR.
- Coupling EGR and gasoline addition is an effective way to break up the trade-off relationship between NO_x and particulates' emissions for pure CTL.

Abbreviations

AHRR	Apparent heat release rate	FTS	Fischer-Tropsch synthesis
AMP	Accumulation mode particle	IMEP	Indicated specific fuel consumption
ATDC	After top dead center	ISFC	Indicated specific fuel consumption
BTDC	Before top dead center	NMP	Nucleation mode particle
CA	Crank angle	SHC	Specific heat capacity
CD	Combustion duration	SOI	Start of injection
CI	Compression ignition	PCR	Premixed combustion ratio
CN	Cetane number	NMP	Nucleation mode particle
CTL	Coal to liquid	TDC	Top dead center
ECU	Electronic control unit	WDF	Wide distillation fuel
EGR	Exhaust gas recirculation		

1. Introduction

The internal combustion engines (ICEs) have been undergoing innovation and development since their birth, continuously promoting the progress of the global economy [1-3]. Applying an electronically controlled high-pressure common-rail injection system permits independent control of pressure establishment and mass of fuel injection, so the flexible adjustment of the boundary conditions in the compression ignition (CI) engine is achieved to acquire better performance and fuel economy. Therefore, the CI engine has a relatively wide application field as power machinery, especially in industry, agriculture, and transportation. In the face of schemed severe legislation on emissions, there is still challenging to break up the trade-off relation between NO_x and soot emissions of diesel engines on account of mixing-controlled combustion [4-6]. The reason for this phenomenon is that the conventional combustion process for the CI engine creates the locally uneven distribution of the mixture and temperature in the cylinder: the high initial temperature of flame is surrounded by excess air leading to NO_x generation while fuel spray is wrapped by high temperature flame leading to soot formation [7, 8]. Although many manufacturers are trying to equip vehicles with advanced after-treatment systems, these complex systems increase costs and cannot solve the fundamental contradiction. Therefore, advanced combustion concepts and technologies have become the hope of breaking through emission limitations, and the demands for pollutants reduction accompanied by higher thermal efficiency compel the researchers to develop superior combustion strategies as soon as possible. Aiming to overcome poor air-fuel mixing, it is the future trend to actively introduce the premixed combustion mode into CI engines. These new combustion concepts are classified into low-temperature combustion (LTC) [9], including homogeneous charge compression ignition (HCCI) [10], premixed charge compression ignition (PPCI) [11], and reactive controlled compression ignition (RCCI) [12] as representatives. The typical LTC adopts the ultra-high injection pressure and heavy EGR to promote the premixed combustion [13, 14], which is aimed at keeping the local combustion temperature sufficiently low and reduce the local fuel-rich area in the cylinder to maintain the low level of emissions.

More importantly, energy has always been a prerequisite for human civilization, so petroleum resources are vital to society's economic growth and sustainable development [15, 16]. Liquid fossil fuels are still the main supply for ICEs as an energy source due to their high energy per unit density and facilitation of storage and transportation, so there is a contradiction between the shortage of fossil fuels and the growing demand for the energies. In allusion to the severe global energy problems [17-19], searching for new alternative fuels has gradually attracted people's attention [20, 21], such as hydrogen energy, biomass fuels, and synthetic fuels, which have the potential to be suitable for existing ICEs [22-26]. Meanwhile, it is essential to develop an energy diversification strategy based on the local resource structure to secure the national energy supply. Coal to Liquid (CTL), which uses coal as raw material, has been commercialized in countries with abundant coal reserves, providing a practical approach to solving the oil supply problem with domestic coal resources, such as South Africa, Indonesia, and China [27-30]. For the above countries with relatively high coal consumption, it is strategic to apply CTL through Fischer-Tropsch synthesis (FTS) technology to ensure clean and efficient coal utilization and guarantee energy security [31-33]. The FT chemical processing as an indirect conversion path [34] makes the various natural resources (such as coal, natural gas, and biomass) converted into intermediate gas products firstly, and then above intermediate gas products are transformed into liquid hydrocarbon fuel [35]. Simultaneously, the CTL as the FTS product is designed to meet or exceed the required specifications of commercial diesel [36, 37]. As a high-quality and clean alternative diesel fuel, CTL with a high cetane number (CN) is mainly composed of saturated alkanes, and it is basically free of sulfur and aromatic compounds. Because of the desirable properties of CTL, it can be used directly or blended with traditional diesel in any proportion to reduce dependence on fossil fuels, and lower fuel consumption also can be found by CTL [38-40]. Many researchers have proven that FT fuels have broad application prospects as a promising alternative fuel for CI engines [31-36]. It is demonstrated that the application of CTL as an alternative fuel not only has a positive effect on traditional pollutants from CI engines, such as total hydrocarbons (HC), carbon monoxide (CO), nitrogen oxides (NO), and particulate matter (PM) [41, 42], but also produces lower unregulated emissions (formaldehyde, acetaldehyde, and ozone formation potential) than diesel [43, 44]. Valentin Soloiu et al. did the research employing a Box-Behnken design matrix to investigate the correlation between thermal characteristics of Fischer-Tropsch coal-to-liquid fuel in relation to low temperature heat release (LTHR), ignition delay, and combustion delay within the negative temperature coefficient region (NTC) [45]. Vicente Bermúdez et al. tested gaseous emissions and fuel consumption with five different fuels in a light-duty diesel engine with EURO IV, and it concluded that the use of FT fuel causes lower regulated and unregulated emissions and fuel consumption than diesel [43]. The reduction in carbonyl compound emissions was evaluated by Bin Hao et al. using diesel engines running on FT diesel fuel synthesized from coal (CFT), and the result showed that the use of CFT resulted in a remarkable reduction in carbonyl emissions and ozone formation potential of the carbonyl compounds present in comparison with using diesel fuel [44]. An FT fuel has been experimented using a light-duty diesel engine under the New European Driving Cycle (NEDC) by O Armas et al. [46], and there are noticeable reductions in THC, CO, and particle mass compared to conventional diesel. Furthermore, extensive research on the production and application of CTL has been carried out at the economic and technical level, making a more comprehensive evaluation of FTS technology [47-53]. The findings indicate that the liquid fuels by FTS provide a feasible way that is viable both technologically and economically. The advanced process of CTL-based poly-generation schemes combined with co-production mode will guarantee a better solution to environmental protection and resource utilization, greatly enhancing the industrialization of FTS.

