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In-cylinder Combustion and Soot Evolution in the Transition from Conventional CI mode to PPC

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Abstract:

The present study intends to explore the in-cylinder combustion and evolution of soot emission during the transition from conventional compression ignition (CI) combustion to partially premixed combustion (PPC) at low load conditions. In-cylinder combustion images and engine-out emissions were measured in an optical engine fueled with low octane heavy naphtha fuel (RON = 50). Full cycle engine simulations were performed using a three-dimensional computational fluid dynamics code CONVERGE™, coupled with gas phase chemical kinetics, turbulence, and particulate size mimic soot model. The simulations were performed under low load conditions (IMEP ~ 2 to 3 bar) at an engine speed of 1200 rpm. The start of injection (SOI) was advanced from late (-10 CAD aTDC) to early fuel injection timings (-40 CAD aTDC) to realize the combustion transition from CI combustion to PPC. The simulation results of combustion and emission are compared with the experimental results at both CI and PPC combustion modes. The results of the study show a typical low-temperature stratified lean combustion at PPC mode, while high-temperature spray-driven combustion is evident at CI mode. The in-cylinder small intermediates species such as acetylene (C₂H₂), propargyl (C₃H₃), cyclopentadienyl (C₅H₅) and polycyclic aromatic hydrocarbons (PAHs) were significantly suppressed at PPC mode. Nucleation reaction of PAHs collision contributed to main soot mass production. The distribution of soot mass and particle number density was consistent with the distribution of high-temperature zones at CI and PPC combustion modes.

Keywords

Low Octane Gasoline; Partially Premixed Combustion; Soot Particles;

Introduction

The conventional compression ignition (CI) engines are widely used due to the merits of higher compression ratio, zero throttling losses, and higher combustion efficiency when compared to spark ignition (SI) engines. However, the emissions of particulate matter (PM) and nitrogen oxides (NO_x) possess a major challenge. Therefore, after-treatment systems are needed to meet the stringent emission regulations [1], which are more expensive. Some advanced low temperature combustion (LTC) concepts have been proposed in order to reduce the emissions of PM and NO_x emissions. In conception, the LTC strategy has the obvious advantage of improved in-cylinder homogeneity and low in-cylinder combustion temperature, which mitigates the soot and NO_x emissions. Improvement in efficiency and fuel consumption are the added advantage for the LTC combustion concepts.

Homogeneous charge compression ignition (HCCI) is an ideal and promising LTC strategy, which can achieve a homogeneous mixing of fuel and air at early fuel injection timing. Dec et al. [2] presented a comprehensive overview of HCCI technology and claimed that fuels such as gasoline, diesel, and bio-fuels could be operated in HCCI mode. Hyvönen et al. [3] demonstrated the spark assisted HCCI combustion in a multi-cylinder variable compression ratio (VCR)-HCCI engine and pointed out that intake air temperature and compression ratios can be used as actuators to control combustion. Bessonette et al. [4] studied the effect of fuel properties on HCCI combustion in a heavy-duty (HD) engine. The results showed that HCCI engine had diesel-like higher thermal efficiency because of the shorter combustion duration and lower heat transfer losses. Furthermore, the emissions of PM and NO_x were suppressed due to lean combustion at LTC operating condition. However, there are still many challenges with HCCI combustion that are required to be solved. First, controlling combustion (combustion phasing) is a challenge due to multiple auto-ignitions. Second, HCCI is a closer to constant volume combustion that can result in a higher heat release rate at short durations. The resultant rapid pressure rise rate limits the load expansion and operating limit for HCCI engines. Third, HCCI combustion has different requirements for fuel reactivity at different engine load operating conditions. High reactivity fuels are required at low load while low reactivity fuels are required at high load [4].

To solve the limitations of HCCI, partially premixed combustion (PPC) is proposed, which is an intermediate between CI and HCCI combustion. PPC adopts flexible direct injection strategy along with fast thermal management (FTM) of intake condition in which the fuel/air mixture preparation, combustion phasing, rate of heat release (RoHR) and rate of pressure rise (RoPR) are closely coupled to the start of injection (SOI). [5, 6, 7]. In general, the SOI for PPC is later than that of HCCI but earlier when compared to CI combustion so that premixing time is prolonged. Johansson et al. [5] investigated diesel PPC in a heavy duty (HD) engine under different engine operating conditions. The results showed that NO_x and PM emissions could be simultaneously reduced when the engine was operated at lower compression ratio and higher EGR rate closer to stoichiometric condition at loads of 8bar, 12bar, and 15bar IMEP gross. However, there was a decline in combustion efficiency and an increase of unburned hydrocarbons (UHC) and carbon monoxide (CO) emissions during the conventional NO_x-soot trade-off departure. Kalghatgi et al. [9, 10] investigated PPC operation in a CI engine fueled with diesel and gasoline fuels. The results showed that

gasoline-fueled tests achieved lower PM and NO_x emissions as compared to diesel. Furthermore, the optimum research octane number (RON) for gasoline fuel was found to be between 75 and 85 for their experimental operating conditions. Similarly, most of the other studies report that low RON gasoline is more suitable for operation in CI engine under PPC mode when compared to high RON gasoline as the latter is prone to auto-ignition problems at low load condition when compared to the former. Naphtha, a low RON fuel in the gasoline boiling range of 30-180°C, has shown good potential for widening PPC operating range [11, 12, 13, 14]. The authors have previously carried out experiments in an optical diesel engine and found that these low RON gasoline-like fuels have a significant potential for clean and efficient PPC operations [15].

