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## Article

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## Research paper

## Challenges in charge preparation and combustion in homogeneous charge compression ignition engines with biodiesel: A review

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## ABSTRACT

Homogeneous charge compression ignition (HCCI) engines have high efficiency and lower  $\text{NO}_x$ -soot emissions than diesel and gasoline engines. However, these engines suffer from high HC, CO and unregulated emissions and combustion phasing control. The fuel chemistry, charge dilution and fuel injection timing strongly control HCCI combustion. The oxygenated renewable biofuels, such as biodiesel and alcohol, can be used to control combustion. The use of laser diagnostic techniques can provide insight into the combustion chamber for the air–fuel charge formation and combustion studies. The charge stratification achieved using intake air conditioning, dilution or varying the fuel reactivity can overcome the challenges associated with HCCI engines. The review paper examines the critical challenges in the HCCI engine, use of the renewable fuels and their impact on engine-out emissions.

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## 1. Introduction

Compression ignition (CI) engines are preferred for transportation and power generation due to their high efficiency. However, the depleting petroleum reserves and heavy nitric oxides ( $\text{NO}_x$ ) and particulate matter (PM) emissions are major concerns for their continual use in the transport sector (Reitz, 2013). The charge heterogeneity in CI engines is responsible for  $\text{NO}_x$ -soot formation and a waste of useful part of fuel into emissions (Han et al., 2014). The homogeneity of air–fuel charge depends on fuel injection strategies and physiochemical properties of the fuel (Bohl et al., 2017). Exhaust gas recirculation (EGR) is used in CI engine to reduce  $\text{NO}_x$  emissions, however, it leads to increase of hydrocarbon (HC), carbon monoxide (CO) and smoke emissions (De Serio et al., 2017). Hence, a trade-off is observed in  $\text{NO}_x$ -soot emission formation in CI engines. The after-treatment devices such as a selective catalytic reactor (SCR) and a diesel particulate filter (DPF) are used to reduce  $\text{NO}_x$  and soot emissions respectively. The DPF and SCR are costly sub-systems, require frequent regeneration over a period of time, and impose fuel penalty (Reitz, 2013). Hence, there is a strong requirement to reduce the fuel consumption and emission from these power producing devices by improving the in-cylinder combustion.

Biofuels, renewable in nature and produced locally, can be a sustainable alternative to petroleum-based fuels. They are found

to reduce engine-out emissions due to the presence of an oxygenated component in their composition. However, physical–chemical properties, such as high viscosity, stability, and volatility are a hurdle to their direct use in engines. The  $\text{NO}_x$  emission is found to increase with the higher percentage of biodiesel in blends with diesel in CI engine (Boggavarapu and Ravikrishna, 2013).

The biodiesels are composed of various fatty acid methyl esters or ethyl esters (FAME/FAEE) and their composition varies with the source (species) and location (Knothe, 2005). The biodiesels can be broadly categorized, based on a dominant component in their composition, into two types viz. saturated (e.g. Myristate, Palmitate, stearate) and unsaturated (e.g. Oleate, Linoleate, Linonenate). Saturated biodiesels, consisting of a high percentage of saturated fatty acids, show overall physio-chemical properties similar to that of diesel but with poor cold flow performance (Hoekman et al., 2012). The unsaturated biodiesel has higher viscosity and surface tension compared to diesel (Knothe and Steidley, 2005). The increased  $\text{NO}_x$  emission with use of biodiesel in CI engine is correlated to physical properties (viscosity, surface tension, bulk modulus), atomization, formation of air–fuel mixture and presence of inbound oxygen (Benjumea et al., 2010). Higher  $\text{NO}_x$  emission is attributed to higher bulk modulus, and shorter ignition delay (ID) due to high cetane number (CN). The unsaturated biodiesel produces higher  $\text{NO}_x$  emissions compared to saturated biodiesels (Lapuerta et al., 2009). It is essential to develop a combustion technology which is not sensitive to the composition of biodiesel and require minimum after-treatment devices for emission reduction.

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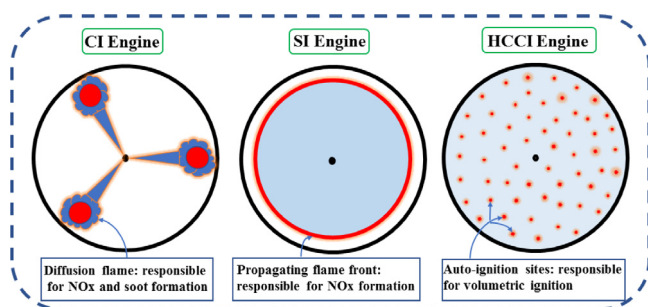


Fig. 1. Combustion in CI, SI and HCCI engine (Saxena and Bedoya, 2013).

The lean combustion based homogeneous charge compression ignition (HCCI) engine is found to be effective to reduce both soot and  $\text{NO}_x$  emission simultaneously (Dec, 2009). This review paper summarizes state of the art research on HCCI engine combustion in light of the use of biodiesel in CI engine. The following section discusses the characteristics of HCCI engine charge formation, combustion, and emissions. This will be followed by a review of regulated and unregulated emission from HCCI engine with biodiesels. The use of fuel reactivity, stratification and biodiesel for extending operating range of HCCI engine is analyzed. The optical diagnostic techniques used in HCCI combustion research are discussed in the last section.