However, diesel-like fuels' high reactive autoignition characteristics make it hard to control combustion rates and suffer from inhomogeneous mixture formation, so single diesel-like fuels may not be the ideal choice for advanced combustion mode to realize premixed combustion operation [54, 55]. Because the combustion and emission performance of ICEs are closely related to the physical and chemical properties of the fuel, optimizing the fuel composition structure and modifying the fuel's physical and chemical properties can meet the requirements of efficient and clean combustion processes. Research institutions in various countries have invested much effort in improving fuel characteristics [56-58]. The studies found that fuel with low distillate and cetane number (CN) had great potential to form a homogeneous mixture and control the combustion process. Therefore, it is expected that these two different fuels with opposite but complementary properties can improve fuel adaptability, reaching a better compromise in the new combustion mode [56]. The ideas of wide distillation fuel (WDF) have been proposed to find a way out of a difficulty about the trade-off between NO_x and soot emissions in CI engines [60]. WDF is the fuel with a wide distillation range from the initial boiling point of gasoline to the final boiling point of diesel. Fortunately, as a petroleum product widely used in ICEs, low-reactivity gasoline can suppress ignition sufficiently, and its high volatility does a favor to the homogenization of fuel-air, so gasoline has been expected as a suitable choice as diesel fuel additives which is the available means to obtain WDF. Diesel and gasoline blends may also change spray characteristics due to the changed physical properties, finally impacting the in-cylinder mixture formation, engine combustion, and emissions [61]. As the gasoline fraction increases, the density, surface tension, and kinematic viscosity of blends will also decrease, and their evaporation and low-temperature fluidity are improved [62, 63]. Hence, diesel and gasoline blends have higher volumetric injection rates, earlier injection starts, and shorter injection delays [64]. H.Fujimoto et al. confirmed that the addition of the high-volatility fuels promotes the evaporation of low-volatility fuels, shortens the spray penetration distance, and enlarges the spray cone angle [65]. Compared with the volatility of the fuel, the CN of fuels also plays a more critical role in physical and chemical processes in the cylinder. Zhong et al. experimented with the HCCI combustion quality of gasoline and diesel blend fuels, and they had referred to the new combustion technology as Dieseline firstly [66, 67]. Dieseline utilizes better volatility of gasoline to prepare a homogeneous mixture and high CN of diesel to be suitable for CI. Han et al. [68] used diesel and gasoline blends in the LTC strategy and found that adding gasoline could decrease the reliance on heavy EGR usage and combustion temperature reduction better-controlling emissions. Chen et al. [69] studied adding gasoline into diesel/biodiesel blends at various speeds and loads under different EGRs to reduce NO_x and soot emissions. Chaudhari et al. [70] obtained clean combustion and improved efficiency with diesel-gasoline blends, and the engine load-carrying capacity can be extended without decreasing the engine performance by combining low and high reactivity fuels. In conclusion, it has been generally recognized that fuels with properties between diesel and gasoline are feasible and potential fuels for current advanced CI engines [71–73].

To sum up, fuel design and composition reconstruction have gradually become an essential path for efficient and clean combustion of ICEs. The CTL with higher CN is blended with gasoline with the resistance to autoignition and high volatility, adjusting reactivity and distillation range of blended fuels. Up to now, some scholars have adjusted the properties of Fischer-Tropsch fuel by adding n-butanol in different ways to achieve better combustion and emission performance [74]. However, adding gasoline to CTL forming WDF needs to be more comprehensively investigated, and this research is essential for its practical application in the future. The objective of the present study is to explore combustion performance and emission characteristics with different blending ratios of CTL/gasoline blends in a CI engine, while the variation in the start of injection (SOI) along with the addition of exhaust gas recirculation (EGR) permits achieving clean combustion with CTL and CTL/gasoline blends. In addition, there is a close relationship between PM emissions and harmful effects on human health attracting people's attention, and the structure, surface chemistry, and reactivity of soot particles are very susceptible to fuel formulation and engine operating parameters [75, 76], so it is necessary to measure and analyze particle characteristics. To better understand the particulate matter of CTL/gasoline blends, this study measured the exhaust particle size distribution (PSD) and analyzed the mass and number concentration of particles produced by the WDF based on the CTL. The significance of this work is to adjust the physical and chemical properties of CTL, an excellent alternative fuel, providing an alternative technical approach to achieve partially premixed compression ignition.

2. Experimental system and test procedure

2.1 Experimental engine and apparatus

An inline four-cylinder CI engine equipped with high-pressure common-rail and turbocharging is used as the test engine in this study. The original engine is modified into a single-cylinder CI engine with controllable combustion boundary conditions to control the fuel injection and intake parameters flexibly, and only the third cylinder is equipped with independent intake, exhaust, and fuel injection systems. **Table 1** shows the detailed specifications of the test engine, and **Fig. 1** makes clear a schematic diagram of the test platform. The test platform mainly includes a modified CI engine, fuel injection control system, intake system, dynamometer, oil-water temperature control system, combustion parameters analysis system, and emissions data collection system. An electronic control unit (ECU, NI2106) is adopted to realize data monitor and online dynamic adjustment of injection parameters. In addition, the independent intake parameters are adjusted through a self-designed simulated supercharging system, and the high-pressure air from the compressor is stabilized by a two-stage pressure surge chamber, entering the third cylinder of the test engine finally. The intake pressure of the engine is flexibly adjusted between 0 MPa and 0.3 MPa (gauge pressure) by a pressure sensor and a flow-limiting valve. The EGR system stabilizes the pressure of the exhaust surge chamber by changing the opening degree of the exhaust back-pressure valve, and then controls the EGR rate by adjusting the EGR valve.

Table 1. Engine specifications

Category	Properties
Geometric compression ratio	17.1
Cylinder diameter / mm	95.4
Piston stroke / mm	104.9
Connecting rod length / mm	162
The number of nozzles holes	7
Injector orifice diameter / mm	0.12
Oil jet cone angle / (°)	12
Swirl number	0.97
The shape of combustion chamber	ω

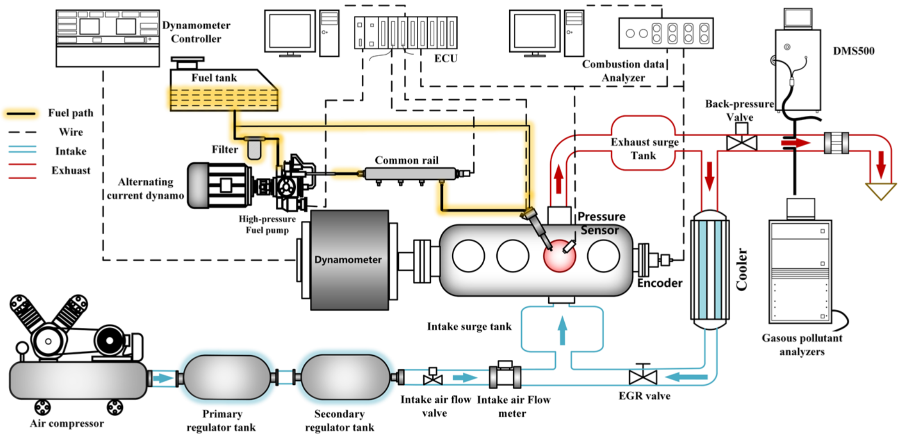


Fig. 1 Schematic diagram of experimental setup

The emissions measuring system is set up to collect pollutants data from the exhaust gas. NO_x concentration analyzer (Cambustion CLD 500), A fast HC analyzer (Cambustion HFR 500), and a fast CO&CO_2 analyzer (Cambustion NDIR 500) measure the concentration of NO, THC, and CO_2 separately. An electron particle spectrometer (Cambustion DMS500) is used to acquire the particle size distributions (PSDs). According to the PM diameter, the particle size is divided into three categories, namely nuclear mode particles (NMPs) (<35 nm), agglomerated particles (AMPs) (>35nm), and ultrafine particles (<100 nm). The combustion parameters acquisition and analysis system are composed of a piezoelectric sensor (Kistler 6052C) and a data acquisition system, which collects the in-cylinder pressure and other combustion parameters. For each operating point, 200 cycles of data are collected and averaged to eliminate measurement errors. The main parameters of the equipment are shown in **Table 2**.

Table 2. Main equipment

Category	Measuring instruments	Manufacturer	Accuracy
Dynamometer	CW260	CAMA	Torque: ± 0.5 NM Speed: ± 2 r/min

Rotary encoder	S4001	Bangman	—
Fuel flow meter	FX-100	ONO-SOKKI	± 0.12%
Air flow meter	20R100	TOCEL	± 1%
Cylinder pressure	6052C	Kistler	± 1%
CO&CO_2	NDIR500	Cambustion	< 2% FS/hour
HC	HFR500	Cambustion	< 1% FS/hour
NO_x	CLD500	Cambustion	< 5 ppm/hour
Particle	DMS500	Cambustion	—

2.2 Methodology and test conditions

CTL and gasoline are selected as primary fuels in this study, and adding gasoline in different proportions to the CTL to make blended test fuels. In addition, the conventional diesel is attached as a reference fuel. **Table 3** compares the main physical and chemical properties of test fuels in this work. The CTL used is a hydrocarbon fuel obtained from coal as a raw material through the FTS, which consists of 95.8% chain alkanes, 3.4% cycloalkanes, 0.5% polycyclic aromatics and 0.3% monocyclic aromatics in molar percentage. In addition, this study adopts CTL/gasoline blended fuels defined as CGXX, of which the XX represents the ratio of the calorific value of gasoline to the total calorific value of the fuel. For example, the blended fuel composed of 60% CTL and 40% gasoline (by calorific value) is defined as CG40. Moreover, the engine operating conditions are shown in **Table 4**.