Many studies [16-22] were performed to understand the in-cylinder soot particle formation and oxidation. Schraml et al. [21] investigated the in-cylinder soot formation and oxidation process by planar laser-induced incandescence of soot (soot-PLII) and found that the initial soot appeared at 2 CAD aTDC. They reported the maximum intensity around 6~8 CAD aTDC and subsequently, the intensity was reduced to zero at 26 CAD aTDC and later. Bobba and Musculus [22] used soot-PLII and planar laser-induced fluorescence of polycyclic aromatic hydrocarbons (PAHs-PLIF) to investigate the in-cylinder distribution of PAHs and soot in an HD diesel engine under LTC operation. In particular, the effect of fuel-bound aromatic components on the soot formation was analyzed. Compared to the representative diesel fuel with 27% aromatics, a non-aromatic fuel showed an increased residence time of PAHs before the initial soot formation. Nevertheless, the two fuels yielded nearly identical spatial and temporal evolution of PAHs-PLIF and soot-PLII at LTC conditions.

Computational fluid dynamics (CFD) simulations coupled with gas phase chemical kinetics complements experimental measurements by providing quantitative information on fuel oxidation and emission formation processes, especially for the evolution of soot particles [16, 23-26]. Appel et al. [16] investigated the soot formation with detailed chemistry in laminar-premixed flames of C₂ hydrocarbon. Kawatani et al. [27] found that soot particulate makes up to approximately 60% of PM. Frenklach [28] developed a soot model that includes four major steps: nucleation of soot particles through gas-phase species, particle coagulation, particle surface reactions (surface growth and oxidation) and particle agglomeration. Hu and Pang et al. [29-31] developed two reduced chemical kinetic mechanisms of primary reference fuel and toluene reference fuel to simulate the in-cylinder combustion and soot particle formation. These mechanisms were validated for low and high temperature reactions with engine combustion data at HCCI and CI conditions. Although injecting fuel at high injection pressure and multiple fuel injection strategies under LTC conditions can reduce the soot mass concentration, the particle number (PN) is difficult to be controlled below the limit of EURO VI (PN < 6x10¹¹ particles/km) [32-35]. Such nanoparticles with a size smaller than 300 nm are known to be a health hazard [36-38].

Based on the above discussion, it is clear that a more detailed analysis of chemical pathways is needed to explain the evolution of small intermediate species of soot. Furthermore, quantitative prediction of soot mass and size distribution during the in-cylinder combustion process is of paramount importance in engine studies. Therefore, in the present study, the evolution of small key intermediate species, PAHs, soot mass, and particle number density during the combustion transition from conventional CI combustion to PPC is investigated for heavy naphtha fuel (RON =50). An updated gas-phase PAHs chemistry mechanism is integrated into the CFD engine model to build the soot model. AVL optical diesel engine is used to obtain the experimental history of in-cylinder pressure, RoHR, and levels of engine-out emissions. High-speed combustion images are used to evaluate the calculated in-cylinder combustion characteristics. The simulation results are compared with the engine experimental results to obtain better insights into the transition in combustion mixture homogeneity and soot particle evolution from CI to PPC condition.

2. Experimental setup and measurements

A single-cylinder AVL optical diesel engine was used for the experimental study, which was operated at CI and PPC conditions. Figure 1 shows a schematic diagram of the experimental engine setup. The engine was operated at a low speed (1200 rpm) and low load condition (IMEP = 2 to 3 bar) with a compression ratio of 9.5, considering the safety and operational limitation of the optical engine. Other specifications of the engine are listed in Table. 1. The piston bowl was made of quartz and the in-cylinder combustion process was captured from the bottom through a mirror. Photron FASTCAM SA4 high-speed color camera was used to capture the combustion process. The high-speed combustion video was recorded at the rate of 10000 frames/s with a pixel resolution of 768 x 512 and aperture of 1.4. A total of 150 continuous cycles were recorded with the trigger for camera set at -20 CAD aTDC. The injection timing, injection pressure, and the injection pulse (duration) were controlled by the AVL electronic control unit (ECU). AVL piezoelectric pressure transducer with a sensitivity of 20pC/bar was used to record the in-cylinder pressure, which was installed in the cylinder head. The cylinder pressure data were recorded in 0.2 crank-angle increments (sampling frequency of 36 kHz). AVL AMA i60 emission analyzer measured the engine-out exhaust emissions such as CO, CO₂, and NO_x. The soot mass concentration was recorded by the AVL Micro Soot Sensor, which had high sensitivity (resolution 0.01 µg/m³, detection limit 1 µg/m³) and wide measuring range (1-1000 mg/m³). ETAS LA4 lambda meter records the lambda data and the engine was operated at lean condition with a global operating equivalence ratio of 0.2.