## 2. HCCI engines

The HCCI engine uses lean homogeneous air–fuel mixture which is ignited by compression of the charge (Yao et al., 2009). The lean charge reduces the overall and local temperature leading to low  $\text{NO}_x$  and soot emission. Onishi et al. (1979), developed a ported 2-stroke, controlled auto-ignition engine, for stable lean-burn combustion as one of the initial attempt towards HCCI engines. They reported a reduction in fuel consumption and emissions. Najt and Foster (1983), extended the lean burn combustion to a 4-stroke single cylinder engine.

### 2.1. HCCI operation

The difference in operation of CI, spark ignition (SI) and HCCI engine is explained in Fig. 1. The combustion in CI engine is heterogeneous and flame is around the spray plume and combustion is initiated by auto-ignition of fuel. In the case of SI engine, combustion is initiated by spark and flame propagates in a homogeneous mixture. The HCCI engine operation combines the features of SI and CI engine where homogeneous mixture combustion is initiated by auto-ignition. Due to instantaneous volumetric heat release, the combustion duration is short and shows a high pressure rise rate (PRR). The operating characteristics of HCCI engines are shown in Fig. 2. At high load operations, the requirement of high equivalence ratio advance the ignition timing and results in high heat release rate (HRR), increases the  $\text{NO}_x$  emissions, and knocking. High equivalence ratio leads to poor cycle-to-cycle stability, and limits the maximum combustion phasing (Dec, 2009). At low load, incomplete combustion and combustion instability is observed under cold start condition with the use of high EGR. The combustion efficiency decreases and HC emission increases significantly for equivalence ratio ( $\phi$ )  $\leq 0.2$ .

It is observed that the high equivalence ratio requires a intake manifold that can handle high temperature charge. Also, the present microprocessor based feedback control system and associated hardware is unable to control the transient load HCCI operation within a few engine cycles and requires further development (Agarwal et al., 2017; Fathi et al., 2017).

### 2.2. HCCI combustion

The HCCI combustion exhibits two-stage ignition (Fig. 3) with high chemical reactivity fuels (HRF), such as diesel and biodiesel. The first stage of heat release is referred as low-temperature heat release (LTHR) where kinetic reactions are slow and occur at a temperature below 850 K with small (7%–10% of total) heat release (Zheng et al., 2001). The rest of fuel energy is released when the charge temperature is beyond 950 K during the high-temperature oxidation reaction, also referred as high-temperature heat release (HTHR). The LTHR and HTHR are separated by a negative temperature coefficient (NTC) regime (as shown in Fig. 3). The overall reaction rate exponentially decreases during the NTC region even with the increase in overall charge temperature (Battin-Leclerc, 2008). The combustion phasing thus depends on NTC region as it controls the start of main heat release rate. The charge reactivity during NTC period can be controlled by a pressure–temperature history of charge, EGR and by low reactivity fuel (LRF) such as gasoline, ethanol in case of dual fuel engine (Zhao, 2007).

### 2.3. Combustion chemistry

The oxidation of hydrocarbon in low-temperature zone is a complex process that involves various chain branching and chain propagation reactions (Gaffuri et al., 1997). A spontaneous ignition occurs when the number of chain branching initiation reactions is less than the chain termination reactions. The chain branching leads to isomerization reaction where straight-chain alkanes are converted to branched isomers by a heating process. This provides several radicals to start the auto-ignition reaction. The timing and amount of heat release in first stage ignition depend on the size and structure of the fuel molecule, and pressure–temperature history of charge. A better understanding of HCCI combustion of diesel and biodiesel fuels require detailed knowledge of the properties of individual components along with their chemical kinetics.

#### 2.3.1. Diesel

Diesel is a mixture of a number of hydrocarbon molecules that includes 41% paraffin, 30% cycloparaffins and 29% aromatics by volume (Battin-Leclerc, 2008). Paraffins and naphthalenes are saturated HC molecules. Aromatics and olefins are unsaturated HC which can convert carbon–carbon ( $\text{C}=\text{C}$ ) covalent or aromatic bond to a single bond by adding hydrogen (H) atom to the adjacent carbon (C) atom (Singh et al., 2017). A large fraction of long, straight-chain alkane (n-paraffins) species in diesel is responsible for two-stage characteristic ignition chemistry (Lapuerta et al., 2014). A long chain saturated HC molecule has high CN and short ID.

#### 2.3.2. Biodiesel

Biodiesel is a mixture of fatty acid esters. The CN of biodiesel increases with the increase in aliphatic chain length and decreases with increase in the number of double bond or branching in the chain (Knothe et al., 2003). The linolenic methyl ester, found in many biodiesels, has least CN while stearic methyl ester has highest. The oxidation of esters with aliphatic chain  $\text{C}_n > 4$  is observed to be similar as that of n-heptane/iso-octane (Coniglio et al., 2013). At low temperature, chain branching occurs by addition of oxygen molecule ( $\text{O}_2$ ) through ketohydroperoxide species. The chain propagation reaction of hydroperoxyalkyl (QOOH) species increases with the increase in ambient temperature. This leads to the formation of cyclic ether and olefinic molecules through ketohydroperoxide species (Coniglio et al., 2013).

