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Methane Direct Injection in an Optical SI Engine - Comparison between Different Combustion Modes

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Abstract

Natural gas, biogas, and biomethane are attractive fuels for compressed natural gas (CNG) engines because of their beneficial physical and chemical characteristics. This paper examines three combustion modes - homogeneous stoichiometric, homogeneous lean burn, and stratified combustion - in an optical single cylinder engine with a gas direct injection system operating with an injection pressure of 18 bar. The combustion process in each mode was characterized by indicated parameters, recording combustion images, and analysing combustion chemiluminescence emission spectra. Pure methane, which is the main component of CNG (up to 98%) or biomethane (> 98 %), was used as the fuel. Chemiluminescence emission spectrum analysis showed that OH* and CN* peaks appeared at their characteristic wavelengths in all three combustion modes. The peak of OH* and broadband CO2* intensities were strongly dependent on the air/fuel ratio conditions in the cylinder. Lower OH* and CO2* intensities were observed with lean air/fuel mixtures because under these conditions, more air was present, the combustion reactions were slower, and the cylinder pressure was higher. CN* was formed by the spark plasma and was detected over a particularly long period when using a dual coil ignition system. The intensities of the OH* and CN* signals correlated when using this ignition system. Combustion image analysis showed that the flame had a wrinkled boundary in stoichiometric and lean burn modes and was especially distorted in stratified mode. No yellow soot luminescence was observed during homogeneous combustion. However, the emission spectra and combustion images acquired during stratified combustion showed that soot formation occurred due to the presence of fuel-rich areas with inadequate mixing in the cylinder. The difficulty of maintaining stable fuel injection, achieving proper air/fuel mixing, and ensuring stable flame propagation in lean air/fuel mixtures increased cycle-to-cycle variations. However, the homogeneous lean burn and stratified combustion modes achieved significantly lower indicated specific fuel consumption values than stoichiometric combustion.

Introduction

Methane Based Gaseous Fuels in SI Engines

Industry and the transportation sector are the dominant sources of CO2 pollution. Authorities around the world are therefore introducing increasingly stringent regulations to reduce the CO2 emissions of on- and off-road light and heavy vehicles. For example, the U.S. Environmental Protection Agency has established a new strategy for passenger cars to reduce CO2 emissions to 143 g/mile (~89 g/km) by 2025 [1]. The European Union has also introduced strict regulations, requiring vehicle manufacturers to reduce fleet CO2 emissions to 95 g/km by 2020; even more strict limits have been proposed for the period after 2030 [2]. One way for vehicle manufacturers to comply with these requirements is to replace conventional fossil fuels with alternative liquid and gaseous fuels and biofuels.

Consequently, researchers in industry and academia are strongly incentivized to develop cleaner transport technologies such as alternative fuels to reduce the fuel consumption and exhaust gas emissions of internal combustion engines (ICE). Natural gas is a leading alternative fuel for vehicles [3] and it is the second most widely used alternative fuel in the transport sector [4]. It consists primarily of methane (CH4), which accounts for between 75% and 98% of natural gas composition. In addition, it often contains higher hydrocarbons such as ethane, propane, and butane, as well as non-combustible diluents such as nitrogen or carbon dioxide [5]. The composition of natural gas can vary depending on its geographical source, time of the year, and its treatment during production. The combustion flame temperature increases with the increased content of higher hydrocarbons in natural gas, resulting in increased NOx and CO2 emissions. Higher hydrocarbons also reduce knock resistance. To compensate for this, the spark timing must be retarded, which reduces engine
power and efficiency [6, 7]. The methane number (MN) quantifies the ability of a methane-based gas to resist to knock in an ICE. Most commercial natural gas has an MN above 70, while pure methane has an MN of 100. The MN of natural gas is inversely proportional to its content of higher hydrocarbons. According to European automakers, the MN of natural gas should be at least 70 (regular grade) at all points of sale because lower grade gas reduces engine performance. For example, the use of MN65 gas instead of MN70 gas in a boosted compressed natural gas (CNG) spark ignition (SI) engine reduced performance by ~10% [8].

Another attractive renewable fuel for ICEs is biogas or biomethane. Biogas can be produced from many kinds of waste including municipal waste, agricultural waste, and food waste. Raw biogas is typically methane-rich with a high content of CO₂ and some additional impurities, like N₂. Its contents of CH₄ and CO₂ typically range from 50% to 70% and 30% to 50%, respectively. Because of its content of CO₂ and N₂, biogas has a lower calorific value than natural gas: pure CH₄ has a calorific value of 36 MJ/m³ under standard conditions whereas the calorific value of biogas with a CH₄ content of 60–65% is only 20-25 MJ/m³ [9]. The use of raw biogas with a high CO₂ content in ICEs profoundly affects the combustion process: it reduces the rate of heat release (RoHR), combustion duration and flame speed decreased as the gas mixture’s CO₂ content increased [11]. Biogas must therefore be upgraded before use to remove CO₂ and other impurities (notably water, hydrogen sulphide, and siloxanes) [12]. Cleaned and upgraded biogas, or “biomethane”, can have a CH₄ content above 90% and can be mixed with natural gas to serve as an alternative to gasoline or diesel fuel [13]. If specific cleaning technologies are used, methane contents above 98% are achievable [12, 14]. Table 1 compares the physicochemical properties of methane and gasoline [15, 16].

Because methane has a higher octane number (RON>120) and higher knock limits than gasoline, it can be used at substantially higher compression ratios (CR) than are used in modern turbocharged gasoline engines. Theoretical relationship between Otto cycle thermal efficiency, compression ratio and the specific heat ratio shows that with increasing CR, engine efficiency increases [17]. Therefore, the high compression ratios achievable when burning natural gas offer opportunities to increase the thermal efficiency of ICEs [3, 18]. Accordingly, a downsized 1 liter 3-cylinder engine achieved a lower brake-specific fuel consumption when running on CNG with elevated compression ratios than when running on gasoline [19]. In another study, raising the CR from 10:1 to 13:1 increased engine efficiency by ~4% [20].

An additional advantage of methane is that it has a simpler molecular composition and a higher H/C ratio than gasoline [16]. Therefore, the CO₂ emissions from a vehicle using CNG could be ~25% lower than those from a gasoline-fueled vehicle for a given input of energy into the combustion chamber under stoichiometric conditions [13, 21]. This is comparable to the reductions in CO₂ emissions achievable using hybrid engines.