The engine speed is maintained at 1400 r/min, and the load rate was about 60% (the indicated mean effective pressure (IMEP) was about 1.0 MPa). The calorific value contained in the fuel injected into the cylinder is fixed at 1704 J/cycle for all test fuels. In this experiment, the proportion of gasoline in the blended fuel is set as 20%, 40%, and 60%. The start of injection (SOI) is selected from 2 °CA before the top dead center (BTDC) to 14 °CA BTDC, and the EGR rate is set to 0% and 30%. It is worth mentioning that the EGR rate is defined as the ratio of the CO_2 content in the intake and the exhaust of the engine, and the CO_2 content was measured by the exhaust gas analyzer. When the EGR rate is adjusted by controlling the mass of introduced exhaust gas, the pure air flow rate into the intake remains at 43kg/h unchanged. During the experiment, the temperature of coolant is controlled at 358±2 K.

Table 3. Main properties of tested fuels

Fuel property	CTL	Diesel	Gasoline	CG20	CG40	CG60
Density (25°C) / ($kg\cdot m^{-3}$)	757.0	840.0	690	743.6	730.2	716.8
Cetane number / (CN)	75.4	52.9	13	62.92	50.44	37.96
Low calorific value / ($MJ\cdot kg^{-1}$)	43.07	42.69	42	42.87	42.64	42.42
Total Aromatics / %	≤ 0.8	≤ 3.6	0	≤ 0.64	≤ 0.48	≤ 0.32
Sulfur content / 10^{-6}	0.38	3.7	~	~	~	~
Viscosity (25°C)/ ($mm^2\cdot s^{-1}$)	2.14	3.64	0.59	1.83	1.52	1.21
Theoretical air-fuel ratio	14.96	11.2	14.7	14.91	14.86	14.8

Table 4. Engine operating conditions

Category	Properties
Engine speed / ($r\cdot min^{-1}$)	1400 ± 2

IMEP / (MPa)	About 1.0
Injection pressure / (MPa)	100 ± 2
SOI / (°CA BTDC)	2-14
EGR ratio / (%)	0 and 30
Inlet air temperature / (°C)	25 ± 1
Cooling water temperature / (°C)	80 ± 1

For the DMS500, the concentration is expressed as a concentration size spectral density in dN/dlogDp (/cc), with units of N (/cc). It allows easy integration over any size range to give a total particle concentration, and the calculation equation is as follows:

$$N = \int_{D_{P1}}^{D_{P2}} \frac{dN}{dlog(D_P)} dlog(D_P)$$

At the same time, given a size/number distribution, it is desirable to calculate in real-time a mass concentration, so the conversion equation is as follows:

$$Mass(\mu\mathrm{g}) = \text{Density Factor} \cdot Dp^{power\ factor}$$

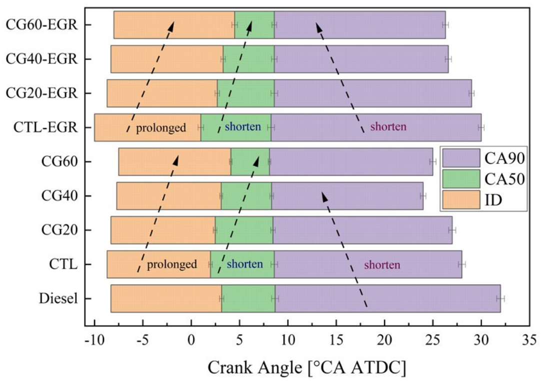
For diesel engine agglomerates, research shows that the DMS500 mass calculation gives good agreement with gravimetric techniques using a density factor of $2.2{\cdot}10^{-15}$ and a power factor of 2.65.

3. Results and discussions

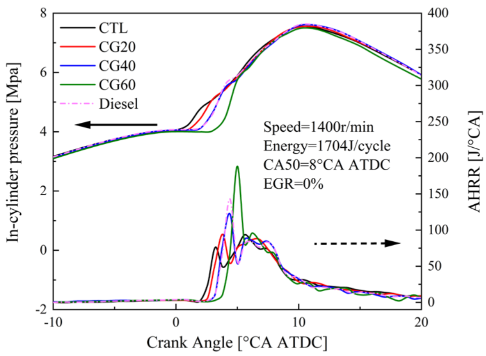
3.1 Effects of CTL/gasoline blends on combustion and emissions characteristics

As a potentially alternative diesel fuel, CTL is demonstrated to improve the combustion and emissions performances of the CI engine. However, the higher fuel reactivity of CTL makes itself fast auto-ignite and has poor fuel-air mixing, so it is hard to attain a higher premixed combustion mode. To optimize the feature of CTL by adjustment of physicochemical properties and fuel components, adding gasoline with low CN and high volatility to CTL forming WDF is a potential and feasible solution to improve fuel adaptability to future CI engines.

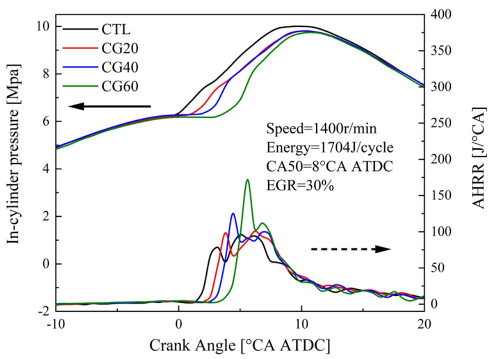
3.1.1 The effects of CTL/gasoline blends on the combustion performance



(a) Combustion phasing



(b) In-cylinder pressure and HRR with 0% EGR



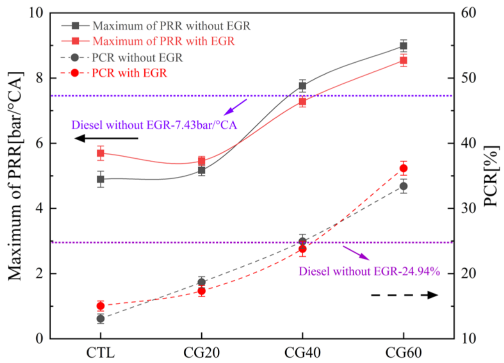
(c) In-cylinder pressure and HRR with 30% EGR

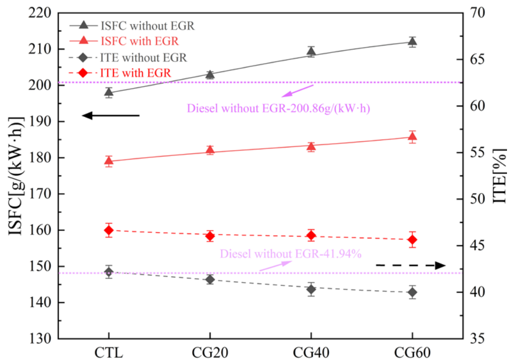
Fig. 2 In-cylinder pressure, the HRR, and the combustion phasing of different fuels at 0% and 30% EGR rates