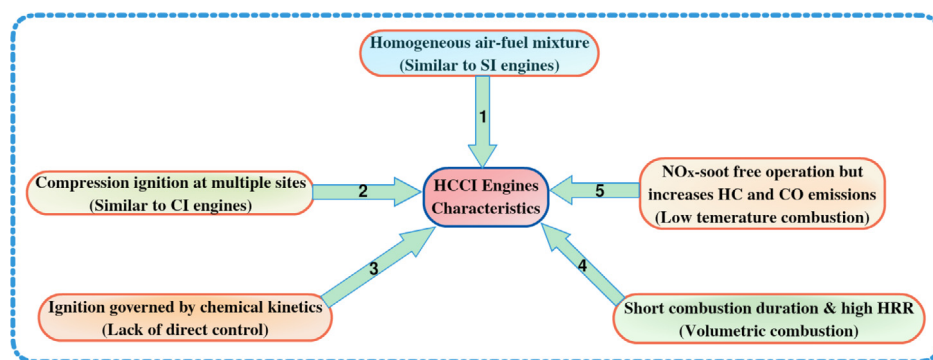


Fig. 2. Operating characteristics of HCCI engines.

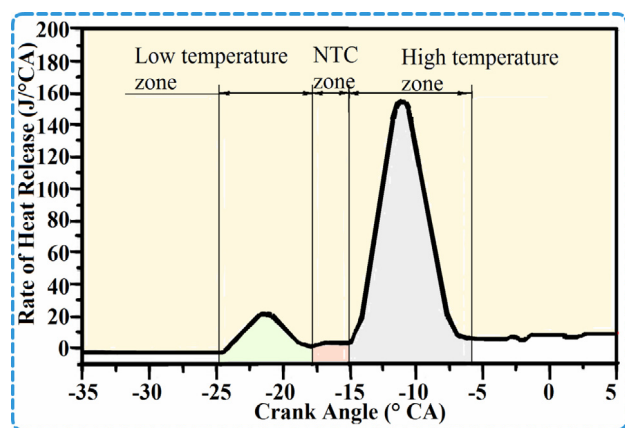


Fig. 3. Heat release curve of n-heptane HCCI combustion (Yao et al., 2009).

The LTHR and HTHR are initiated by the decomposition of ketohydroperoxide at low temperatures and rapid dissociation of hydroperoxide ( $\text{H}_2\text{O}_2$ ) into the hydroxide (OH) radicals. The oxidation mechanism involves temperature dependent competition between chain propagating channels and chain branching channels for esters with aliphatic main chain (Fisher et al., 2000). During the low-temperature reactions, temperature and pressure sharply increase over a limited temperature range (around 500 K). However, the reaction stops before combustion is complete due to decreased reactivity in NTC region (Westbrook and Dryer, 1984). The alkyl and alkyl ester peroxy radical isomerization to hydroperoxy alkyl and hydroperoxy alkyl ester radical reactions play a key role in the low-temperature chemistry and controls the auto-ignition of fuel (Ranzi et al., 1995). The number of double bonds and its location in the main aliphatic chain reduce the reactivity of unsaturated methyl esters during low-temperature region but increase the reactivity in the high-temperature zone (Lai et al., 2011). Majority of aliphatic chains in biodiesel shows various un-saturated chemical species that hinders the internal isomerization of a molecule.

### 2.3.3. Gasoline

Gasoline has nearly 42% saturated hydrocarbons, which are dominant with single sigma bond branched alkanes. Gasoline exhibits high volatility due to a high percentage of small hydrocarbon molecules,  $\text{C}_4\text{--C}_9$  (85%). However, smaller molecule requires higher energy to break the bonds, thus reducing the fuel reactivity (Battin-Leclerc, 2008).

### 2.3.4. Alcohol

Alcohol affects the hydroxyl (OH) radical formation, decreasing the concentrations of reactive intermediate species (formaldehyde ( $\text{CH}_2\text{O}$ ), OH, hydroperoxyl ( $\text{HO}_2$ )). Due to the attached hydroxyl group, alcohols reduce the low-temperature combustion reactions, increase the ignition delays, reduce the combustion temperatures and soot emissions (Komninos and Rakopoulos, 2012). However, an increase in carbon monoxide (CO) emissions is observed with alcohol-based fuels due to carboxylation (Maurya et al., 2018).

Based on the above discussion, it is observed that the start of ignition depends on fuel reactivity and is driven by chemical kinetics. The saturated and unsaturated fatty esters in biodiesel are found to impact low-temperature reaction. The alcohols are found to decrease low-temperature combustion reactions, although it helps in improving atomization of diesel and biodiesel.