The use of natural gas as a fuel presents several challenges. First, its combustion enthalpy per unit mass is higher than that of gasoline or diesel. However, its energy density per unit volume is much lower [22], so the volume of gaseous fuel required to achieve stoichiometric conditions is much greater than the corresponding volume of liquid gasoline. In port fuel injection (PFI) systems, the ICE’s volumetric efficiency is reduced by occupied air volume in the intake manifold, which reduces engine performance [23]. Despite this drawback, PFI is still currently the dominant technology for injecting gaseous fuel into engines for light duty vehicles [24].

Direct injection (DI) systems could be used to achieve a better torque and power output as well as higher volumetric efficiencies that could rival those of gasoline engines [21]. Several recent industrial and academic research projects have examined the use of natural gas (methane) DI systems in experimental single cylinder engines and standard vehicle research engines [20, 24, 25, 26].

Varying injector and injection parameters such as the injection pressure, spray angle, injection timing, and injection duration will influence the combustion process, emissions formation, and the potential to operate the engine in different combustion modes. In particular, the injection pressure is a key variable in a DI system. High CNG injection pressures are typically used in diesel engines with high compression pressures, especially in liquified natural gas (LNG) systems where liquid gas is vaporized and then injected at a pressure of 250-300 bar [27, 28]. CNG systems with very high injection pressures require either a fuel tank designed for a very high pressure or a fuel tank designed for a lower pressure together with a multi-stage compressor. If the latter approach is chosen, the compression work will reduce the overall engine efficiency [22]. Several studies have shown that lower gas injection pressures of ~16-30 bar are sufficient to deliver the quantity of fuel required for stable engine operation, even under full load [20, 21, 24].

The effects of lower injection pressures (7.5 and 12 bar) on engine performance have also been studied. Comparatively long injection times are needed to inject sufficient fuel under these conditions. This reduces volumetric efficiency because part of the injection occurs during the intake stroke [28]. The start of the injection also has a marked influence on the combustion duration, the flame development angle, and the engine’s indicated thermal efficiency. Longer flame development angle and combustions duration can occur due to earlier injection timings. At very late injection timings, the cylinder back pressure limits the mass of fuel that can be injected.

### Table 1: Physicochemical properties of gasoline and methane [15, 16].

<table>
<thead>
<tr>
<th>Properties</th>
<th>Gasoline</th>
<th>Methane</th>
</tr>
</thead>
<tbody>
<tr>
<td>Formula</td>
<td>C₈H₁₈</td>
<td>CH₄</td>
</tr>
<tr>
<td>H/C ratio</td>
<td>1.9:1</td>
<td>4:1</td>
</tr>
<tr>
<td>Molecular weight [gmol]</td>
<td>98</td>
<td>16</td>
</tr>
<tr>
<td>Density at NPT [kg/m³]</td>
<td>720</td>
<td>0.67</td>
</tr>
<tr>
<td>Autoignition temperature [°C]</td>
<td>350</td>
<td>540</td>
</tr>
<tr>
<td>RON</td>
<td>94</td>
<td>&gt;120</td>
</tr>
<tr>
<td>Stoichiometric A/F ratio [kgair/kgfuel]</td>
<td>14.6</td>
<td>7.24</td>
</tr>
<tr>
<td>Laminar burning velocity in air [m/s]</td>
<td>0.45</td>
<td>0.43</td>
</tr>
<tr>
<td>Lower heating value [MJ/kg]</td>
<td>43.5</td>
<td>50</td>
</tr>
</tbody>
</table>

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Consequently, the difference between the injection and compression pressures must be large enough to ensure that sufficient fuel is injected during the injection period [29]. The injector’s positioning (top or side injection) can also influence the mixing of the injected gas and the air flow in the combustion chamber [24, 26].

Some studies have evaluated the performance of CNG DI systems using combustion modes including homogeneous stoichiometric, homogeneous lean burn, and stratified lean burn. The use of DI instead of PFI increased indicated thermal efficiency at low engine loads and under homogeneous lean combustion regimes [30]. In another study, fuel consumption during stratified operation with DI was found to be lower than during homogeneous combustion with DI because of reduced pumping losses [31]. As noted above, the use of CNG DI systems reduces CO₂ emissions [26, 32]. In addition, it can reduce NOₓ emissions relative to those of gasoline engines [33]. Notably, CNG DI yielded a lower smoke opacity value and lower soot formation under both stoichiometric (λ=1) and homogeneous lean (λ=1.3) conditions when compared to DI with gasoline. The low soot formation was attributed to the use of a gaseous fuel with no C-C bonds [32]. At high fuel injection pressures (100 to 150 bar), no smoke was generated when using natural gas as the fuel under lean conditions (λ=1.1), even when large quantities of fuel were injected [34]. Particle number measurements in a downsized CNG DI engine revealed that its emissions did not exceed critical levels [35]. A comparison of gasoline DI, gasoline PFI, and CNG DI showed that the gaseous fuel yielded the lowest particle number concentration. However, the emissions of particles smaller than 10 nm from the CNG DI engine were higher than those from a gasoline DI engine under lean conditions [32]. While these studies have provided important results, there have been no direct comparisons between different combustion modes (stoichiometric, lean burn, stratified) in a methane DI engine with a consistent experimental setup and constant testing conditions. In addition, there is a need to better understand the methane combustion process and emissions formation under mentioned combustion modes.

### Emissions Spectra during Combustion

Emissions spectrum analysis can be applied as for studying the combustion of diverse fuels because the fluorescence of combustion products can reveal details of combustion process behavior. In SI IC engines, atomic recombination during the spark discharge phase causes the formation of diatomic molecules and excited radicals such as CN*, OH*, NO* and NH*. CN* is formed by the recombination of C and N generated in the plasma and is generally identified by the *Violet System* bands (*B*′Σ→*X*′Σ*) which cause emission at 380 - 390 nm, with a highest peak at 388 nm. There are also weaker bands at 359 and 420 nm due to Δν = ±1 transitions. However, the overlap of emission bands makes it difficult to distinguish between species against the background of plasma emissions. NH* can be detected between 330 and 340 nm (*A*′Σ→*X*′Σ*), with a strong peak at 336 nm. CN*→NH* balance can be related to the local A/F ratio. N₂* can emit weakly at ~316 nm and ~337 nm due to C*Π→B*Π (0, 1) and (0, 0) transitions. In addition, N₂* emission can be characterized due to transitions in the 330-435 nm range [36, 37, 38]. However, it has been noted that the emissions of N₂* and N₂* can be very unstable [38, 39]. During combustion, chemical reactions in the flame give rise to excited species (OH*, CH*, C₂*, and CO₂*), which are the main chemiluminescent emitters in hydrocarbon flames. In addition, hydrocarbon flames often contain solid bodies (e.g. soot and ash) that emit so-called black body radiation. The key reactions in the formation of these species are presented in Table 2 along with their characteristic wavelengths [36, 40, 41].