Fig. 2 displays the variations of combustion phasing, in-cylinder pressure, and apparent heat release rate (AHRR) of test fuels at different EGR rates. In order to eliminate the influence of different combustion phasing, adjusting injection timing keeps CA50 constant at 8 °ATDC for all test fuels. The CAXX represents the crank angle (CA) at which the accumulated heat release reaches XX% of the entire cycle heat release. Ignition delay (ID) is defined as the time between SOI and CA10, and CA10-CA90 represents the combustion duration (CD). **Fig. 2(a)** shows the combustion phasing of different fuels. The higher CN and chemical reactivity indicate better ignitability, so the CTL can reach the ignition conditions earlier than diesel with increases of the temperature and pressure in the cylinder before combustion, making the CTL have a shorter ID than conventional diesel. What also can be found is that the CD of conventional diesel is the longest. Compared with the CTL, the poor reaction conditions during the later diffusion combustion process make the diesel with low-reactivity burn more slowly. Even if conventional diesel has similar CN with CG40, CG40 has better volatility, which is beneficial to the formation of the combustible mixture in the later combustion, avoiding the phenomenon of combustion tailing. It also can be seen from **Fig. 2(a)** that SOI gradually advances as the gasoline proportion in the fuel decreases to maintain the same CA50 at a 0% EGR rate. For high CN fuels as CTL, their branched chain reaction in the low temperature range, including extraction of hydrogen atoms from the fuel, oxygenation, isomerization reactions and the decomposition of ketone hydro peroxide molecules, produces a large amount of active free radicals such as OH, triggering a cold flame reaction at a certain temperature and leading to premature ignition eventually [73]. Compared with pure CTL, the CD is gradually shortened, and ID is prolonged after adding gasoline, and this trend becomes more prominent with the gasoline ratio increasing. The essence of this phenomenon is that adding gasoline as an inert additive changes the reaction pathway. There are weakened and slowed low-temperature reactions for WDFs inhibiting OH formation reaction and reducing the exotherm in the low-temperature range, so ID is prolonged by CTL/gasoline blends. Besides, due to the lowest fuel reactivity of CG60, the main combustion heat release process is postponed to the stage of the expansion stroke accompanied by the piston descending, slightly prolonging the CD compared to CG40.

After introducing 30% EGR, the injection timing of pure CTL needs to be drastically advanced to be consistent with CA50 without EGR, so the starting point of the heat release is also significantly earlier. The sharp contrast is that the injection timing of CTL/gasoline blends only needs to be advanced slightly at a 30% EGR rate to keep the same CA50 at 8 °ATDC. The above phenomenon can be attributed to that EGR gas makes in-cylinder oxygen content and temperature lower during the compression stroke, severely deteriorating the mixing condition of fuel and air for CTL with higher CN and poor volatility. Currently, hugely advanced injection timing is beneficial to form combustible mixtures to ignition for CTL. The gasoline addition with better volatility promotes spray breakup, atomization, and evaporation processes of the CTL, which can effectively optimize the combustion condition and reduce necessary mixing times before the start of ignition, so there is a slightly advance of injection for CTL/gasoline blends at a 30% EGR. The EGR leads to a decrease in the oxygen concentration, so the probability of collision between fuel and oxygen molecules decreases during the chemical reaction process, inhibiting the reaction rate and prolonging the ID and CD. However, CG60 minimizes the impact of EGR on CD. Under EGR operating conditions, ID still prolonged with the increase of gasoline content, while CD showed the opposite trend. As the gasoline proportion increases, the amount of later combustion is gradually reduced because of larger premixed combustion volume shortening CD. Compared to the pure CTL with obviously advanced injection timing, the CD of CG40 and CG60 was also reduced by 24.71% and 31.99%, respectively. Therefore, the combustion process is not sensitive to EGR by CTL/gasoline blends; in other words, and the wide distillation blended fuels are better tolerated to EGR than pure CTL.

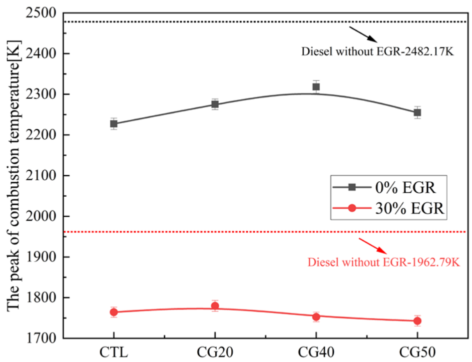
Comparing to the In-cylinder pressure and AHRR curves of conventional diesel in the **Fig. 2(b)** attached as a control group, there are reduced peak values of in-cylinder pressure and AHRR for the CTL. In addition, since the CN of conventional diesel is close to CG40, these two fuels exhibit similar combustion process. **Fig. 2(b)** shows that all test fuels have similar peaks of the in-cylinder pressure when the CA50 and the total calorific value of the injected fuel remain the same without EGR. However, as the gasoline content increases, the peak value of AHRR rises sharply. It can be concluded that gasoline plays a role in promoting premixed combustion in the early combustion stage. Due to gasoline with lower CN, increasing the gasoline ratio makes the start of heat release lag behind the CTL in turn. What can be seen from **Fig. 2(c)** is that the in-cylinder pressure has increased significantly after the introduction of EGR, mainly because that the EGR rate is adjusted by introducing the exhaust flow while fixing the inlet flow by means of the simulated supercharge system, increasing intake pressure and charge at a high EGR rate. It is necessary to significantly advance the injection timing of pure CTL at a 30% EGR rate, keeping the CA50 unchanged. Hence there is a significant increase in the in-cylinder pressure peak, but the peak of premixed combustion still maintains the lowest level compared with CTL/gasoline blends. The changing trend of the AHRRs is consistent with no EGR for four test fuels, and premixed combustion has dominated the entire combustion process as the gasoline proportion increases. In conclusion, adding gasoline moderately reduces the fuel cetane number of CTL extending the timescale of the mixture ignition chemistry, and reduced distillation temperature and improved volatility lessening the required timescale for mixture formation.





(a) Maximum of PRR and PCR at 0% and 30% EGR rates

(b) ISFC and ITE at 0% and 30% EGR rates



(c) The peak of combustion temperature

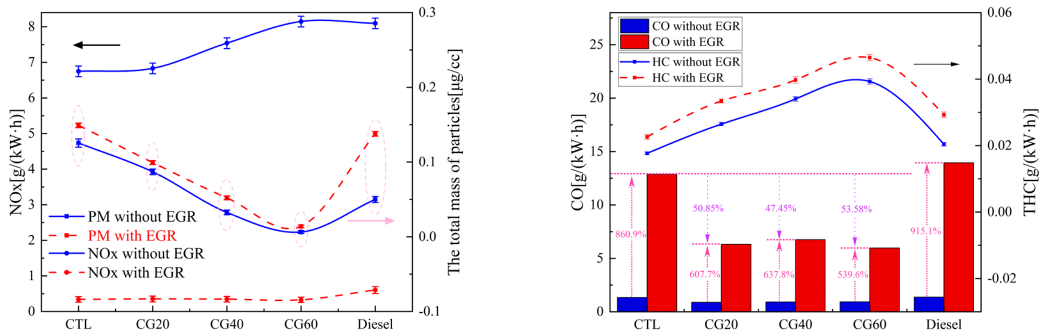
Fig. 3 Combustion characteristic of different fuels at 0% and 30% EGR rates