### 2.4. $\text{NO}_x$ -soot emissions

The emission formation in combustion depends on temperature, equivalence ratio and combustion chemistry. Fig. 4 shows the zone of  $\text{NO}_x$  and soot formation for combination of local temperature and equivalence ratio. The SI engine operates around an equivalence ratio of 1. SI engine operates near to  $\text{NO}_x$  emissions zone (Gan et al., 2011). The CI engine combustion is diffusion type giving the entire range of equivalence ratio. This puts local condition prone to both  $\text{NO}_x$  and soot emissions (Maurya et al., 2018). The HCCI engine operates at low equivalence ratio around  $\phi < 0.45$ . This also ensures low local temperature leading to very low  $\text{NO}_x$  and soot emissions. Low equivalence ratio in combustion with EGR further reduces the local temperature and hence  $\text{NO}_x$  formation. The adiabatic flame temperature curve is also shown in Fig. 4. The adiabatic flame temperature and equivalence ratio suggests the  $\text{NO}_x$ -soot free operating regime for engines operating in zone left of the adiabatic curve. The use of biodiesel in CI engine has shown an increase in  $\text{NO}_x$  emissions. The  $\text{NO}_x$  emissions decreases with increase in EGR for biodiesel-fueled pilot ignited HCCI engine (Zheng et al., 2006). The absence of aromatics content and the presence of oxygen in biodiesel reduces the soot, CO and hydrocarbon emissions (Subramanian, 2017). The biodiesel shows high CN and advances the combustion. Hence, to achieve premixed charge before the start of combustion, high levels of diluted charge is required that provides longer ID and avoids the  $\text{NO}_x$  formation.

The soot increases with deposition of fuel on combustion chamber walls during fuel injection (Maurya et al., 2018). A sufficient degree of premixed combustion results in negligible soot formation while an increase in charge stratification increases the soot formation (De Zilwa and Steeper, 2005). It is observed that the soot forms only in regions where OH radicals are absent (Yao et al., 2009). The decreased oxygen concentrations in the mixture are found to be responsible for soot formation (Liu et al., 2004).

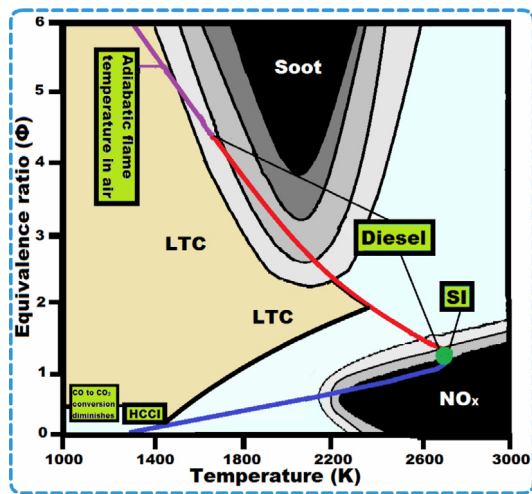


Fig. 4.  $\phi$ -T map for NO<sub>x</sub> and soot emissions in diesel combustion (Dec, 2009).

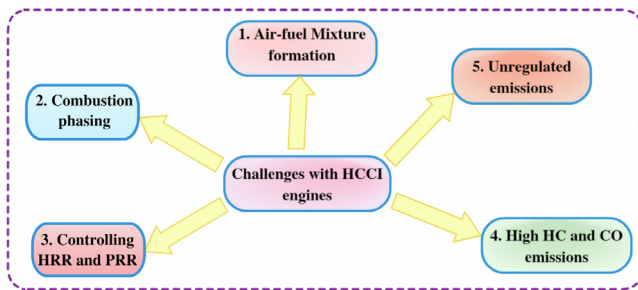


Fig. 5. Challenges with HCCI engines.

### 3. HCCI challenges

The actual power train application of HCCI engines is restricted by few challenges and needs to be solved before implementation. The challenges with HCCI engines are shown in Fig. 5.

#### 3.1. Mixture formation

A homogeneous charge preparation is challenging to achieve in HCCI engine over a range of operating condition due to a short time span and ambient conditions inside the combustion chamber. The charge formation can be achieved internally or externally by mixing fuel with intake air outside the combustion chamber. The Fig. 6 shows various methods used for air-fuel mixture formation in HCCI engine.

##### 3.1.1. External mixture preparation

It is the most effective and simple technique to achieve the premixed homogeneous charge with highly volatile fuels (Mau-rya et al., 2018). The volatile fuels such as gasoline, alcohols, dimethyl ethers (DME), acetylene, hydrogen, compressed natural gas (CNG), and biogas are introduced in the intake manifold. A fumigation technique can be utilized to vaporize the diesel and biodiesel fuels by heating at higher temperatures and mixing with the intake air to achieve the homogeneous charge (Agarwal et al., 2017). The low volatility of some fuels can lead to wall wetting (Pandey et al., 2018). The external mixture preparation is a simple method for homogeneous charge formation with volatile fuel. However, it provides little control over the start of ignition at various operating conditions.

##### 3.1.2. In-cylinder (internal) mixture preparation

Internal mixture preparation can be achieved by early and late direct injection of fuel inside the combustion chamber. The early fuel injection timing refers to fuel injection during early compression stroke while late injection means injection near top dead center (TDC) or at the end of compression stroke. The charge formation depends on fuel properties, injection timing, spray development, evaporation, and diffusion in compressed air (Baumgarten, 2006; Gupta and Agarwal, 2016). The early fuel injection during the compression stroke is limited by longer tip penetration and wall impingement which leads to an increase in HC, CO emissions and dilution of lubricating oil (Zhu et al., 2013; Lu et al., 2011; Yu et al., 2017). The use of split injection, and fuel preheating with significant EGR can improve the air-fuel mixing (Singh and Agarwal, 2012; Turkcan et al., 2018).

In the late injection strategy, fuel is injected close to TDC (3–5° after TDC) where ambient gas densities and temperatures are high. These ambient conditions enhance the fuel evaporation and diffusion leading to short ID. The in-cylinder mixture can give good control over HCCI combustion though it depends on various parameter which are difficult to control.