After the spark discharge, the flame kernel is initiated, and different light-emitting species can be detected. At high temperatures, the fuel’s (e.g. methane) thermal decomposition causes chain initiation where pool of OH* (also O* and H*) radicals develops [42]. OH* appears during flame kernel (also appears during the spark discharge) due to A-X transition at vibrational band (0, 0) at 306 nm and at 309 nm wavelength [36, 38, 43]. The OH* radical is in the primary combustion zone via the chemical reaction shown in Table 2. OH* forms in chain branching and propagating reactions during hydrocarbon oxidation [44]. OH* is thus a marker of combustion reactions. The intensity of its emissions depends on the local temperature and A/F ratio. It is stated that the locations with high OH* intensity values can characterize the A/F ratio near to the stoichiometric conditions [45]. Tests with several fuels showed that the OH* band due to the flame kernel consistently appears at 309 nm. OH*, O*, H* play key roles in the initiation of combustion because they are the main species responsible for breaking C-H bonds in hydrocarbon fuels [38]. Importantly, calibrated chemiluminescence techniques can give quantitative OH* measurements. OH* is being produced by a variety of mechanisms of energetic reactions depending on the flame chemistry. The most likely hydroxyl producing reactions result in electronic ground state OH. The majority of OH radicals do not give chemiluminescence, e.g. the production of ground state OH (reaction H+O₂→O+OH) in lean methane/air flame produces million times more OH than in the reaction of CH+O₂→CO+OH* [46]. CH* and C₂* emit blue and green light, respectively. CH* is primarily formed by the reaction of C₂H with O₂, and its chemiluminescence is most intense at ~431 nm (*A*Δ→*X*Δ*Π*). CH* loses its energy in few ways - through spontaneous fluorescence (chemiluminescence) or through the physical

### Table 2

<table>
<thead>
<tr>
<th>Species</th>
<th>Reactions</th>
<th>Wavelength, nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>OH*</td>
<td>CH + O₂ → CO + OH*</td>
<td>282.9, 308.9</td>
</tr>
<tr>
<td>CH*</td>
<td>C₂H + O₂ → CO₂ + CH*</td>
<td>3871, 431.4</td>
</tr>
<tr>
<td>C₂*</td>
<td>CH₂ + C → C₂* + H₂</td>
<td>473.7, 516.5</td>
</tr>
<tr>
<td>CO₂*</td>
<td>CO + O + M → CO₂* + M</td>
<td>350 - 600</td>
</tr>
<tr>
<td>CO₂*</td>
<td>CO₂ → CO₂ + hv</td>
<td></td>
</tr>
<tr>
<td>CO₂*</td>
<td>CO₂ + M → CO₂ + M</td>
<td></td>
</tr>
</tbody>
</table>
quenching (collisions). Levels of CH* are strongly temperature-dependent; it is most abundant at high temperatures. It is also very unstable and reacts quickly with other molecules [45]. C2* (~516 nm) radicals are mainly present in rich flames [47].

CO2* is characterized by a continuous broadband emission, resulting in the “background” signal observed from 350 nm to over 600 nm [48]. It is a three-body reaction (Table 2) which involves CO and O in their electronic ground state which leads to the formation of electronically excited CO2*. Reaction 2 of CO2 formation (Table 2) is responsible for the light emitted by CO2* and reaction 3 is quenching step. It is stated that with higher OH* intensities the signal of CO2* is also getting higher due to higher OH* contribution in CO oxidation [38, 41].

Soot (particulate matter) is an important combustion product. Soot formation depends on the local stoichiometry, temperature, pressure, and air/fuel mixing. An engine's soot emissions depend on the competition between soot formation and oxidation processes. In a DI gasoline engine, soot chemiluminescence due to pool fires on the piston surface was detected at a high wavelength (650 nm). The emission spectra also revealed OH* formation regions [49]. Experiments with butanol and gasoline in an SI DI engine revealed a rising signal intensity at higher wavelengths (510 nm) due to soot luminosity [50]. Gaseous fuels (e.g. 100% CH4) can also have a dominated background radiation from soot [51]. The soot radiance and soot temperatures may peak a few crank angle degrees before the heat-release rate. Combustion temperatures can vary widely, with profound effects on soot oxidation; at their maximum, they can be as high as ~2400 K, under which conditions soot is rapidly burned. Conversely, the soot temperature can fall to only ~1500 K in regions where pool fires form during the late stages of combustion, which may be too low for soot oxidation [49]. The cooler the flame, the greater the tendency to soot [52]. Studies on CH4 pyrolysis via high temperature mechanisms revealed that ethane and ethylene form quickly, with acetylene forming later [42, 53]. Two competing processes occur in sooting flames: the formation of soot precursors via acetylene pyrolysis, and the oxidative degradation of these precursors by OH* [49, 50]. High temperatures increase the rates of both processes, but the rate of oxidation increases more rapidly [52]. The higher the temperature, the lower the tendency of premixed flames to soot. Oxidative attack on soot precursors does not appear to occur in diffusion flames. However, the pyrolysis rate still increases with temperature, leading to a higher rate of soot formation [52]. The tendency to soot also depends on the fuel’s C/H (carbon to hydrogen) ratio, which determines the OH concentration, and its O/C (oxygen to carbon) ratio [52, 54].

Because of its simple structure, CH4 has a relatively simple combustion reaction mechanism that can be described using only 53 species and 400 reaction steps (GR13.0 mechanism); for comparative purposes, the mechanism for iso-octane includes 860 species and 3,606 steps [55, 56]. Methane also has a unique tetrahedral structure with high C-H bond energies, and therefore has unique combustion characteristics [57]. Notably, the chemiluminescent intensities of natural gas (composition - 94% CH4; 6% diluents) flames are half those of comparable propane or iso-octane flames [47, 58]. A detailed optical analysis of methane (100% CH4) combustion in an ICE showed that methane DI under stoichiometric and lean conditions resulted in a lower chemiluminescence emission intensity than gasoline PFI [15]. OH*, CO2, and small amounts of CH* chemiluminescence were detected at typical wavelengths during methane (100% CH4) combustion in a PFI system [16]. Additionally, it was claimed that OH* radicals in natural gas flames have a flatter chemiluminescent intensity than CH* [47, 58]. Chemiluminescent emission originates from electronically excited reactive species. Natural gas is known as a clean burning fuel, so its radiant emission is predominantly due to chemiluminescence [59]. However, researchers noted that methane (100% CH4) and air flames can also have a dominated background radiation from soot in the visible spectrum as a black body radiation [51]. Additionally, previous research work of methane (100% CH4) DI stratified combustion revealed that the soot formation was due to fuel rich areas in the stratified lean burn conditions and the results were also supported by the spectrometer measurements [60].