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Figure 1. (a) Schematic diagram of the engine setup

(b) optical piston bowl

Table 1. Single cylinder optical diesel engine Specifications

Bore	85mm	
Stroke	90mm	
Compression ratio	9.5	
Nozzle diameter	0.18mm	
Injector holes	7	
Injection angle	145°	
Intake Valve	Open	30 CAD bTDC
	Close	45 CAD aBDC
Exhaust Valve	Open	50 CAD bBDC
	Close	25 CAD aTDC

Table 2. The experiment conditions

Engine speed	1200rpm	
Injection pressure	800bar	
Injection timing	- 10 ~ - 40 CAD aTDC	
Intake temperature	90°	
Intake air pressure	1.5 bar	
Coolant temperature	80°	
Lubricant temperature	80°	
Global equivalence ratio	0.2	

Table 3. Fuel Properties

Property	Heavy Naphtha [15]	PRF50 [15]
Density (kg/m ³)	760	687
LHV (MJ/kg)	44.9	44.43
Viscosity (m ² /s)	0.4	0.45
IBP (°C)	40	99
RON	46	50
H/C ratio	2.15	2.27
Derived Cetane number	41	40.1

The test conditions are shown in Table 2. The fuel injection timing was varied from -10 CAD (aTDC) to -40 CAD (aTDC) to study the transition in combustion from CI to PPC mode. The fuel at ambient temperature of 23° was injected at a pressure of 800 bar. Intake temperature and pressure were raised to compensate for the lower compression ratio. Further, the intake air was boosted so that the pressure at TDC was maintained at 35 bar under the motoring operation condition as reported in previous

studies [15]. The reported intake air temperature was chosen in such a way that the coefficient of variance was below 5% at all fuel injection timings.

Throughout the experimentation, the FuelMEP was kept constant at 5.1 bar and estimated as follows.

$$\text{FuelMEP} = \frac{M_f \cdot Q_{LHV}}{V_d} \quad (1)$$

where, M_f is the injected fuel mass (mg), Q_{LHV} is the lower heating value, and V_d is the displacement volume in m^3 . For the CFD simulations, the physical properties of heavy naphtha were incorporated to the code and the primary reference fuel, PRF50 (50% iso-octane with 50% n-heptane by volume), was selected as a surrogate to represent the chemical behavior of the naphtha fuel. The physical and chemical properties of naphtha and PRF50 are shown in Table 3.

3. Model description

3.1 Numerical setup

The three-dimensional CFD code (CONVERGETM) is widely recognized for engine simulations [39-45], which is used to simulate the in-cylinder combustion process and soot particle evolution in the present study. The full engine geometry developed for CFD study is shown in Figure 2 (a) and the mesh-generated in-situ is shown in Figure 2 (b). The initial base grid was set to 4mm in all three directions. The local grid was refined by adaptive mesh refinement (AMR) technique according to the absolute value of temperature gradient (5K) between the neighboring cells. The AMR of level 5 (minimum cell size = $\text{dx}/2^5$) refined the cells to a minimum size of 0.125 mm to capture the events with higher resolution. The renormalized group (RNG) k-epsilon model [46] was used to describe the in-cylinder turbulent flow field with Reynolds averaged Navier Stokes (RANS) approach. The “blob” injection model [47] and modified Kelvin-Helmholtz/Rayleigh-Taylor (KH-RT) model [48] were used for the fuel injection and spray breakup. The droplet vaporization was modeled by the multi-component evaporation model [49] through mass conservation equations. The effect of recirculation inside the droplets was accounted by effective thermal conductivity model [50]. A square rate shape injection profile was used during the fuel injection and the spray model used here had been validated before for ECN spray A conditions [51] and was not shown here for brevity. The spray model constants used are included in the Appendix Table A1. The chemistry solver, SAGE [52], calculated the reaction rates for each elementary reaction based on the detailed reaction mechanism, while CFD solved the transport equations. A multi-zone model based on well-stirred reactor model was used to accelerate the calculation, which divided the cells for 0.05 ϕ and 5 K, respectively [53]. More details about the models used can be found in the CONVERGE manual [53].

Figure 2. (a) Three-dimensional model of AVL optical CI engine (b) Geometric mesh of the computational domain

3.2 Chemical kinetic mechanism

Previous studies have pointed out that soot particles are derived from the important soot precursors such as small olefins and alkynes molecules and PAHs that are generated from the pyrolysis of the injected fuel during the combustion process [29-31, 43-45, 54-56]. Therefore, the accurate prediction of gas-phase pollutants and soot is strongly dependent on the reliability of the chemical kinetic mechanism. The author built a reduced gasoline fuel mechanism including PAHs formation/oxidation reactions that contain 85 species and 232 element reactions [43, 44, 57, 58]. This chemical kinetic mechanism contains low-temperature reactions, high-temperature reactions and PAH reactions.

The low-temperature reactions used in the current mechanism were derived from Andrae [59] model, Tsurushima [60] model and Jia [31, 61] model as described in blue box of Figure 3 (a). It primarily describes the reaction process of the two kinds of paraffin such as n-heptane and iso-octane molecules undergo the first O_2 addition reactions, isomerization reactions, and the second O_2 addition reactions to form the hydroperoxy alkyl peroxy radicals. These intermediate species then break down into ketohydroperoxide (KETO) and hydroxyl radicals. It should be noted that the small olefins such as ethylene and propylene, and aldehydes such as formaldehyde and acetaldehyde are the important intermediate species during the low-temperature process. Therefore, the KETO is broken down into C_5 or C_6 alkyl radicals (R' denotes C_5 or C_6 alkyl radicals) as shown in Figure 3 (a)

according to Tsurushima [59] mechanism. Then these alkyl radicals are broken down into small olefins of propylene and ethylene, and also into aldehydes represented by CH₂O.