Fig. 3 depicts the combustion characteristic of four test fuels at 0% and 30% EGR rates, including that **Fig. 3(a)** shows maximum of pressure rise rate (PRR) and premixed combustion ratio (PCR), **Fig. 3(b)** shows indicated specific fuel consumption (ISFC) and indicated thermal efficiency (ITE), and **Fig. 3(c)** shows the peak of combustion temperature. The combustion characteristics of conventional diesel fuel are given as baseline in **Fig. 3**. This paper defines the premixed combustion ratio (PCR) as the ratio of the cumulative heat release of the premixed combustion endpoint (the crank angle corresponding to the minimum value of the heat release acceleration) to the total cumulative heat release. PRR maximums and PCRs maintain an upward trend with the increase in gasoline content seen from **Fig. 3(a)**. For the pure CTL with high CN, the shortest ID keeps the lowest peak of AHRR, making the smallest PCR with or without EGR. It also illustrates that creating a homogenous mixture requires a very long mixing timescale, but the chemical ignition timescale is very short for CTL. The addition of gasoline reduces the reactivity of the blends and promotes the mixing process of fuel and air, so more combustible mixtures are produced during the longer ID due to the higher gasoline content in CG40 and CG60, and these mixtures are ignited simultaneously with the ignition of the CTL, considerably enhancing the PCR, and this improvement is strengthened with the improved gasoline content. Thus, the faster premixed combustion speed and shorter combustion duration significantly elevate the PRR maximums of CTL/gasoline blends, which is also the reason for the shortening of the period during the middle and late combustion stages. It can be found that PCR increases linearly with the increase of gasoline content at no EGR, and the PCR of CG20, CG40, and CG60 fuels increase by 42.74%, 90.67%, and 155.50% compared with the pure CTL, respectively. Moreover, there are similar cylinder pressure and AHRR curves shown in **Fig. 2(b)** because diesel and CG40 have similar CN, so the maximum of PRR and PCR of these two fuels are very close. However, the pure CTL's tolerance to EGR is weak, so it is needed to sharply advance injection timing after introducing 30% EGR, making maximum of PRR and PCR higher than no EGR. The diffusion combustion of CTL also dominates the whole combustion process of CG20 due to the low gasoline content. Nevertheless, CG40 and CG60 have lower reactivity and higher volatility improving the mixing process and prolonging the chemistry timescale, which allows more time for fuel vaporization and consequently yields better homogenization of the fuel, air, and residual gas mixture at 30% EGR. Meanwhile, the mixture will pack more EGR gas during the fuel-air mixing process smoothing the combustion rate, so the PRR maximums of CG40 and CG60 with obviously delayed start of combustion are lower than that of 0% EGR by 6.19% and 5.01% severally. ISFC and ITE show the contrary tendency as the gasoline ratio in the fuel rises from **Fig. 3(b)**. It can be seen from the figure that the ISFC of CTL is lower than that of diesel, because the CTL's ID is shorter improving the propensity for constant volume combustion. In this study, due to the regulation of adjusting EGR, the higher intake pressure brings extra work after introducing EGR, improving ITE. With or without EGR, as the proportion of gasoline in the fuel increases, it can be seen that the ITE decreases and the ISFC increases gradually. It is mainly due to the higher latent heat of vaporization for gasoline, and the proportion of gasoline improved leads to the increase of the locally fuel-lean area leading the incomplete combustion. It is worth noting that, after the introduction of EGR in this test, the ISFC of CTL/gasoline blends are closer to the pure CTL.

Fig. 3(c) is a comparison of in-cylinder peak temperature for different fuels. The peak combustion temperature of CTL is significantly lower than that of conventional diesel, mainly due to the large proportion of diffuse combustion and almost contenting no aromatic hydrocarbons with high adiabatic temperature. Even if CG60 has a larger PCR, a delayed heat release process and a more uniform mixture are not conducive to the generation of local high temperature, and this phenomenon is especially obvious after the introduction of EGR. Since the EGR reduces the intake oxygen concentration and introduces more three-atomic molecules (H_2O, CO_2), increasing the specific heat capacity (SHC) of the working fluid in the cylinder, the combustion temperature drops significantly at 30% EGR. Diffusion combustion of CTL is still dominated for CG20, so the peak temperature is at the same level. For CG40 and CG60, the peak temperature is lower than that of pure CTL and diesel. The above phenomenon is because the expansion of the fuel distillation range is conducive to the homogenization of the components in the cylinder and a large amount of exhaust gas is wrapped in the mixture, which reduces the combustion temperature and makes the combustion process softer.

3.1.2 The effects of CTL/gasoline blends on the emission characteristics

It is not difficult to see from the above discussion that the addition of gasoline to CTL is conducive to the active introduction of premixed combustion mode. Simultaneously, CTL/gasoline blends extend chemistry timescale and increase fuel-air mixing rate in favor of balance between the mixing timescale and chemistry timescale. Therefore, this section analyzes and studies the gaseous pollutants and particulates' emissions of CTL and CTL/gasoline blends.



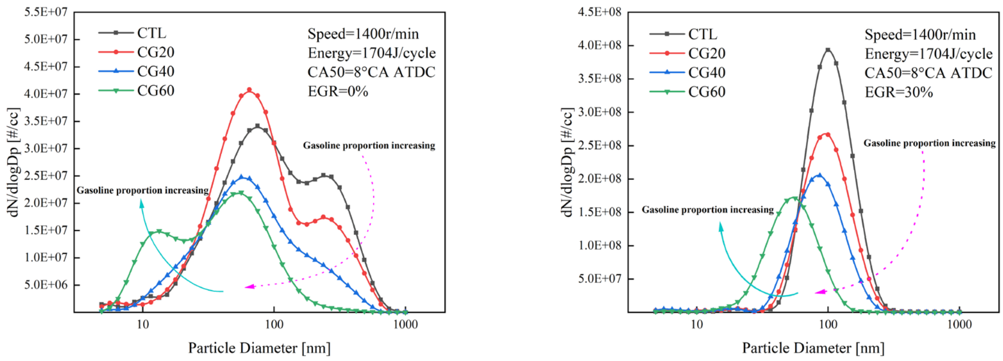
(a) NO_x and particle

(b) THC and CO

Fig. 4 Comparison of the NO_x, THC, and CO emissions for different fuels

Fig. 4 displays the comparison of the NO_x, THC, and CO emissions for test fuels. It is possible to see from **Fig. 4(a)** that adding 20% gasoline does not cause a significant change in NO_x, but NO_x emissions of CG40 and CG60 rise continuously. The main reason is that the increase in gasoline ratio significantly improves the fuel and air mixing process and PCR providing more local oxygenrich conditions and higher temperature, making for NO_x generation. Due to the highest in-cylinder temperature of diesel, its NOx emissions are close to CG60. Fortunately, 30% EGR introduced can effectively suppress NO_x formation and keep NO_x emissions at low level due to lower in-cylinder temperature and oxygen content slowing down combustion speed. The NOx emissions of the four test fuels are significantly lower than that of diesel with 30% EGR. CG40 and CG60 promote the homogenization of fuel, air and exhaust gas, while EGR slows down the combustion rate granting more time to transfer heat to cooler regions, so these factors help avoid local build-up of temperature avoiding NO_x formation. For the particulates' emissions, emissions of particles masses have risen after introducing EGR, and this phenomenon is very significant for diesel especially. The EGR makes oxygen content and temperature decrease extending more part of combustion into the expansion stroke. The nucleation mode particles formed cannot be oxidized effectively, but surface addition reactions are still favored as they don't require high temperatures [74], ultimately deteriorating particles' emissions. Compared to CTL, CG20, CG40, and CG60 can reduce the masses of particles' emissions to 30.78%, 74.01%, and 94.97% at the condition of sufficient oxygen content in the cylinder. There also is a linear decline of particles' emissions as the gasoline proportion increases at a 30% EGR rate. Adding gasoline with lower reactivity into CTL can prolong the ID, and gasoline with better volatility improves the atomization and evaporation process of blends, promoting the quality of the fuel-air mixture during the chemical preparation period. The above benefits of CTL/gasoline blends are remarkable after a drastic deterioration of combustion conditions caused by the EGR. It is evident that the improvement degree of CG60 to the total mass of particles is maintained at above 90% with or without EGR compared to pure CTL. Above all, CTL/gasoline blends combined with EGR can break the trade-off relationship between NO_x and PM, and the increase in the gasoline proportion can make particulates' emissions more resistant to EGR.