Emissions spectra can be related to combustion parameters such as the rate of heat release. However, they are also sensitive to variables such as the equivalence ratio, turbulence, fuel properties and pressure [40]. Najm et al. [61] argued that CH*, OH* and C2* are unreliable flame markers because CH*, OH* and CO2* chemiluminescence signals can vary with the equivalence ratio even when the heat release is constant [62]. However, other studies showed that the chemiluminescence intensity due to OH* and CH* radicals, and the background intensity due to CO2*, can be considered good indicators of the rate of heat release [58]. It was also suggested that OH*, CH* and C2* radicals formed in the reaction zone can be considered good indicators of the heat release pattern and the reaction rate in premixed flames [63]. However, high turbulence in the cylinder can alter the relationship between emission spectra and the heat release rate. Higgins et al. [51] and Najm et al. [61] therefore argued that CH*, CO2* and OH* are not suitable indicators for global heat release determination under highly turbulent combustion conditions.

This review of the literature on natural gas/methane combustion processes and emission spectra shows that not all investigations have examined a range of combustion processes in ICE, and there is a particular lack of studies on different combustion modes in natural gas/methane DI systems. Due to different type of researches and versatile results of DI system, there is still lack of information on comparison related with combustion imaging and emissions chemiluminescence in ICE between three main combustion modes: homogenous stoichiometric, homogenous lean burn and stratified lean burn. Previous studies on gas DI [64] focused on the DI system and engine adjustable parameters such as the spark timing, injection timing, and injection duration. These earlier studies [60] also focused mainly on stratified combustion at low engine speeds and several combustion process time steps. This work builds on the earlier study by comparing three different combustion modes using a low pressure (18 bar) methane direct injection system in an optical single cylinder engine. Experiments were conducted to determine how the combustion processes evolve over time (expressed in CAD) and to explain their behavior by drawing on analyses of emission spectra. Key goals were to capture
images of the combustion processes, analyze the formation of chemiluminescent emissions, and compare key derived parameters for different combustion modes.

**Experimental Setup**

**Optical Measurement Setup**

Optical combustion measurements were performed using an AVL optical single cylinder engine whose specification is given in Table 3. As shown in Figure 1, optical access to the combustion chamber was provided via a glass ring between the cylinder head and a liner, and via a Bowditch-type flat piston. This configuration enables optical access from the bottom of the piston along the cylinder axis via a mirror that was positioned at the bottom of the elongated piston skirt. The cylinder head was also fitted with a quartz glass cylinder, so combustion could be observed from the side.

Images of methane combustion processes were captured with a high-speed camera through a quartz glass window (bottom view). The bottom view images were diverted through a 45° mirror towards the video cameras through a beam splitter. The beam splitter was an uncoated quartz plate reflecting ~10% of light intensity. The beam splitter directed view into a high-speed camera capturing the combustion and into a spectrometer which was connected to a second high-speed camera. Spectrometer had 180 mm focal length and grating of 200 lines/mm. The central wavelength was 475 nm and the spectral resolution was 0.7338 nm/pixel. The combustion images were captured with an Ametec Vision Research Phantom Miro M310 camera with a Nikon 105 mm f/2 lens. The resolution of the combustion images was 512x480, the exposure time was 98.46 μs, and the sampling rate was 7,407 frames per second. The intensified signal from spectrometer was recorded with an Ametec Vision Research v7.1 high-speed video camera. A mercury reference lamp was used to calibrate the wavelength spectrum based on the lamp’s known wavelength peaks and intensities. The calibration wavelengths of the mercury lamp were between 200 nm and 550 nm. The region of the combustion chamber visible to the spectrometer was limited by a vertical slit that covered the area between the valves in the center of the spark plug. This region is indicated by the red rectangle in Figure 2. The green rectangles in this figure surrounding the spark plug (central area) and a region below the spark plug show the areas in which emission spectra were acquired.

Spectrometer images were captured at a resolution of 656x456 pixels with a 110 μs exposure time and a sampling rate of 7,407 frames per second. Emissions spectra and video footage were recorded over 10 successive combustion cycles. The ability to record higher number of cycles was limited by the internal memory of emission spectra high speed camera. However, additional combustion images from bigger amount of cycles (from 80 to 100) were captured for all combustion modes with the same Ametec Vision Research Phantom Miro M310 high speed camera in order to proceed with a flame area analysis. Figure 3 shows a raw flame image captured during stoichiometric combustion (~3.5 bar IMEP) and its post-processed counterpart.

**TABLE 3** Optical engine specification

<table>
<thead>
<tr>
<th>Specification</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Displaced volume</td>
<td>0.487 dm³</td>
</tr>
<tr>
<td>Stroke</td>
<td>83 mm</td>
</tr>
<tr>
<td>Bore</td>
<td>90 mm</td>
</tr>
<tr>
<td>Connecting rod</td>
<td>139.5 mm</td>
</tr>
<tr>
<td>Compression ratio</td>
<td>9.86:1</td>
</tr>
<tr>
<td>Number of valves</td>
<td>4</td>
</tr>
<tr>
<td>Fuel injection pressure</td>
<td>18 bar</td>
</tr>
</tbody>
</table>


**FIGURE 2** Combustion chamber of the experimental engine showing the areas visible to the spectrometer and in which emission spectra were captured (enclosed within the red and green rectangles, respectively).
Image illustrated the good agreement between the detected and real flame contours. The combustion image and emission spectrum analysis were carried out using Matlab software code.

**Fuel Injection System and Operating Conditions**

The engine was equipped with a specially designed prototype DI injector for gaseous fuels that opened outwards using a solenoid actuator. The injector’s operating pressure was 18 bar and it produced an A-shaped jet with a cone angle of 60 degrees (Figure 4).