#### 3.2. Combustion phasing

The combustion phasing is critical in HCCI engine and occurs in a narrow crank angle window near TDC. The combustion phasing is characterized by CA50, the crank angle position where 50% of the total heat release occurs (Kalghatgi and Head, 2006). The combustion phasing in CI engines is controlled by the timing and quantity of fuel injected. However, in HCCI engines fuel is premixed and lean, hence the start of combustion is influenced by low-temperature chemistry of the fuel. The molecular structure of fuel, residence time, equivalence ratio, and temperature-pressure history of intake charge decide the rate of low-temperature oxidation reactions. The control on combustion timing is difficult in HCCI engine due to LTHR dependence on fuel chemistry and ambient conditions (Saxena and Bedoya, 2013). If the main heat release (2nd stage ignition) occurs too early during a compression stroke, combustion phasing is advanced, that may lead to an increase in the operating noise and knocking. On the contrary, delay in heat release by retarded ignition with lean charge increases the probability of misfiring and leads to very high HC emissions with reduced engine performance.

Ignition timing is advanced with high intake air temperature and pressure, which promotes the chemical kinetics (Hasan et al., 2018). The combustion timing advances for high reactivity fuels. The low reactive fuels have a lower specific heat ratio and require a higher temperature to start the auto-ignition. The variation in gross indicated mean effective pressure (IMEP), CA10 (crank angle position where 10% of the total heat release occurs) and engine misfire increases with highly retarded combustion phasing. It is important to maintain the combustion phasing between knocking and stable misfire limits for the operating range of the engine (Lawler et al., 2018).

#### 3.3. Heat release rate (HRR) and pressure rise rates (PRR)

The volumetric ignition of complete charge in HCCI engine leads to rapid HRR that may lead to knocking combustion. A lean air-fuel ratio with high EGR helps to reduce HRR with stable combustion. However, at high loads, combustion results in high HRR due to higher equivalence ratio. The high HRR limits the maximum BMEP of an HCCI engine (Olsson et al., 2001). The maximum brake mean effective pressure (BMEP) of around 18 bar is achieved for a light duty CI engine with turbo-charging. A naturally aspirated HCCI engine shows maximum indicated mean

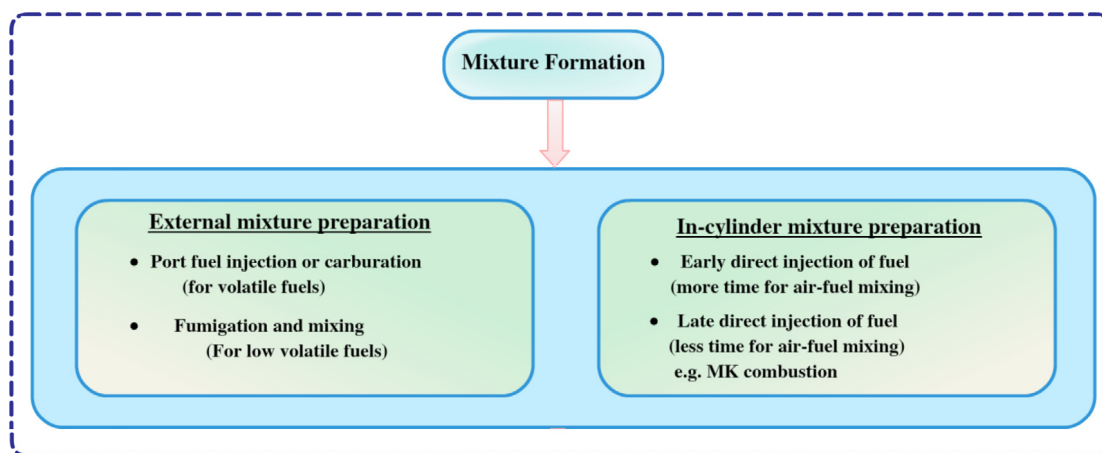


Fig. 6. Mixture formation in HCCI engines.

effective pressure (IMEP) of 5 bar for 1000–1500 RPM (Saxena and Bedoya, 2013). The addition of water in gasoline HCCI, extends the engine operating range from maximum allowable IMEP of 3.5 to 10 bar but requires a higher intake pressures (2.4 bar) at high load (Valero-Marco et al., 2018).

A high PRR causes acoustic resonance, knocking, and reduces the combustion duration. Thermal stratification within bulk gas governs the sequential auto-ignition and reduces the PRR at high load condition (Dec, 2009). A decrease in combustion temperature slows the chemical kinetic reactions and reduces the PRR. Olsson et al. (2001) reached up to 16 bar BMEP with dual fuel turbocharged engine at 1000 RPM and maximum efficiency up to 41.2%. The use of split injection in ethanol-gasoline fueled HCCI-DI engine shows a decrease in maximum pressure and heat release rate (Turkcan et al., 2018). A combination of fuel injection strategy with dual fuel is required to control HRR and PRR in HCCI engine.