The high-temperature process strongly relates to the high-temperature heat release that mainly contains the H₂/CO chemistry and C₁ and C₂ chemistry [31]. This chemistry describes the beta-scission reactions and the oxidation of the small alkyl radicals, as shown in the red box of Figure 3.

Figure 3. Major reaction branches of n-heptane/iso-octane

Figure 4 shows the PAHs sub-mechanism that describes the reactions for molecules beyond benzene (A1) up to four rings (A4). Benzene is the initial PAH that is critical to the subsequent PAHs growth process through H-abstraction and C₂H₂/C₄H₄-addition (HACA). R1-R3 play an important role in A1 formation for temperature below 1500K, while R4 becomes dominant in A1 formation for temperatures over 1500K [63, 64].

Previous studies [62, 65-68] found that acenaphthylene (A2R5, C₁₂H₈) and indenyl (C₉H₇) play an important role in bigger PAHs formation. R5-R8 were added into the current mechanism used in the study.

Figure 4. Major reaction branches for formation of PAHs [44, 58]

Choi and Chung [69] found the synergistic effect on PAHs and soot formation in the counterflow diffusion flames for gasoline surrogate fuels. After that, Yu et al. [70] investigated this synergistic effect and defined the reaction pathways. Therefore, these synergistic reactions were added to the current mechanism, as shown in Table 4. The current mechanism was validated for the

low-temperature characteristics of ignition delays for n-heptane and iso-octane in a shock tube [71-75]; as well as, for the high-temperature characteristics of laminar flame speeds for the same fuels in a flat flame adiabatic burner [76-78]. In addition, this mechanism was also validated for small intermediate species (acetylene, ethylene, CO, CO₂ and H₂O) and PAHs mole fractions in iso-octane and n-heptane flames respectively [79, 80]. More details about the current mechanism can be found in the literature [43, 44, 58].

Table 4 Rate coefficient for Reactions in Arrhenius form $k=AT^n\exp(-E/RT)$

No	Reactions	A(cm ³ /mol/s)	n	Ea(cal/mol)	Ref
R9	$C_6H_5CH_3+H=C_6H_5CH_2+H_2$	6.47E+00	4	3394	68, 69
R10	$C_6H_5CH_3+H=A1+CH_3$	1.20E+13	0	5100	68, 69
R11	$A2CH_3+H=A2+CH_3$	3.85E+06	2.2	4163.5	68, 69
R12	$C_6H_5CH_2+C_3H_3=C_{10}H_{10}$	1.51E+75	-17.9	39600	68, 69
R13	$C_{10}H_{10}+H=C_{10}H_9+H_2$	1.00E+13	0	0	68, 69
R14	$C_{10}H_9+H=A2+H_2$	1.00E+14	0	0	68, 69
R15	$P2- +C_2H_2=A3+H$	4.60E+06	2	7300	68, 69
R16	$A3-4 + C_2H_2 = A4 + H$	1.40E+29	-3.36	17800	68, 69
R17	$C_6H_5CH_2+C_9H_7= A4+2H_2$	2.00E+13	0	2000	68, 69

3.3 The detailed soot model

The particulate size mimic model (PSM) [81, 82] based on the discrete sectional method was used to describe the soot particle size distribution function (PSDF). In the PSM, each particle size section is bounded by its minimum and maximum volume, $v_{i,min}$ and $v_{i,max}$, respectively:

$$v_{1,min} = v_{MIN} \quad (2)$$

$$v_{i,min} = v_{i-1,max}, \text{ for } i>1 \quad (3)$$

$$v_{i,mean} = \frac{v_{i,min}+v_{i,max}}{2} \quad (4)$$

v_{MIN} is the volume of the smallest soot particle, defined as the volume of the soot precursor (0.4e-27 m³ for pyrene, A4, in the current study), v_{MAX} is defined as the volume of the biggest soot particles (approximately 100 nm in diameter). The total volume fraction Q_i for each section will be:

$$Q_i = \int_{v_{i,min}}^{v_{i,max}} q_i(v) dv \quad (5)$$

where q_i is the distribution function of v for each section. The distribution function $q_i(v)$ for v is presented in the first-order polynomial:

$$q_i(v) = q'_i v + q''_i \quad (6)$$

$$q'_i = \frac{k_i^+ + k_i^-}{2} \quad (7)$$

Where k_i^+ and k_i^- are the slopes of the left and right boundaries of $q_i(v)$ respectively. These values are calculated so that the soot streams transported through different sections are conserved. q''_i is then calculated as:

$$q''_i = \frac{Q_i}{v_{i,min}-v_{i,max}} - q'_i v_{i,mean} \quad (8)$$

The maximum boundary is increased using the following non-linear formulation to obtain high computation efficiency.

$$v_{1,MAX} = v_{MAX} + v_{C2} \quad (9)$$

$$v_{i,MAX} = (v_{MAX} + v_{C2}) \left(\frac{v_{MAX}}{v_{MIN} + v_{C2}} \right)^{i-1} \quad (10)$$

The current soot model is derived based on the detailed soot model developed by Frenklach and Wang [28, 83] as shown in Figure 5. Pyrene (A4) is used as the soot precursor and leads to soot inception. Subsequently, soot particles grow into the final mature particle through the steps of PAHs condensation, soot surface growth by C₂H₂, particle coagulation, and soot surface oxidation processes via oxygen (O₂) and hydroxyl radicals (OH) [45].