As mentioned before, CH₄ is usually the dominant constituent of CNG and biomethane, therefore all experiments were performed with pure CH₄ as the test fuel (MN=100). This minimized the possibility of variation in results due to variation in gas composition, which usually depends on the source of the gas and the technology used in its production. Gas was supplied from a bottle pressurized to 200 bar. A gas reducer was used to reduce the pressure to 18 bar, and gas was supplied to the DI injector via a Micro Motion ELITE Coriolis mass flow meter CMF010M323NB, which had a measurement accuracy of ±0.35%. The injector was controlled using the LabView Direct Injection system module, which can control an injector’s opening time on a scale of milliseconds (ms).

Three combustion modes were tested at three different engine loads, at an engine speed of 1500 revolutions per minute (rpm) (Table 4). For all tested conditions, the spark timings were selected when the brake torque was maximum and the combustion was the most stable. To achieve proper mixing, the fuel was injected during early intake stroke in stoichiometric and lean burn combustion modes. However, under stratified conditions, the fuel was injected very late in the compression stroke to achieve mixture stratification and to maximize combustion stability. The injection duration depended on the combustion mode: it was always shorter in homogeneous lean burn mode than in stoichiometric mode because the former mode requires less fuel to achieve a given load point. Much greater injection durations were used in stratified combustion because gaseous fuel was injected during the compression stroke at a high back-pressure. Also, to inject the fuel at a higher load and maximize the pressure difference between the fuel injection system and the cylinder compression pressure, the fuel was injected at the highest possible pressure - 18 bar. As mentioned previously, injection pressures of 18 bar are sufficient for operation at full load [20, 24].

A standard single coil ignition system was used for the homogeneous stoichiometric and homogeneous lean burn combustion modes to ensure that the ignition conditions matched those in current CNG vehicles using these combustion modes. However, pilot tests of stratified combustion with a single coil ignition system revealed combustion stability issues resulting from excessive misfiring. Therefore, for stratified mode experiments, a dual-coil ignition system was used to ensure acceptable combustion stability. The spark duration was increased by using a multi-strike system that allowed the spark plug to discharge several times per cycle; in total, the ignition system could perform 6 strikes per combustion cycle at an engine speed of 1500 rpm. Methane has a high ignition temperature and a low flame speed [57], so different combustion modes require tailored ignition systems, especially when using lean air/fuel mixtures. It was also investigated by Polcyn et al. [65] that CNG engine working in leaner conditions and with a standard (single coil) ignition system, experienced ~10% coefficient of variation of IMEP (CoVIMEP) and the ignition system with increased discharge energy and duration could reduce the variations with up to 5%.

The engine’s cylinder pressure was measured with a Kistler 6053CC60 pressure sensor (sensor sensitivty 20 pC/bar) and all pressure data was sampled using an AVL Indicom data acquisition system. The air/fuel ratio was determined by measuring the oxygen content of the exhaust gas, using a Horiba Mexa-110, which determines the air/fuel (A/F) ratio with an accuracy of ±0.1.

**Results**

**Indicated Parameters**

Fuel consumption and cylinder pressures were sampled during the experiments. The fuel mass flow was measured 3 times at each load point for each combustion mode. The average fuel mass flow per unit time together with calculated
error bars and average indicated specific fuel consumption data are presented in Figure 5.

As expected, the lowest fuel consumption and the lowest indicated specific fuel consumption (ISFC) were achieved in stratified combustion mode due to the lower pump losses and lower heat losses. The ISFC for the stratified combustion was 192.19 g/kWh at around 2.5 bar IMEP, and it rose to 206.55 g/kWh at ~5 bar IMEP. At the lowest load, the ISFC for stratified combustion was 30.7% lower than that for stoichiometric combustion. However, at the highest load, the difference in ISFC between stoichiometric and stratified ISFC was 9.3%.

The fuel flow and ISFC results for homogenous lean burn mode were in between those for the other two modes; the ISFC in lean burn mode was 11.7% lower than that for stoichiometric combustion. However, at the highest load, the difference in ISFC between stoichiometric and stratified ISFC was 9.3%.

The fuel consumption and ISFC for three combustion modes and three engine load points are shown in Figure 5.

Cylinder pressure traces were also captured for the three combustion modes. Three measurements spanning 50 combustion cycles each were performed for each mode, so 150 combustion cycles were analyzed for each mode overall. Averaged pressure traces for these cycles are shown in Figure 6.

The figure shows results obtained at the ~3.5 bar IMEP load point. The highest cylinder pressures occurred during homogeneous lean burn combustion, for which the peak pressure was 26.64 bar. The lowest peak pressure (22.48 bar) was observed under stoichiometric conditions. The peak pressure occurred between ~8 and ~10 crank angle degrees (CAD) after top dead center (aTDC). Because the lean burn and stratified combustion modes use lean A/F mixtures, they require comparatively large amounts of air in the cylinder during the intake stroke. Therefore, less throttling was applied in these modes, which increased the pressure in the air intake manifold (Table 4).

RoHR curves for the three combustion modes at the ~3.5 bar load point are shown in Figure 7. Under lean burn conditions, the RoHR starts increasing earlier than under
stoichiometric conditions. This can be attributed to the earlier ignition timing in the former mode, which results in an earlier initiation of combustion. Combustion occurs over a longer period of time in lean burn mode because the A/F mixture was lean, and the methane burned more slowly. The RoHR peak in the lean burn case was lower than in the stoichiometric case. Both stoichiometric and lean burn combustion modes had a very long heat release period at the end of a combustion process. The RoHR curve for the stratified case can be divided into three phases. The primary minor phase occurred just after ignition, between 40 CAD before top dead center (bTDC) and 20 CAD bTDC, and was due to very slow combustion in rich mixture zones.

This was followed by the main combustion phase, in which the flame propagates in the locally flammable mixture around the spark towards the cylinder walls, reaching a high combustion speed. The final phase is the post-oxidation stage, which has a lower RoHR but a longer duration. Particulate matter can form at this time in regions with locally rich mixture and in pool fires on the piston surface [66]. However, previous studies showed that no pool fires occurred during stratified methane combustion. Instead, soot formation occurred mainly in the bulk flame due to insufficient mixing [60].

### Combustion Stability Analysis

Measurements showed higher CoV IMEP values with gas DI system in present work (Table 4). Overviewed researchs of methane combustion in ICE showed that a CoV IMEP of ~2% can be achieved with a CNG PFI system at 2000 rpm and 5 bar IMEP [16]. CNG DI systems can also achieve relatively stable combustion, with CoV IMEP values of 2% to 3% at 2000 rpm part load [24]. To explain higher variation seen in this work, Figure 8 presents box plots showing the variation in the peak cylinder pressure between cycles for each combustion mode at ~3.5 bar IMEP.