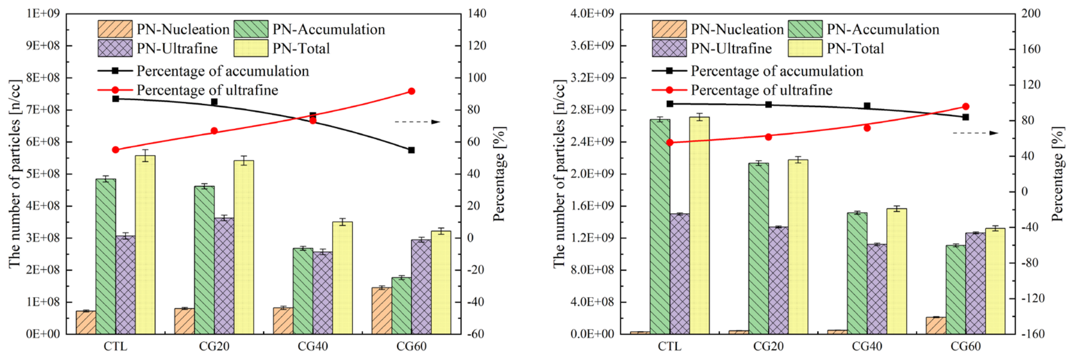
Fig. 4 (b) presents the variations in HC and CO emissions for CTL and CTL/gasoline blends. Due to the introduction of EGR, the combustion temperature and oxygen concentration in the cylinder decrease, resulting in a significant increase in THC and CO emissions compared to that of no EGR. As shown in the figure, the THC emissions gradually increase, but CO emissions gradually decrease as the gasoline blending ratio increases. As the proportion of gasoline increases, the longer ID makes more fuel adhere to the cylinder wall and the locally fuel-lean regions are more easily formed, so THC shows an increasing trend. The CO emissions have close relations with combustion quality, which mainly depends on the fuel-air mixture formed for CI engines. After the introduction of EGR, the CO emission of diesel increased by 9.15 times even exceeds the CTL, indicating that the combustion of diesel is deteriorated significantly and conventional diesel has poor adaptability to EGR. Fortunately, the CTL/gasoline blends can greatly improve PCR maintaining a higher combustion quality and shortening CD, so the CO emissions are inhibited ultimately. CO emissions are relatively low in the absence of EGR, but it increases after introducing EGR. The reduction in CO of CTL/gasoline blends is maintained at above 50% compared with pure CTL. Compared with no EGR operating, the decrease in in-cylinder oxygen content owing to EGR leads to an apparent growth in CO emissions by 8.61 times for the pure CTL. However, CO emissions of CG20, CG40, and CG60 at 30% EGR are 6.08, 6.38, and 5.40 times higher than no EGR, respectively, so this upward trend of CO emissions has been alleviated by adding gasoline to CTL. The above results prove again that CTL/gasoline blends can weaken the sensibility of the combustion process to the lacking oxygen in the cylinder.



(a) PSD at 0% EGR (b) PSD at 30% EGR

Fig. 5 Comparison of CTL and CTL/gasoline blends on particle size distribution at 0% and 30% EGR

Fig. 5 displays the particle size distributions (PSDs) of CTL and CTL/gasoline blends at 0% and 30% EGR rates. As shown in **Fig. 5 (a)**, the PSDs of four test fuels show bimodal distribution at 0% EGR. As the proportion of gasoline increases, both peaks of the particle numbers gradually shift to smaller particle sizes. Especially, CG60 keeps the first and the second peaks at around 13nm and 50nm, and the particles larger than 36.52nm decrease immensely. Due to the early flame zone chemistry has a strong influence on the total surface area of the first particle [74], a larger proportion of diffusion combustion makes more locally high-temperature and oxygen-lean regions during the combustion process, strengthening the formation of particles with the larger size for CTL. Nevertheless, CTL/gasoline blends can improve the quality of the fuel-air mixture, eliminating locally fuel-rich regions to the maximum extent, which is not conducive to the inception of particles suppressing the AMPs emissions and shifting particles towards NMPs with smaller sizes. The introduction of 30% EGR results in a unimodal distribution in the PSDs curve from **Fig. 5 (b)**, and the magnitude of the particle number increased from 10^7 to 10^8. Introducing EGR reduces the oxygen concentration and forms many oxygen-lean areas in the cylinder, deteriorating the combustion process and increasing the availability of suitable THC fragments. The fuel in some oxygen-lean areas fails to contact with oxygen fully and undergoes the process of crack, cyclization, and dehydrogenation under high-temperature conditions, finally forming carbonaceous particles with loose and porous surfaces. These porous carbonaceous particles whose specific surface area is large easily absorb THC and grow continuously and accumulate each other, generating more AMPs with larger sizes. Fortunately, as the proportion of gasoline added to CTL continues to rise, this upward trend of particulates' emissions can be effectively suppressed. The CTL/gasoline blends can not only optimize the spray characteristics, but also rely on the volatility of gasoline to form more combustible mixtures during the prolonged ID, which greatly enriches the quality of the fuel and air mixture. It is not helpful to the formation of particles inception for CTL/gasoline blends. In conclusion, as the gasoline proportion increases in the CTL/gasoline blends, the negative impact of EGR on PM has significantly been alleviated.



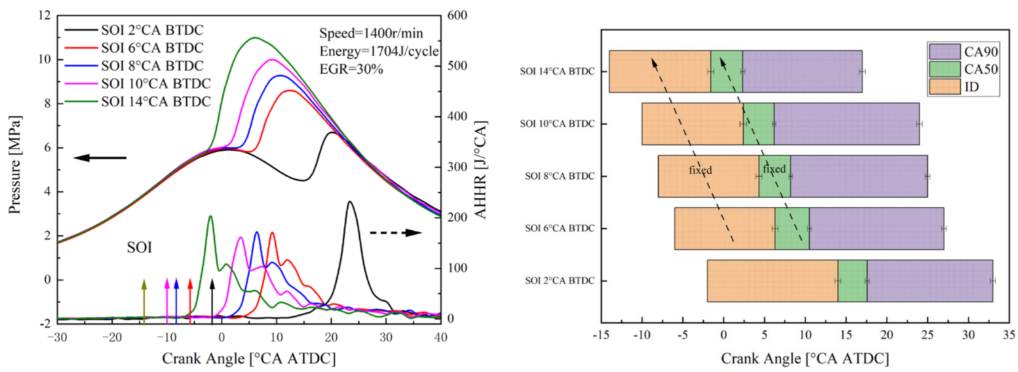
(a) 0% EGR (b) 30% EGR

Fig. 6 Comparison of CTL and CTL/gasoline blends on different modes of particle at 0% and 30% EGR

Fig. 6 manifests the comparison of CTL and CTL/gasoline blends on different modes of the particle at 0% and 30% EGR. Regardless of EGR, the changes in particle characteristics show consistent features as the proportion of gasoline increases. The number of total particulates and AMPs both decrease significantly with the increase of the gasoline content, and the proportion of AMPs has gradually diminished. As shown in **Fig. 6 (a)** at 0% EGR, the AMPs proportion in CTL has reached 87.00%, but CG60 has dropped the AMPs proportion to 54.84%. Compared with CTL, the total number of particles of CG20, CG40, and CG60 is lessened by 2.72%, 37.10%, and 42.24%. The proportion of ultrafine particles rises with the gasoline proportion increasing, which explicates that CTL/gasoline blends induce the particle size to be smaller. It can be viewed from **Fig. 6 (b)** that the numbers of total particles and AMPs are closer, indicating that the AMPs have dominated the entire particle size distribution at 30% EGR. Only by increasing gasoline proportion to 60% is there a substantial increase in NMPs, and the ultrafine particles account for the most of total particulate number. Due to the introduction of EGR, the reduced oxygen content in the cylinder precipitate imperfect combustion, so the combustion in the cylinder gradually deteriorates. The possibility of the formation of locally high-temperature areas with oxygen-deficient in the cylinder is increased, which makes more primary particles generated, aggregated, and condensed, and more THC emissions also promote the growth of particles. It is generally believed that NMPs are mainly formed in the process of exhaust gas cooling and dilution, and their components are mainly soluble organic matter (SOF) and some ultrafine carbon cores [75]. Therefore, the number of AMPs whose adsorption of unburned HC is enhanced increases, inhibiting the NMPs generation. Eventually, the number of particles has risen sharply, and the proportion of AMPs has also increased at 30% EGR. CTL/gasoline blends are beneficial to the homogenization of fuel and in-cylinder charge as an increase of gasoline content, so there are improved quality of fuel-air mixture and a large amount of premixed combustion, significantly restraining the formation of large-size particles. After introducing EGR, the improvement of particles' emissions by adding gasoline is more prominent. Compared with CTL, the total particle numbers of CG20, CG40, and CG60 are reduced by 19.65%, 42.21%, and 51.28%.