Figure 5. Soot particle model with PAHs as soot precursors [45, 83].

The soot inception is based on Smoluchowski equation [84] with PAH species (pyrene, A4 is used in the current model) as:

$$\dot{Q}_{i,pi} = 2v_{PAH}\beta_{fm,pi}(v_{PAH})N_{PAH}^2 \quad (11)$$

v_{PAH} is the volume of the PAH species, $\beta_{fm,pi}$ is the collision coefficient for the PAH species, and N_{PAH} is the number density of the PAH species. It should be noted that nucleation is regarded as the first section, while the source term for other sections is zero. Soot condensation mainly describes the coagulation process of PAH species on the soot particle surface, which leads to form larger soot particles. The condensation source term is calculated as follows:

$$\Delta Q_{i,cond} = v_{PAH}N_{PAH} \int_{v_{i,min}}^{v_{i,max}} \beta_{fm,cond}(v_{PAH},v)n(v)dv \quad (12)$$

The soot surface reaction is modeled by the widely-used description is the hydrogen abstraction acetylene addition (HACA) mechanism [85]. The reaction pathways of the HACA mechanism are given below:

In the above, $C_{soot,i}H$ represents an active site on the soot particle surface with a terminal H-C bond, and $C_{soot,i}^*$ represents the active soot particle after the dehydrogenation reactions. The soot surface growth process is described by reactions (1-3) and the soot oxidation is described by reactions (4-5). The surface growth rate $\Delta Q_{i,sg}$ and oxidation rate $\Delta Q_{i,ox}$ for each section can be formulated as [86]:

$$\Delta Q_{i,sg} = \alpha v_{c2}^{\frac{3-\theta}{3}} (k_d - k_{rev}) \left(\frac{3}{3+\theta} q_i \left(v_{i,max}^{\frac{3+\theta}{3}} - v_{i,min}^{\frac{3+\theta}{3}} \right) + \frac{3}{\theta} q_i^u \left(v_{i,max}^{\frac{\theta}{3}} - v_{i,min}^{\frac{\theta}{3}} \right) \right) \quad (13)$$

$$\Delta Q_{i,ox} = \alpha v_{c2}^{\frac{3-\theta}{3}} (k_{O2} - k_{OH}) \left(\frac{3}{3+\theta} q_i \left(v_{i,max}^{\frac{3+\theta}{3}} - v_{i,min}^{\frac{3+\theta}{3}} \right) + \frac{3}{\theta} q_i^u \left(v_{i,max}^{\frac{\theta}{3}} - v_{i,min}^{\frac{\theta}{3}} \right) \right) \quad (14)$$

Where θ is the fractional dimension of the soot, α is the fraction of active sites on soot particle surface and k is the reaction rate coefficient.

The physical collision of small soot particles can result in formation of larger soot particles, which is described by the soot particle coagulation and is calculated as [86]:

$$\Delta Q_{i,coag} = \sum_{v_{i,min} < (v_k + v_j) < v_{i,max}} (v_k + v_j) N_k N_j \beta_{coag}(v_k, v_j) + \sum_{(v_k + v_j) < v_{i,max}} v_k N_k N_i \beta_{coag}(v_k, v_i) - v_i N_i \sum_{(v_k + v_j) > v_{i,max}} N_i \beta_{cond}(v_i, v_j) - 2v_i N_i N_i \beta_{coag}(v_i, v_i) \quad (15)$$

Each section in the PSM model is solved as a global transport passive:

$$\frac{D(\dot{Q}_i/\rho)}{Dt} = \Delta \left(\frac{\mu}{SC} \Delta \left(\frac{\dot{Q}_i}{\rho} \right) \right) + \dot{S}_{Qi} \quad (16)$$

$$\dot{S}_{Qi} = \dot{Q}_{i,pi} + \Delta Q_{i,cond} + \Delta Q_{i,sg} + \Delta Q_{i,ox} + \Delta Q_{i,coag} \quad (17)$$

Where, SC is the Schmidt number and $\dot{S}_{Qi}$ represents the source term for each section. The section source term is coupled with the species source term and solved using SAGE solver.

4. Result and discussion

4.1 Full cycle engine model validation

The CI and PPC modes were differentiated by the change in slope of CA50 (crank angle at which 50% of total heat is released) with respect to SOI. As shown in Figure 6, CI mode shows a positive heat release whereas a negative slope is witnessed at PPC mode. The transition from positive to negative slope happens at -25 CAD aTDC, which represents the transition point in combustion regime from CI combustion to PPC. Notably, it takes a dip at -35 CAD aTDC before it starts to increase again. The observed pattern in CA50 suggests decoupling of combustion phasing from SOI at early injection timings. Thus, SOI is sensitive to ignition at late fuel injection timing and not at early fuel injection timings. The comparison of CA50 between the experiments and simulations are satisfactory and the trends are well reproduced by the simulations. CA10 (crank angle at which 10% of total heat is released) is regarded as the start of combustion in the present study. Similar to CA50, the start of combustion also matched between the experiment and simulation.