The red lines show the median peak pressure (50th percentile) for each mode. The upper and lower blue box contours indicate the upper (75th percentile) and lower (25th percentile) quartile boundaries. The black lines (whiskers) show the range of the remaining datapoints and red dots represent outliers [67]. It is clear that the CoV IMEP was highest during stratified combustion and lowest during stoichiometric combustion.

The high variation in the cylinder peak pressure caused variation in the work done on the piston during the expansion stroke, leading to variation in the engine torque. Similar results were observed under low (~2.5 bar) and high load (~5 bar) conditions.

Further analyses of flame propagation were conducted to better explain the high CoV IMEP values and variation in peak cylinder pressure. Figure 9 shows the flame area as a function of time (in CAD) for each combustion mode at lowest (~2.5 bar) and highest (~5 bar) loads.

Results for two load cases are presented to show how load affects the variation in flame development. Each green dot indicates the detected flame area for a single cycle at the indicated time (results for 80 to 100 cycles are shown for each time point). The black line shows the trend in the average flame area as a function of time in CAD. The secondary y axis and blue dots show the coefficient of variance for the flame area. It is clear that at low engine load (~2.5 bar), the variation in early flame development (from -25 to -10 CAD) is higher than in the high load case (~5 bar) under homogenous conditions. The high variation during the early flame formation period affected the later stages of combustion, increasing the spread of the cylinder pressure peaks. Characterization of the early stages of combustion was more challenging in the stratified case because the combustion flame was more uneven and the high energy spark plasma discharge persists for longer, complicating the detection of flame contours. However, flame detection was consistently achieved during the later stages of stratified combustion, revealing that the variation in flame area under stratified conditions was greater than in any other tested mode, in keeping with the previously discussed CoV IMEP values. In general, the lean combustion conditions used in the
lean burn and stratified combustion modes appear to have increase the CoV IMEP.

Additionally, early flame development appears to have taken much longer under stratified conditions than for homogeneous air/fuel mixtures. In general, lean conditions increase both the time needed for initial flame development and the duration of the rapid flame development period. Dilute fuel mixtures produce lower flame propagation speeds and reduce the rate of heat release, and misfires are more likely under very lean conditions [68, 69].

The observed instability during early flame formation may be due to the influence of the DI injector, the technical characteristics of the ICE, or the properties of the fuel. As noted in the Introduction section, several injector characteristics (including the injection pressure, injection timing, and injection duration) as well as swirl/tumble characteristics can influence the combustion process and its stability. Adequate stability under low load conditions is easily achieved with gasoline DI systems but is much more challenging with gas DI systems and the likelihood of success depends strongly on the geometries of the cylinder head and piston and charge motion. Combustion stability can also be influenced by shot to shot variation in fuel injection due to the injector’s characteristics. Figure 10 shows the relation between CoV IMEP and the injection duration for the three combustion modes at different engine loads.

For example, stoichiometric combustion was more stable at high load than at low load. This may be because the injection duration at low load was less than 1 ms; such a short duration could give rise to higher shot to shot variation between combustion cycles. The variation in the injected fuel mass may explain the variation in early flame development between combustion cycles. It has been argued [24] that shorter injection durations should be avoided because they may create non-linear transient flow zones. Comparatively long injection durations were used in this work to ensure sufficient fuel would be injected at higher loads. The CoV IMEP decreased as the injection duration increased.

The high observed cycle-to-cycle variation could also be due to inefficient A/F mixture formation and the physical properties of methane. Gaseous fuels generally have less inertia than liquid fuels, so gas plumes quickly lose momentum, resulting in poor fuel penetration because of strong aerodynamic drag under high pressure conditions [70]. It should be noted that CoV IMEP should be as low as possible to maintain adequate driveability [17, 21].

**Combustion Emission Spectroscopy**

Combustion chemiluminescence measurements were performed for all three combustion modes - homogeneous stoichiometric (λ=1), homogeneous lean burn (λ=1.4), and stratified lean burn (λ=2.2) - at 1500 rpm engine speed and ~3.5 bar IMEP engine load.

The wavelength spectrum was calibrated with known wavelength peaks and their intensities from a reference Mercury lamp. The main regions of the emission spectrum considered in this work are the OH* (~306-309 nm), CN* (~388 nm), CO2* (broadband from 350 to 550 nm) and soot formation (broadband 350 to 550 nm) regions, but there is also some discussion of NH* and CN* emissions. Combustion emission spectra were mainly acquired in an area near the sparkplug. However, some spectra were acquired ‘below’ the spark plug (see Figure 2) to better characterize soot formation during stratified combustion. Combustion images and emissions spectra acquired during an earlier study [60] indicated that soot formation occurred in stratified mode predominantly in the central and lower areas of the cylinder. This was primarily attributed to the movement of air in the cylinder and the composition of the air/fuel mixture in these areas of the combustion chamber.

As mentioned, a single coil ignition system was used in the stoichiometric and lean burn combustion modes, but a dual coil ignition system with a longer spark discharge timing was used in stratified combustion mode. Previous work showed that a long lifetime plasma covered a greater volume of the cylinder and generated highly stochastic spark plasma deflections due to in-cylinder air motion, which improved combustion stability [60].

Figure 11 shows averaged spectra for ten combustion cycles in each combustion mode. Figure 11 a) shows spectra acquired in all three combustion modes at the spark timing. During spark break up, extensive dissociation and ionization occur during the breakdown phase [37]. In all cases, the most intense peak in the spectrum is at ~381 nm and likely corresponds to the formation of the CN* (0, 0) radical appearance during the spark discharge. CN* is usually present at 388 nm with additional satellite peaks at 359 nm (1, 0) and 421 nm (0, 1) [36]. Accordingly, several satellite peaks were detected at ~354 nm and ~418 nm. However, the wavelengths of these peaks were shifted slightly compared to the literature values, presumably because of the spark plasma’s high temperature. The intensity of the CN* peak is known to increase with the plasma temperature [43], which in turn depends on the applied ignition energy [37].