3.2 Effects of fuel injection strategy on the combustion and emissions of the CTL/gasoline blends engine

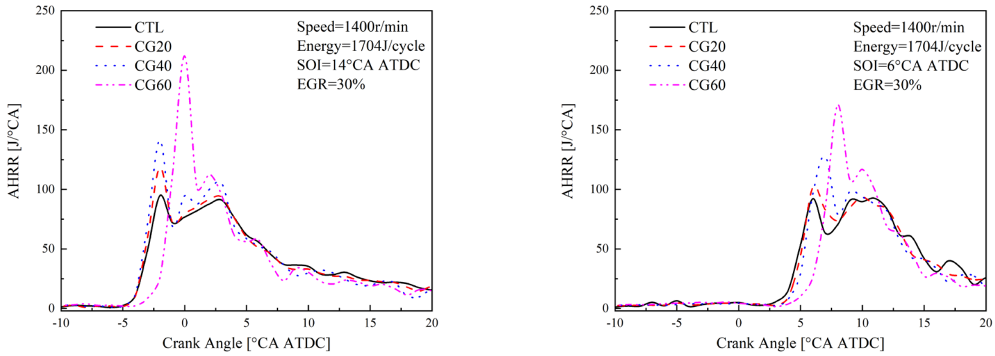
To further study the adaptability of CTL/gasoline blends to changes in combustion phasing, this section details combustion performance and exhaust emissions characteristics of CTL and CTL/gasoline blends at different injection timings combined with EGR achieving clean combustion.



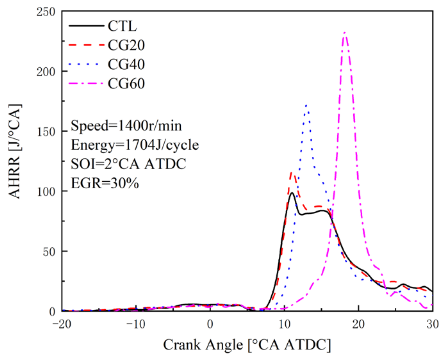
(a) In-cylinder pressure and AHRR (b) Combustion phasing

Fig. 7 Comparison of combustion characteristics at different SOI for CG60

Fig. 7 shows the influence of SOI on the combustion characteristics for CG60. The research on injection timing is carried out at a 30% EGR rate, mainly to avoid tremendous combustion noise and mechanical load causing by the earlier injection timing. As shown in **Fig. 7 (a)**, when the injection timing is set to 2°CA BTDC, the combustion process is mainly postponed to the expansion stroke of the CI engine, which is not suitable for the ignition of CG60 with low CN. Therefore, the start of heat release is very lagging, and the ID is greatly extended, delaying the CA50 significantly. Under the circumstances, a separation between the end of the injection and the start of the combustion is achieved, and air and fuel mix extensively prior to combustion. Hence, there is the highest peak value of AHRR whose curve tends to be unimodal, and it shows that the combustion process basically eliminates the diffusion combustion. With the continuous advance of the SOI, the maximum cylinder pressure increases because of better constant volume degrees. It also can be found from **Fig. 7 (a)** that the ID and the period of CA10 to CA50 remain almost constant, so the CA50 gradually advances linearly closer to the top dead center (TDC) with the change of injection timing from 6°CA BTDC to 14°CA BTDC from **Fig. 7 (b)**. The AHRR curves move forward and maintain the same shapes, but there is a higher peak value of AHRR as the SOI advances to 14°CA BTDC.



(a) SOI of 14 °CA BTDC (b) SOI of 6 °CA BTDC



(c) SOI of 2 °CA BTDC

Fig. 8 Comparison of AHRRs between CTL and CTL/gasoline blends at different SOI

To clearly elaborate the combustion characteristic of fuels with different distillation range at various SOI, **Fig. 8** shows the comparison of AHRR between CTL and CTL/gasoline blends at SOI of 14 °CA BTDC, 6 °CA BTDC and 2 °CA BTDC. As the proportion of gasoline increases at the fixed SOI, the AHRR peak gradually becomes higher, but the start of heat release of CG60 is significantly behind mainly because the lowest fuel reactivity prolongs the ID obviously. When the SOI is relatively advanced, the start of heat release for CG20 and CG40 is basically the same as CTL, but their AHRRs have higher slopes at SOI of 14 °CA BTDC from **Fig. 9 (a)**. Because 30% EGR reduces the oxygen concentration and combustion temperature in the cylinder, which leads to the deterioration of the combustion atmosphere, but the addition of gasoline improves the formation of the combustible mixture during the preparation of chemistry, optimizing the ignition conditions and significantly increasing PCR which contributes to fast combustion speed. With the postponement of SOI, the increase in the gasoline proportion makes the start of heat release gradually delayed from **Fig. 8 (b)** and **(c)**. When the fuel injection is closer to the TDC, a relatively higher cylinder temperature and pressure are more suitable for ignition and heat release of CTL with high CN, so fuel reactivity has become more prominent for the start of heat release at SOI closer to TDC. The lagging injection timing at 2 °CA BTDC makes the AHRR peak of CG40 and CG60 significantly increase. At the same time, the boundary between premixed combustion and diffusion combustion of CG40 has been obscured, and it is radically close to full premixed combustion for CG60.

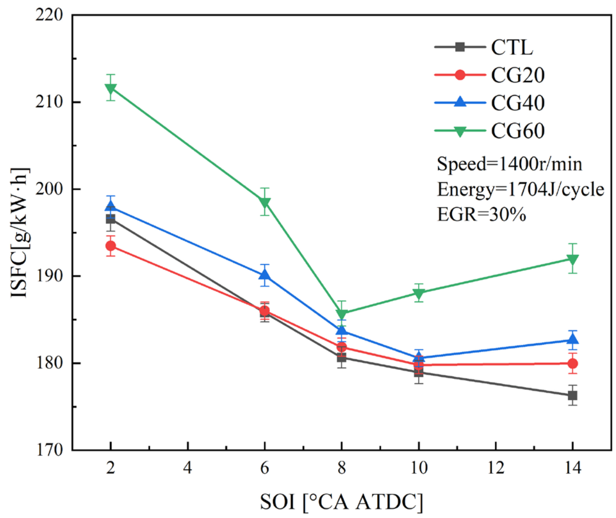
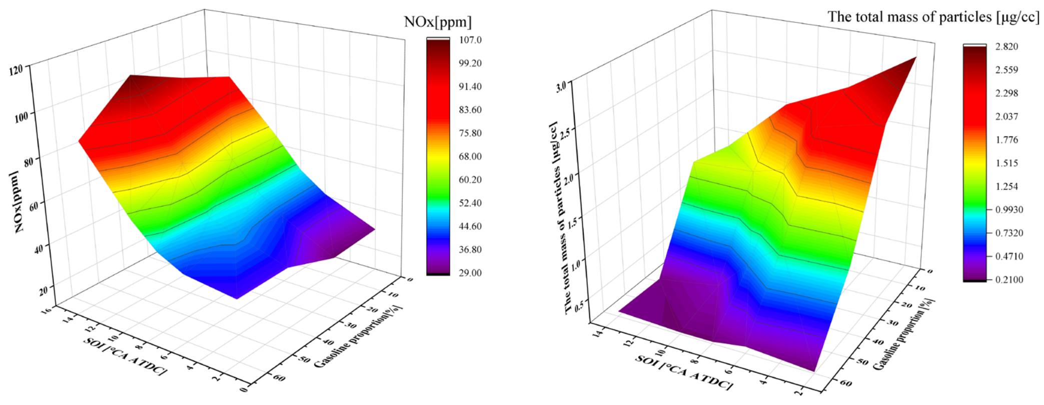