Figure 6. Comparison of derivative of CA50 with SOI, CA10 and CA50 between experiments and simulations at different SOIs.

The predicted in-cylinder pressure and RoHR by the three-dimensional CFD model are compared with the averaged experimental pressure data under CI and PPC conditions. The shaded region (Figure 7) indicates the minimum and maximum pressure range obtained from the experiment for 150 cycles. The peak in-cylinder pressure and RoHR at PPC mode are lower when compared to CI mode, given the indicated mean effective pressure (IMEP) and combustion efficiency are decreased as SOI is advanced for the same compression ratio of the engine. As shown in Figure 7, the in-cylinder pressure and ROHR predicted by the present simulation methodology have reasonable agreement with the measurement results at CI and PPC modes. There are two peaks in the measured RoHR trace, where the first small peak indicates the low-temperature reaction (LTR) region in the early stage of combustion and the second higher peak corresponds to the high-temperature reaction (HTR) region. Although the simulated peak RoHR is over-predicted, the experimental behavior of the two stages combustion has been captured well in the simulations. The level of the above agreement justifies the use of the current simulation approach and models to reflect the in-cylinder combustion process. The over-prediction of heat release indicates the need for further optimization of H_2-O_2 chemistry and C_1 and C_2 chemistry. Mehl et al. [87] noted that the flame propagation was mostly affected by the reactions of H/OH radical system. Therefore, the current mechanism needs further refinement as reported above so that the high-temperature heat release could be predicted accurately.

Figure 7. In-cylinder pressure (P) trace and rate of heat release (RoHR) validation under CI and PPC conditions.

The comparison of the measured engine-out emissions and the predicted results from the simulation is shown in Figure 8. Initially, as the SOI is advanced to -15 CAD aTDC, the emissions of NO_x and soot are increased while CO emission is decreased. This is mainly due to the change in combustion behavior. The fuel spray is directed towards the combustion bowl and a rich mixture is burnt when compared to the well premixed mixture at PPC mode. The in-cylinder rich mixture and the subsequent increase in in-cylinder temperature (as shown in the later section) increases the soot and NO_x emission. The in-cylinder maximum temperature is shown in Appendix. It can be seen that the maximum temperature is noted at SOI of -15 and -20 (CI mode). However, at early fuel injection timings, the maximal temperature is decreased due to premixed effect and low temperature combustion. Thus, when the SOI is further advanced to -25 CAD aTDC, there is a sharp reduction in NO_x emission and a slight increase in CO emission due to lower in-cylinder gas temperature. The CO_2 and soot emissions are decreased because of the improved air/fuel mixing. At SOI = -40 CAD aTDC, CO_2 and soot emissions are further decreased as the in-cylinder air/fuel mixture becomes more premixed to reduce the overall fuel to air equivalence ratio (section 4.2). On the other hand, NO_x emission decreases due to low-temperature combustion effect. Overall, soot, NO_x , and CO_2 emissions are decreased and CO emission is increased as the injection is advanced towards PPC. It could be observed that the predicted trends of CO, CO_2 , NO_x , and soot emissions are consistent with the experimental measurements at CI and PPC conditions. The CO emissions are over-predicted in the SOI range of -35 to -40 CAD aTDC; CO_2 emissions are well reproduced at various injection timings. The NO_x emissions are well reproduced except at the injection timing of -15 CAD aTDC while the predicted soot emissions are consistent with the experimental results.

Figure 8. Comparison between experimental and simulated engine-out emissions for CI and PPC conditions

The combustion efficiency and maximum in-cylinder temperature are compared with the simulation results and shown in Figure 9. The simulated combustion efficiency is slightly higher but has the similar trend to the experimental results. As the SOI is advanced from CI to PPC mode, the combustion efficiency gradually drops for both simulation and experiments as IMEP is decreased for the same FuelMEP. This explains the reason for decreased RoHR and increased CO emissions. The increased HC and CO emissions for PPC mode due to decreased combustion efficiency have been reported in literatures before [5, 6, 15, 18]. The maximum in-cylinder temperature at various fuel injection timings is shown in below Figure 9b. It can be seen that the

maximum in-cylinder temperature achieves peak values at late SOI of -15 and -20 (CI mode). The maximum temperature inside the cylinder is higher, which supports the formation of the soot and NO_x emissions. However, at early fuel injection timings, the maximum in-cylinder temperature is decreased due to premixed effect by which the formation of NO_x and soot can be effectively suppressed.

Figure 9. Combustion efficiency and the in-cylinder maximum temperature for CI and PPC conditions

To obtain further insight into the in-cylinder combustion process, the captured high-speed combustion images from the optical engine experiments are compared with the simulation results, as shown in Figure 10. The experimentally captured images and the simulation results are analyzed at the combustion phasing of CA50. By this comparison, the combustion behavior between the CI and PPC combustion is understood. In the experimental combustion image, the dotted white circular line indicates the boundary of the piston bowl. The blue color in images is derived from the blue channel of the high-speed color camera (FASTCAM SA4), which has a peak spectral wavelength range of 440 ~ 500 nm. Here, the natural luminosity of the images is correlated with the mixture homogeneity. As SOI is advanced to early fuel injection timings, the intensity of the images decreases. The weak luminous images at early fuel injection timings indicate enhanced premixed effect towards low temperature combustion. The simulations were able to reproduce the experimental trend qualitatively. Isolated combustion clouds are observed in CI mode, which moves towards the center of the piston, as the injection is advanced. A typical spray driven combustion is obvious at CI mode in both experimental and simulation results. In PPC mode, the fuel is injected into the squish zone and combustion is observed in the squish region. This is not evident in the experimental combustion image because the optical access for the squish region is not available, given a full optical piston is not used in the present study.