CN* intensities in the homogeneous stoichiometric and homogeneous lean burn modes were similar, but that in stratified combustion mode (when the dual coil ignition system was used) was much higher. The dual coil ignition system allows the spark glow to persist for a longer time, so the CN* peak remains visible in the emission spectrum for the stratified combustion case even when the piston has reached TDC (see Figure 11 b). In addition to CN*, the spark discharge also generates OH* radicals. The OH* peak at ~306 nm is visible in the spectra for all three combustion modes at the spark
During the arc phase, atomic recombination leads to diatomic molecule formation and the emergence of CN*, OH*, and NO* [37]. An earlier study [72] presented similar results, with OH* radical emissions emerging due to the high energy of the spark glow. The R and P branches of OH* (A-X transitions) occur at 306 and 309 nm respectively [43]. In addition, NH* (0,0) is likely to be formed during the arc phase, giving rise to peaks at ~330 nm [36]. NH* has previously been detected during CH₄ combustion at the spark timing [45], and a separate investigation showed that its formation depends on the spark timing by demonstrating that its presence only became apparent at around the spark timing even when the spark timing was varied [37], and that it always co-occurred with CN* and OH* emission peaks. Further evidence for NH* and CN* formation during the arc phase is provided in Figure 11 b), which shows an emission spectrum acquired later in the combustion process when the piston has reached TDC. The peaks corresponding to the mentioned species are still present at the appropriate wavelengths in the stratified combustion case because of the prolonged ignition under stratified conditions. However, these peaks have disappeared in the stoichiometric and lean burn cases because of the absence of the spark glow. The only peak visible at shorter wavelengths in the latter two modes at TDC is the OH* peak.

During spark discharge, chemical reactions are initiated in the region of high radical density, and OH* fluorescence is usually observed together with CH* fluorescence [43]. However, no clear evidence of CH* radical emission at ~431 nm was detected in the spectra for any of the combustion modes in either the early or the later stages of combustion. This could be due to the comparatively simple molecular structure of methane. Earlier experiments [45] showed that the intensity of CH* emission during 100% CH₄ combustion is much weaker than during gasoline combustion. This may be because gasoline consists of more complex hydrocarbons with higher molecular weights, resulting in the formation of more CH* during combustion.

The spectra for the homogeneous combustion modes at the later stage of the cycle feature clear peaks at around 308-310 nm. These peaks are characteristic for OH* and they indicate that high-temperature combustion is occurring, combustion reactions have been initiated, and the flame kernel is developing. The highest peak is observed in the stoichiometric combustion spectrum because a greater quantity of fuel is present and the combustion temperature is higher. The peak intensity was lowest in the lean burn case because of the small amount of fuel injected, the low combustion temperature, and the slower combustion process.

The background between 350 and 550 nm is dominated by emission from CO₂*. Broadband CO₂* emission is detected as blue light during combustion. The broadband intensity was highest under stoichiometric conditions. This can be attributed to the larger amount of fuel injected and combusted under stoichiometric conditions, which increases CO₂ formation and raises the gas temperature, both of which increase the intensity of the CO₂* signal. The intensity of CO₂* emission was much lower under lean burn and stratified conditions because less fuel was injected, and larger amount of air was present, so the temperature in the cylinder was lower. Similar explanations were proposed by Aleiferis et al. [63], who stated that the burned gas temperature in lean mixtures was lower than that in stoichiometric mixtures. Moreover, in lean mixtures, the dissociation of CO₂ was inhibited and the level of flame chemiluminescence was lower. Background broad-band emission can also be due to chemiluminescence from other sources such as HCO* bands (from 250 nm to 410 nm) or HCHO* bands (from 340 nm to 523 nm). These broadband bands were detected during CH₄ combustion in an optical SI engine fitted with a PFI system [16]. However, in present experimental research if HCO* or HCHO* emission is present it will be overlapped by emission from CO₂* and other species with a stronger emission. The peak observed at 540 nm in stoichiometric and lean burn combustion is probably due to emission from CuOH* [48] formed during combustion from trace amounts of Cu released by piston ring wear.

Additional analyses of the CN* and OH* emission spectra were conducted to clarify their behavior during combustion. The mean intensity of the CN* band as a function of time (expressed in CAD) is plotted for each combustion mode in Figure 12.

Green points show the spread of an emission peak at ~381 nm wavelength during the CAD timing from 10 recorded combustion modes. The red line shows averaged value of these points.

The spark discharge process can be divided into three phases: breakdown, arc discharge, and glow discharge. During breakdown, highly conductive plasma forms between the electrodes. All molecules in this spark channel are dissociated and ionized at a high plasma temperature. The plasma temperature decreases due to plasma cooling and recombination when the ionization and dissociation energies are transformed into thermal energy [43]. As mentioned previously, diatomic species such as CN* and OH* are formed by atomic recombination during this phase [27]. The duration of the glow discharge can be increased by using an ignition system with secondary coil assistance. In such systems, the glow discharge has lower power but more energy [43].
The abundance of CN* presence can also be related to the fuel concentration. It has been suggested [73] that CN* emission can be a marker of local fuel concentrations during the spark timing. It was determined that with leaner mixtures, the CN* intensity decreased linearly, suggesting that the CN* intensity is directly related to the carbon number density at ignition phase. A convenient expression for estimating the carbon number density at the ignition timing was proposed based on this hypothesis [37]. In this work, the CN* peak intensity was highest at the spark timing and decreased over time. This can be attributed to the decreasing carbon number density around the spark plug during the initiation and development of the combustion process. Similarly, it was noticed that the radiation of the glow gradually diminished and the CN* intensity declined in parallel as the amount of fuel surrounding the spark plug decreased [37, 73].

Figure 13 plots the intensity of OH* emission at 308 nm for the three combustion modes. Blue points represent the spread of OH* emission intensities for individual combustion cycles at each time point, and the red line shows the average OH* intensity over 10 combustion cycles.

The OH* intensities at the spark timing in the stoichiometric and lean burn cases are similar because both of these modes used a single coil ignition system. The OH* peak intensity declined slightly after its initial increase (shortly after the spark discharge) under both stoichiometric and lean burn conditions. However, after a brief period, the OH* intensity started increasing again, indicating that the combustion reactions had begun. It is known that OH* can be detected in the plasma phase and the initial flame kernel. However, the OH* intensity is usually quite low during the early stage of the arc phase but increases as the initial flame kernel develops [43].