### 3.4. HC and CO emissions

Reduced in-cylinder combustion temperature, spray impingement and cold cylinder wall temperatures lead to increased HC and CO emissions in HCCI engines. A longer burn duration leading to partial misfire and expansion quenches in the coldest zone increases the HC and CO emissions. The major source for HC formation is the top ring land crevice of the combustion chamber. The unburnt HC regions are observed along the centerline, squish volume and bowl/central clearance volume (Lu et al., 2011). The nozzle region is also found as a major source of unburnt HC emission (Aronsson et al., 2009). Due to lower combustion temperatures, CO is not completely oxidized to carbon dioxide (CO<sub>2</sub>), which requires temperatures above 1450 K. The CO emissions decrease with increase in premixing ratio, load and fuel–air ratio (Reitz and Duraisamy, 2015). The reduction in HC and CO emissions can be simultaneously achieved by increasing the inlet air temperature (Gowthaman and Sathiyagnanam, 2017) and compression ratio (CR) (Calam et al., 2019). Development of a low-temperature oxidation catalyst is required to control the tailpipe emissions in HCCI engine (Reitz and Duraisamy, 2015).

### 3.5. Unregulated emissions

The HCCI combustion is also reported to have an unregulated emission like aldehyde and ketones which are partially oxygenated organic compounds (Maurya et al., 2018). Aldehydes include Formaldehyde (HCHO) and Acetaldehyde (MeCHO).

Aldehydes are characterized by HC=O and ketones are characterized by RC=O. Aldehydes are carcinogenic and very reactive to form ozone by photochemical oxidation (O'riordan, 2014). Chain carrier radicals in the hydrocarbon combustion are alkyl radicals ( $\dot{R}$ ) and are formed by the cleavages of C–C or C–H bonds of hydrocarbon. The dehydrogenation by O<sub>2</sub> and the presence of other combustion radicals influence this process at high temperatures. Formaldehyde formation is most pronounced at low temperatures and acetaldehyde formation occurs at rich air–fuel mixtures (Wagner and Wyszynski, 1996). The formaldehyde emissions from aliphatic fuels are five times higher than aromatic fuels. Olefins exhibit highest aldehyde emissions while aromatics show lowest (Shore et al., 1993). The aldehyde emissions are found to decrease with the increase in fuel–air ratio and increase with the fuel CN. The oxygenated fuels produce higher aldehyde emission (Wagner and Wyszynski, 1996). The soybean biodiesel up to 30% shows a decrease in aldehyde and ketones in the exhaust than diesel (Lapuerta et al., 2008). The formaldehyde emissions decrease with increase in temperature, pressure and EGR while they increase with the increase in CR (Li et al., 2017). The low-temperature combustion with gasoline–n-butanol blend under the primary individual hydrocarbon emissions include the ethylene (C<sub>2</sub>H<sub>4</sub>), propene (C<sub>3</sub>H<sub>6</sub>), n-pentane (NC<sub>5</sub>), iso-pentane (IC<sub>5</sub>) and aromatic HC.

The carcinogenic and mutagenic action of acetylene (C<sub>2</sub>H<sub>2</sub>) affects the living beings. The polycyclic aromatic hydrocarbons (PAHs) species are found to be precursors for soot formation (Yang et al., 2014). The PAHs, carbonyl compounds, benzene, toluene, ethyl-benzene and xylene (BTEX) are harmful to human health and categorized as “possible carcinogens” (Srivastava et al., 2018). At rich and lean LTC operation NPAH (Nitro-PAH) emissions are much higher than diesel operation.

Based on the literature, it is observed that the formation mechanism, sources, and harmful effects of unregulated emissions from HCCI combustion have not been fully explored and understood yet. It is important to study these unregulated emissions from HCCI combustion and regulate them.

## 4. Fuel reactivity and stratification

The auto-ignition in HCCI engine depends on the fuel chemical kinetics and local equivalence ratio (Zhao, 2007). Single reactivity fuels are unable to control HRR and PRR for high load HCCI operation (Paykani et al., 2016). A low CN fuel retards the start of ignition and shows fast HTHR with negligible LTHR, which results in high cylinder pressure and NO<sub>x</sub> formation. An intake charge with high temperature is required to achieve the optimum

combustion phasing at higher loads for low CN fuel (Bunting et al., 2007). On the contrary, high CN fuels with high equivalence ratio lead to early LTHR and fast combustion. The LTHR rate is constant and it is not affected by the engine speed whereas the HTHR scales with engine speed and combustion duration remains constant (Bunting et al., 2007). Low engine speed favors more advancing in combustion phasing while high engine speed requires substantially decreased LTHR time scale.

A dual fuel reactivity can control the start of ignition and combustion phasing. Fuel reactivity stratification affects the ignition delay, ignition location and growth rate of reaction during the combustion. The fuels with different reactivity can control the global reactivity, hence HRR and combustion phasing (Dempsey et al., 2014). The addition of hydroperoxide in n-butanol, facilitates the formation of hydroxyl and hydroperoxyl radicals in the cylinder. It extends the engine operating range to lower load (with reduced HC and CO emissions) and improves the indicated thermal efficiency (Zhou et al., 2019). The addition of small amount of hydrogen in the mixture DME/CNG fueled HCCI engine, retards the ignition and expands the operating range within the knock limit (Bastawissi et al., 2019). At high load, biodiesel-hydrogen fueled HCCI engine retards the combustion phasing, lowers the peak HRR and reduces the  $\text{NO}_x$ -soot emissions (Khandal et al., 2019). In biogas-diesel fueled HCCI engine, with the increase in  $\text{CO}_2$  the HRR and  $\text{NO}_x$ -soot emissions decrease without affecting the thermal efficiency (Nathan et al., 2010). The LRF requires charge stratification, which indicates that ignition should occur first in an outer region of the combustion chamber. However, the temperature gradient suggests that ignition should occur in the center of the combustion chamber (Kokjohn et al., 2015). Flame propagation from high to low reactivity regions can reduce the PRR. The charge stratification with dual fuel in HCCI engine is obtained using port injected gasoline and direct injected diesel. Dual fuel HCCI shows the ability to operate at low EGR levels and controls combustion phasing during transient operations (Divekar et al., 2016).