Figure 10. Comparison of high-speed color images and simulated in-cylinder temperature contour under CI and PPC conditions

4.2 Prediction of in-cylinder combustion and soot particulates

4.2.1 Temperature-equivalence ratio ($T-\phi$) distribution

Figure 11 presents the in-cylinder local distribution of temperature and equivalence ratio (ϕ) at CA50 and CA90 (the crank angle when 50% and 90% of fuel energy released) at different fuel injection timings. In CI mode (SOI of -10 CAD aTDC), the in-cylinder temperature mainly distributes in the piston bowl and shows seven independent combustion clouds. These results are

similar to the experimental data of Dec et al. [91], indicating the spray-driven combustion at CI conditions. The high-temperature core zones are over 2500K, which is consistent with the typical temperature range for rich mixture zones. At CA90, the in-cylinder high-temperature zones are distributed close to the piston bowl boundary and decrease to ~2200K. Similar to in-cylinder temperature, equivalence ratio also distributes at the outer periphery of piston bowl.

As the SOI is advanced to -20 CAD aTDC, the in-cylinder combustion clouds are still distributed in the piston bowl but moves towards the center of the piston and the high-temperature core zones are reduced to around 2500 K at CA50. The distribution of ϕ shows less than seven clouds in the rich mixture zones, which represents spray-driven combustion. At CA90, the in-cylinder high-temperature zones transit further to the center of piston bowl with the reduced temperature of around 2200K. Here, the distribution of ϕ is less than 1 as most of the fuel/air mixture is burnt out.

Further advancing the SOI to PPC condition (SOI = -40 CAD aTDC), the in-cylinder temperature is reduced to 2000K. The combustion clouds are observed in the crevice as the early-injected fuel spray entrains into the top-land of the piston. Slightly rich fuel/air mixture distributes in the squish while the lean mixture distributes in the piston bowl region. When combustion progresses towards CA90, the in-cylinder high-temperature zones are reduced to below 2000 K and still distributed in the crevice. From the temperature and equivalence ratio distribution, it can be inferred that the typical low temperature stratified combustion occurs at early fuel injection timing. These findings clarify the experimental results of reduced NO_x and soot emissions at PPC mode, as discussed above.

Figure 11. Volume contours of temperature and equivalence ratio (ϕ) at different crank angle during combustion under CI and PPC conditions

4.2.2 In-cylinder soot evolution

Figure 12 shows the predicted in-cylinder small intermediate species and A4 history at CI and PPC conditions. As SOI is advanced to early fuel injection timing (PPC condition), the premixing process of fuel and air is enhanced and the in-cylinder temperature is reduced. As a result, the mass of intermediates (C_2H_2 , C_3H_3 , and C_5H_5) is lower at PPC mode when compared to CI mode. This results in decreased formation of soot precursors (A4) as the reaction pathways (R1~R8) are suppressed [62, 63] (Figure 4). In addition, a lower species mass is produced with the increase of carbon atoms during the combustion process. The formation of these intermediates species at the transition point from CI to PPC (-25 CAD aTDC) occurs earlier when compared to late fuel injection timings (CI mode). Regardless of the advancement of fuel injection timing after the transition zone, the formation of the intermediate species is not advanced and occurs at the same location as that of the transition zone. This gives a notion that fuel spray is directed towards the squish region above the piston top land at early fuel injection timings.

Figure 12. The mass variation of key intermediate species related to PAHs formation at various injection timings.

Figure 13 (a) and 13(b) shows the variation of simulated soot mass and particle number density (PND) during the combustion process at CI and PPC conditions. The results point out that the soot mass and PND at SOI = -15 CAD aTDC are higher initially and decreases subsequently as the fuel injection timing is advanced to PPC conditions. The PND curve is very similar to the history of A4 because soot inception is the formation of the smallest solid soot particles from the collision of gas-phase PAHs species. This result also indicates that the soot nucleation is the main source of particle number.

Figure 13. The calculated in-cylinder evolution of soot mass and particle number during the combustion process at CI and PPC conditions

Figure 14 presents the in-cylinder local distribution of propargyl (C_3H_3) and pyrene (A4) species at CA50 and CA90 for different fuel injection timings. The distribution of C_3H_3 and A4 are noted to be consistent with the distribution of high-temperature zones. C_3H_3 and A4 are burned out in the high-temperature core zones with the temperature over 2200K, which results in the blank areas. As such, the higher mass fractions of C_3H_3 and A4 are primarily distributed in the edge of the high-temperature zones with the temperature below 2200K. The distribution area of C_3H_3 is slightly larger as compared to A4 at CA50. However, at CA90, most of the C_3H_3 is consumed while a considerable amount of A4 is left in the cylinder. As the combustion transits from CI mode to PPC mode, the in-cylinder distribution of C_3H_3 and A4 moves from the piston bowl to the piston top-land. The mass fraction of the two species is reduced, which indicates suppression in formation of soot precursors at PPC regime.