Fig. 9 Influence of SOI on the ISFC of the CTL and CTL/gasoline blends

According to the above discussion, SOI has a noticeable influence on the combustion characteristics of four test fuels, so the fuel economy also needs to be discussed. **Fig. 9** shows the influence of SOI on the ISFC of the CTL and CTL/gasoline blends. It can be found from the figure that as the fuel injection timing is advanced from 2°CA BTDC to 10°CA BTDC, the ISFC of the four test fuels shows a linear downward trend since early injection will cause the CA50 to approach the TDC, improving the propensity for constant volume combustion. When the fuel injection time is close to TDC, adding 20% gasoline helps to improve the quality of the mixture before the start of heat release increasing PCR, and there is no delay in combustion heat release, so the ISFC of CG20 is reduced by 3% at SOI of 2°CA BTDC compared with CTL. When the SOI gradually changes to 14°CA BTDC, the bigger PCR results in a faster combustion rate and maximum of PRR, making most of the heat release maintain at the point before TDC. Therefore, an increase in the negative work that the piston must overcome decreases the fuel economy. Due to a large increase in PCR of CG60, the ISFC starts to increase at injection timing earlier than 8°CA BTDC. However, since CTL is still dominated by diffusion combustion with SOI of 14°CA BTDC, the peak heat release rate of premixed combustion is low, keeping the most combustion process after TDC, which leads to the ISFC still maintaining a decreasing trend. At the same time, it also can be found that the fuel economy of CG20 and CG40 is not significantly inferior to the pure CTL. Owing to the largest proportion of gasoline in CG60, the local area with a lower in-cylinder equivalent ratio expands, resulting in incomplete combustion. Therefore, the ISFC of CG60 is significantly higher than other fuels eventually but keeps SOI at around 8 °CA BTDC to get a better fuel economy.



(a) NO_x emissions (b) Particulates' emissions

Fig. 10 Influence of injection timing on the emission characteristics of the CTL and CTL/n-butanol blends at 30%EGR

Fig. 10 presents the influence of injection timing on the emission characteristics of the CTL and

CTL/gasoline blends at 30% EGR. The increasing NO_x emissions can be seen from **Fig. 10 (a)** with the advanced injection. The reason is that the advanced injection makes the higher in-cylinder pressure and maximum of PRR. NO_x emissions of CG20 and CG40 are close to the level of CTL, and their NO_x emissions only show an increase when the injection timing is the most advanced (SOI of 14°CA BTDC). However, the NO_x emission of CG60 is lower than that of CTL. CTL/gasoline blends have a longer chemistry timescale and a better condition physical preparation before ignition, and this advantage becomes more prominent as the proportion of gasoline increases. These properties are conducive to the homogenization of the components in the cylinder, avoiding local high temperature and smoothing combustion rate. Although CG60 has the highest PCR, its NO_x emission does not increase but decreases. There is a downtrend of particulate mass as the gasoline proportion increases seen from **Fig. 10 (b)**, especially CG60 reduces PM mass by above 79.53% compared with pure CTL. With the advancement of SOI, the particle mass basically also shows a downward trend. The main reason is that the advanced SOI and enlarged gasoline ratio will increase the proportion of the combustible mixture before combustion and eliminate the excessively fuel-rich area, inhibiting the generation of particles essentially. It is proved once again that the CTL/gasoline blends have enhanced tolerance to EGR, and the addition of gasoline is more effective than blindly advancing SOI in improving particulates' emissions at heavy EGR. It can be seen that CG60 is able to decrease the total particulates' mass emissions by above 80% for all SOI compared with CTL. While maintaining the same level of NO_x emissions and ISFC as the pure CTL, CG40 and CG60 can reduce the total particulates' mass emissions by 81.34% at SOI of 10°CA BTDC and 89.54% at SOI of 8°CA BTDC compared to CTL.

All in all, combining gasoline and control of combustion boundary conditions can minimize emissions levels without deteriorating fuel economy and further break the trade-off relationship between NO_x and soot.

4. Conclusions

Combustion processes and emission characteristics are investigated based on a modified single-cylinder CI engine with flexible and adjustable control boundary parameters using CTL and CTL/gasoline blends under different EGR rates. It is an attempt to change the physical and chemical fuel properties of CTL by adding gasoline, meeting the needs of advanced combustion concepts and realizing the integration of gasoline and diesel engines in the future CI engine. The major conclusions can be summarized as follows:

- CTL has higher CN meaning higher fuel reactivity, so the better ignitability benefits shorten the ID. There is a smaller PCR due to poor fuel-air mixture quality CTL, so more diffusion combustion dominates the whole combustion process. The above shortcomings are fully exposed at 30% EGR. To maintain a consistent CA50 as no EGR, SOI must be greatly advanced after introducing EGR. At the same time, EGR has caused a significant increase in particulates' emissions of pure CTL.
- Adding gasoline with lower CN and high volatility to CTL forms WDF, which is conducive to reducing the required mixing timescale and lengthening the chemical preparation timescale. Prolonged ID and improved fuel-air mixture quality bring in a higher PCR increasing the peak of AHRR and fasting the combustion rate for CTL/gasoline blends. In addition, the ISFC rises mildly for CTL/gasoline blends because of the incomplete combustion of gasoline.
- CTL/gasoline blends raise the PCR as gasoline proportion increases rising combustion

temperature, which is conducive to NO_x formation. Fortunately, the combustion temperature in the cylinder was significantly reduced at 30% EGR, and the NO_x emissions of all tested fuels are kept at the lowest level. Simultaneously, the inhibition effect of CTL/gasoline blends on particles' emissions is evident with or without EGR, and CG60 can reduce the emissions of the total particles' mass by reaching above 90% compared to CTL. Coupling between EGR rate and gasoline proportion is an effective way to mitigate the trade-off relationship between NO_x and particulates' emissions.

- The change of SOI mainly affects the combustion phasing, but ID and the shapes of HRR curves are not sensitive to different SOI. However, the combustion process of CG60 basically eliminates the diffusion combustion at SOI of 2°CA BTDC. Although NO_x grows with the advance of SOI, it remains at a low level at 30% EGR. As for PM emissions, simply relying on advanced SOI to remedy the aggravation of particulates' emissions caused by the introduction of EGR has little effect for CTL. However, the addition of gasoline to CTL is a valuable way to alleviate the deterioration of combustion and emissions caused by EGR. In other words, CTL/gasoline blends make combustion performance and emission characteristics have enhanced toleration to EGR. While maintaining the same level of NO_x emissions and ISFC as the pure CTL, CG40 and CG60 can reduce the emissions of total particulates' masses by 81.34% at SOI of 10°CA BTDC and 89.54% at SOI of 8°CA BTDC compared to CTL.

Notes

The authors declare no competing financial interest.

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