The review of the research work on HCCI engine indicates that air-fuel mixture stratification and chemical kinetics of the fuel is important to control the combustion in HCCI engine. It is important to understand the air-fuel mixture formation and chemical kinetics experimentally. Various optical diagnostic techniques are used to study the mixture formation and combustion. The relevant techniques and studies are reviewed in the following section.

## 5. Optical diagnostics

The optical diagnostic is an effective tool to understand the physical and chemical processes in HCCI engines. Gasoline HCCI combustion process is first spectroscopically investigated by Noguchi in 1979 (Noguchi et al., 1979). They detected various intermediate chemical radicals significantly different from conventional gasoline combustion. The laser based diagnostic techniques used in combustion research are broadly categorized in Scattering, Chemiluminescence and Planar Laser Induced Fluorescence (PLIF). The scattering technique involves imaging, scattered light from particles (molecules, gas particles, liquid droplets). Rayleigh scattering, Mie scattering and Raman scattering is used in combustion research. Chemiluminescence involves imaging the light emitted by radicals or particular species. PLIF visualization involves excitation of particular species by laser and imaging their fluorescence.

### 5.1. Scattering

Diesel spray evolution in a wide and narrow-angle injector with multiple injections is determined using Mie scattering (Fang et al., 2009). The narrow-angle injector showed fuel impingement and a large amount of fuel deposition with ignition in central bowl region, higher soot luminosity and lower  $\text{NO}_x$  emissions (Fang et al., 2009). Mie scattering and LIF was used for spray development and fuel distribution measurement of swirl and eight hole gasoline direct injector (GDI) in low-load HCCI engine. The 8 hole injector small packets fuel vapor that lead to lower peak gas temperature and lower  $\text{NO}_x$  emissions (Steeper and De Zilwa, 2007). Musculus (2006) studied liquid fuel penetration, vapor jet penetration and ignition location using Mie scattering, fuel fluorescence and chemiluminescence imaging respectively. They observed an increase in liquid fuel penetration, OH species (throughout diesel jet cross section) and increased soot formation in the downstream head of jet.

### 5.2. Chemiluminescence imaging

The relaxation of the excited combustion radicals produce the chemiluminescence. The radicals such as OH, HCHO,  $\text{CO}_2$  exhibit the chemiluminescence in n-heptane flame (Yu et al., 2013). A strong radiation from the luminous flame in conventional diesel combustion overwhelms the weak chemiluminescence signal, which makes imaging of chemiluminescence difficult. A low soot emission from HCCI engine makes an analysis of chemiluminescence and spectral images relatively simple task (Dec and Espey, 1998). Chemiluminescence intensity and rate of heat release show a strong correlation (Augusta et al., 2006). The chemiluminescence images and spectral analysis of n-heptane and iso-octane mixtures show cool flames for HCCI combustion (Hultqvist et al., 1999). The spectral analysis of HCCI combustion with dimethyl ether showed the formaldehyde derived cool flames in LTHR (Kim et al., 2002). The study of n-heptane distribution using a high speed natural flame luminosity shows the increase in flame propagation and decrease in the combustion reactions with the rise in fuel inhomogeneity (Zheng et al., 2019). The HCCI combustion chemiluminescence images show the multiple locations of single auto-ignition of air-fuel mixtures. The inhomogeneity in temperature or air-fuel mixture fraction increases the probability of random combustion (Charalambides et al., 2018). It is observed that the engine parameters affect auto-ignition timings but not the HCCI reaction pathways.

A reduction in soot concentration is observed for rapeseed biodiesel at low fuel injection pressure using two-color pyrometry (Mancaruso and Vaglieco, 2010). Dec et al. (2006) used chemiluminescence imaging and reported lower PRR in optical HCCI engine by natural stratification in the combustion chamber. Chemiluminescence imaging in HCCI combustion showed stronger turbulence and lower PRR for square bowl piston compared to flat piston due to charge stratification (Vressner et al., 2007). The multiple injection, inlet air preheating and residual gas trapping under HCCI condition show large temperature inhomogeneities which slows the auto-ignition (Aleiferis et al., 2005). The local equivalence ratio variation by fuel injection timing increases the combustion duration and moderates the heat release (Berntson and Denbratt, 2007). Kumano et al. (2007) studied the effect of charge inhomogeneity on HCCI combustion in an optical engine using chemiluminescence. A large part in combustion chamber showed luminescence for a short time due to locally fast combustion under homogeneous condition. The luminescence is observed locally for a non-homogeneous condition with longer combustion duration, lower peak pressure and moderate heat release compared to homogeneous charge (Kumano et al., 2007).

Based on chemiluminescence imaging it can be observed that the fuel injection timing, EGR level, piston shape and intake air preheating can induce the charge stratification to extend the HCCI operating range.