Figure 14 Volume contours of mass fraction of propargyl (C_3H_3) and pyrene (A4) during the combustion process at different injection timings

The in-cylinder distribution of soot mass and particle number density (PND) at CA50 and CA90 for various fuel injection timings are presented in Figure 15. With the advancement in fuel injection timing, the distribution of soot mass is reduced. On one hand, the in-cylinder homogeneity is improved, which reduces the rich zones in the combustion region. On the other hand, the in-cylinder combustion temperature is reduced that suppress the formation of soot precursors, as shown in Figure 10. It is obvious that the distribution of soot mass is similar to that of C_3H_3 at CA50 while high soot mass zones are more consistent with that of A4, as A4 is used as the soot nucleation precursor. The distribution of PND is almost identical to the distribution of A4 and this result is consistent with the in-cylinder mass variation traces of A4 and PND, as shown in Figure 11 and Figure 12. At CA90, the higher soot mass zones are still consistent with higher PND zones while the PND distribution areas become larger than that of soot mass. However, for late fuel injection timings of -10 CAD aTDC and -20 CAD aTDC, the distribution of soot particles and A4 is different, wherein the higher zones of soot mass and PND correspond to lower A4 due to the conversion reaction of PAHs into soot. At PPC conditions, the in-cylinder combustion temperature is further reduced. Thus, the correspondence between soot particles and PAHs becomes increasingly apparent, especially for PND and A4.

Figure 15. Volume contours of mass fraction of soot and particle number during the combustion process at different SOIs.

5. Conclusions

In-cylinder combustion and soot evolution process for heavy naphtha (RON =50) were simulated by the CONVERGETM CFD code during the transition in combustion from CI to PPC at low load operating conditions. The predicted results showed good agreement with the combustion and engine-out emission measurements from experiments. Thus, the present PAHs mechanism and soot model can be used to calculate the in-cylinder combustion, gas-phase pollutants and soot particle formation under different combustion modes. The main conclusions of this study are as follows:

1. The experimental measurements of in-cylinder pressure, rate of heat release, CA 10 and CA 50 were satisfactorily reproduced in the simulations. While the peak RoHR was overpredicted in the simulation, the two-stage combustion behavior with LTR and HTR phases are evident in both experiment and simulation.
2. The combustion phasing is sensitive to SOI at late fuel injection timings (positive slope of $d(\text{CA}_{50})/d(\text{SOI})$) whereas combustion phasing starts to decouple from SOI at early fuel injection timings (negative slope of $d(\text{CA}_{50})/d(\text{SOI})$).
3. The intensity of combustion images decreases with the advancement of fuel injection timing, signifying more premixed low temperature combustion. The distribution of combustion clouds in the piston bowl and crevice region from the simulation is in consonance with the experimental findings at CI and PPC mode.
4. A high-temperature spray-driven combustion occurs at CI conditions with late injection timing of -10 CAD aTDC. The temperature and equivalence ratio distribution gradually weakens as the fuel injection timing is advanced to -20 CAD aTDC. A typical low-temperature stratified lean PPC occurs, as the injection timing is advanced to -40 CAD aTDC. Overall, mixture stratification increases and temperature decreases when SOI is advanced to early fuel injection timings.
5. The in-cylinder intermediate species mass of C_3H_3 , C_5H_5 , and PAHs (A4) are reduced at PPC mode when compared to CI mode due to premixed and low temperature combustion effect. Similarly, the soot mass and particle number distribution are lower at PPC mode; both of which are consistent with the PAH's distribution.
6. The distribution of soot mass and particle number density is consistent with the distribution of high-temperature zones during the combustion process under CI and PPC conditions.

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Abbreviations

A1	Benzene
A2	Naphthalene
A3	Phenanthrene
A4	Pyrene
AMR	Adaptive mesh refinement
C2*	Electronically excited dicarbon
CH*	Electronically excited methylidyne
CO ₂ *	Electronically excited CO ₂
CFD	Computational fluid dynamics
CAD	Crank angle degree
ECU	Electronic control unit
EVO	Exhaust valve open timing
EGR	Exhaust gas recirculation
HACA	Hydrogen Abstraction Acetylene Addition
IVO	Intake valve open timing
IMEP	Indicated Mean Effective Pressure
IBP	Initial boiling point
KH-RT	Kelvin-Helmholtz/Rayleigh-Taylor
LTC	Low-temperature combustion

1			
2			
3	452	LHV	Low heat value
4	453	LD	Light-duty
5	454	OH	Hydroxyl
6	455	OH*	Electronically excited Hydroxyl
7	456	PPC	Partial premixed combustion
8	457	PSM	Particulate Size Mimic model
9	458	PSDF	Particle Size Distribution Function
10	459	PN	Particulate numbers
11	460	PAHs	Polycyclic aromatic hydrocarbons
12	461	PLII	Planar laser-induced incandescence
13	462	PLIF	Planar laser-induced fluorescence
14	463	P _{in}	In-cylinder pressure
15	464	RoHR	Rate of heat release
16	465	SC	Schmidt number
17	466	T _{max}	The maximum mean in-cylinder temperature
18	467	TRF	Toluene reference fuel
19	468	TDC	Top Dead center
20	469	UHC	Unburned hydrocarbon
21	470	φ	Equivalence ratio

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