### 5.3. Planar laser induced fluorescence (PLIF) imaging

The PLIF imaging can be used to image the distribution of various species during combustion. Richter et al. (2000) used the planar laser-induced fluorescence (PLIF) to study the degree of air–fuel mixture homogeneity obtained by port fuel injection and charge preheating. The distribution of HCHO is observed using PLIF imaging during cool flame of iso-octane and n-heptane blend (Collin et al., 2003). The HCHO is formed as an intermediate species during hydrocarbon oxidation at the low-temperature reaction. The early and late fuel injection respectively show more homogeneous and stratified charge distribution before combustion and good spatial resemblance in both cases (Särner et al., 2005). Charge temperature primarily affects HCHO distribution of primary reference fuel blends, while HCHO consumption is affected by its concentration and temperature (Zhao et al., 2004). The PLIF imaging of iso-octane fueled HCCI engine shows a significant production of HCHO during the cool flames and fast consumption during the second stage ignition results in steep pressure rise with short ID (Strozzi et al., 2019). The LIF measurements of a lean mixture in the squish region showed that the high wall heat transfer suppresses the lean mixture oxidation and increases the UHC and CO emissions (Ekoto et al., 2009). The major contributor to CO formation is observed to be squish volume. The source of HC formation is clearance volume stem regions near cylinder centerline and top ring land crevice (Kim et al., 2009). The unburnt HC and CO emissions decrease at high loads with an increase in intake pressure (Colban et al., 2007).

Kashdan and Papagni (2005) studied the PLIF images of HCHO, during early stage cool flame in late-injection diesel HCCI combustion. The combustion phasing is influenced by the ratio of the pilot and main injection fuel mass. The intensity of the PLIF signal from formaldehyde is more due to the homogeneous charge compared to uniform bulky combustion system (UNIBUS) and late injection HCCI (Hildingsson et al., 2005). Split injection increases HCHO lifetime and broader distribution of OH is found in HCCI combustion.

The NO-LIF is used to investigate the NO formation of diesel combustion. It is observed that the jet periphery just after diffusion flame and hot combustion gases are responsible for NO formation (Dec and Canaan, 1998; Jin and Zheng, 2015). De Zilwa and Steeper (2005) reported that each air–fuel packet in HCCI combustion burn as a homogeneous mixture and a strong correlation between in-cylinder charge distribution and emission formation is observed. Sufficient degree of premixed combustion results in negligible soot formation while an increase in charge stratification increases the soot formation levels (De Zilwa and Steeper, 2005). The soot formation detection by two-color pyrometry in LTC conditions showed lower temperatures than conventional combustion. The soot forms only in regions where OH radicals are absent (Yao et al., 2009). The decreased concentrations of oxygen in the mixture are found to be responsible for soot formation (Liu et al., 2004).

The liquid–vapor phase fuel distribution and intermediate combustion species are measured in an optically accessible HCCI engine using laser-induced exciplex fluorescence (LIEF) and chemiluminescence imaging respectively by Kashdan et al. (2004). The piston geometry shows an important role in mixture preparation, combustion and emissions characteristics in late injection strategy. The presence of intermediate species, formaldehyde as

auto-ignition precursor in early stages of cool flames was confirmed by LIEF. The flat piston showed strong PAH fluorescence signals as an indicator for the presence of soot.

The optical techniques are important to understand the mixture formation and combustion in HCCI engine. It is observed that the charge stratification, distribution of LRF and HRF inside the combustion chamber and ignition locations play an important role in heat release and emission formation. Their dependencies can be revealed only through optical diagnostics studies in HCCI engine. There are a few optical investigations in HCCI engine using biodiesels.

## 6. Conclusions

The review article recapitulates the current research in air–fuel mixture formation and combustion mechanism in HCCI engine in light of the use of biodiesels. Various approaches to control the ignition and charge stratification are discussed. Some of the techniques examined are intake air conditioning, fuel injection timing, degree of charge homogeneity, fuel chemistry, and EGR level to control the ignition and reaction timing. The oxygenated fuels, such as biodiesel, advance the combustion timing and exhibit the two-stage combustion. However, alcohol-based fuels are observed to retard the combustion timing and show single stage ignition. The chemical reactivity of the fuel is found to be responsible for the unique two-stage heat release characteristic. The OH and HCHO species formation and consumption decide the start of ignition and combustion duration. The HCCI combustion can achieve low NO<sub>x</sub> and soot emission with high thermal efficiency. However, due to the low combustion temperature, HC and CO emissions are high. The development of low-temperature oxidation catalyst is required to oxidize the HC and CO emissions from HCCI engines. The unregulated oxygenated and non-oxygenated hydrocarbon emissions are found to be high in HCCI combustion. There is an urgent need to address the problem of unregulated emissions from these engines.

The optical techniques are essential tools to understand the charge distribution and combustion inside the HCCI engine. The HCHO and OH radicals are suitable tracers at low and high-temperature reaction respectively. The EGR level influence the lifetime of HCHO and mixture homogeneity, and reduces the combustion phasing and high-temperature ignition. The partial charge or reactivity stratification achieved by low and high reactivity fuel provides better control over the start of ignition and combustion phasing for a transient and high load operation. The investigations on chemical kinetics for biodiesel combustion in HCCI engine are required for improving efficiency.

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