The increase in OH* intensity occurred much earlier and much more quickly under stoichiometric conditions than in the lean burn case. Additionally, the maximum OH* intensity in stoichiometric mode was around 4 times higher than in lean burn mode. The lower abundance of OH* radicals in lean burn mode can be explained in a few ways. First, the pressure traces (Figure 6) clearly show that the pressure in the cylinder was higher with lean air/fuel mixtures because there was less throttling. Other researchers have found that the intensity of OH* and CH* signals declines as the cylinder pressure increases [62]. Secondly, and more importantly, the chemiluminescence intensity is known to depend on the corresponding reaction rates [74], and there is a strong correlation between the OH* intensity and the equivalence ratio [46, 75]; the concentration of the excited radical is directly proportional to the intensity of its emissions [40]. Moreover, it has been stated that the chemiluminescent intensity of OH*, CH*, C2*, and CO2* in methane/air flames depend on the equivalence ratio [76] which explains why the amount of reactions and the reaction rates are decreasing. Therefore, the OH* peak intensity would be expected to be lower under lean burn combustion. Accordingly, the increase in OH* intensity between 20 CAD bTDC and TDC (Figure 13) under lean burn conditions was much lower than under stoichiometric conditions. Since OH* radicals exist in both the flame front and the hot post-combustion gases [46], it may be that the flame speed was lower in lean burn mode because both the maximum OH* intensity and its rate of increase were lower than in

Emission spectrum analysis showed that the CN* peak intensities in stoichiometric mode (with a spark timing of 32 CAD bTDC) and homogeneous lean burn mode (with a spark timing of 38 CAD bTDC) were quite similar, and that high intensity was only sustained for a few crank angle degrees, like the spark plasma. Completely different CN* emission spectra behavior was observed in stratified mode (spark timing 38 CAD bTDC). In this case, the spark plasma persisted for a very long time - from 38 CAD bTDC to ~5 CAD aTDC (Figure 12). As mentioned above, the intensity of CN* emission was higher with the dual coil system than with the standard setup. This was due to the higher ignition energy and longer glow discharge period achieved using the dual coil system and is consistent with other studies showing that CN* intensities increase with the plasma temperature [43].
Homogeneous stoichiometric mode. The laminar flame speed in lean mixtures is known to be significantly lower than in stoichiometric mixtures [77].

In the stratified case, the OH* intensity started increasing at the moment of spark discharge. However, the shape of the OH* intensity curve is very different to that for the stoichiometric and lean burn cases because the OH* radical peak persisted for much longer time in the stratified case; its duration matched the dual coil ignition system's period of activity. Another interesting trend was that the OH* peak intensity increased during the spark glow, indicating that the abundance of excited radicals, the rate of the combustion reactions, and the rate of heat release were all increasing throughout this period. Moreover, the number of peaks in the averaged OH* curve (6 peaks) was equal to the number of spark strikes in the dual coil ignition system. The increase in OH* intensity in stratified mode (Figure 13) correlated with the decline in CN* intensity (Figure 12). These findings are again consistent with earlier reports [32] stating that the CN* intensity is proportional to the local fuel concentration. The results obtained here show that as the carbon number density around the spark plug declines, the OH* peak becomes more intense, suggesting that the amount of unburned fuel was decreasing and the amount of high temperature combustion was increasing in the measured area. This result also confirmed that the rate of the combustion reactions increased over time. However, the OH* intensity in stratified mode was still lower than that in stoichiometric mode. It was previously concluded [37] that very lean and very rich A/F mixtures both tend to reduce OH* intensity. It is known that locally rich regions being formed during stratified combustion, despite the overall leanness of the A/F mixture. In this case, the low intensity of the OH* peak in stratified mode suggests the presence of regions with very high densities of methane fuel. However, it is worth noting that the OH* intensity may be dominated by radicals formed in the plasma of the dual coil system because the spark signal likely overlaps the signal from the combustion process.

Further discussion is about the correlation between OH* and RoHR in order to find out if an OH* emission peak can be a marker of heat release rate and specially to explain the real OH* formation (due to combustion process) in a stratified case where the part of OH* emission from the combustion is overlapped by the OH* emission from the spark plasma. As mentioned in the introduction, the heat release rate depends on many parameters including the fuel type, fuel mass flow rate, air/fuel ratio, temperature, and pressure [47]. However, the CH* and OH* chemiluminescent intensities correlated well with the maximum heat release rates, and publication of other scientists [78] stated that CH* and OH* could be used as RoHR markers in cases where the maximum OH* chemiluminescence coincided with the peak of the heat release rate.

A comparison of the RoHR curves (Figure 7) and OH* emission spectra (Figure 13) in stoichiometric and lean burn modes showed that the RoHR curve started rising at approximately the same time (in CAD) as the OH* intensity, emission peak rise where the combustion chemical reactions started to initiate. The timing of the peaks in the OH* intensity also correlated with the timings of RoHR peaks, which occurred between TDC and 10 CAD aTDC. After that, OH* radical emissions decreased in parallel with the RoHR in both combustion modes. As mentioned previously, the OH* emissions from the spark plasma probably overlapped with those from the combustion process in stratified mode. Since the OH* intensity and RoHR should be correlated in the same way as in the stoichiometric and lean burn cases, it should be possible to identify the real OH* peak and the timing of the maximum combustion rate by examining the maximum of the RoHR curve in stratified mode. Figure 7 shows that in stratified mode, the heat release peaked at ~10 CAD aTDC; therefore, the OH* intensity due to combustion reactions should also be highest at this time point. Shortly after the spark plasma disappeared (at ~5 CAD aTDC; see Figure 13), the OH* intensity fell and then stabilized between 10 CAD aTDC and 15 CAD aTDC when the RoHR was at its highest.
This indicates that the OH* emissions during this interval were mainly due to excited species formed by stratified combustion. However, the hydroxyl radical peak intensity was still much lower than in stoichiometric mode. Later in the cycle, the OH* intensity declined in parallel with the RoHR, showing that the rate of the combustion reactions was falling, the number of reactions was decreasing, less energy was being released, and the temperature in the cylinder was falling. The low OH* intensity in stratified mode presumably indicates a low abundance of hydroxyl radicals, which could reduce the rate of soot oxidation and increase engine-out soot emissions. To evaluate this hypothesis, the higher wavelength region of the emission spectrum was compared to the region containing the OH* peak during the period when detectable emission spectrum was present in the vicinity of the spark plug.

Figure 14 shows the OH* emission (at 308 nm wavelength) and higher wavelength spectrum at ~3.5 bar engine load. The green circles represent the observed OH* intensity at the indicated time points in 10 individual combustion cycles, and the blue line represents the average of these values. As noted previously, the broadband emission starting at ~350 nm is due to CO2* chemiluminescence under stoichiometric and lean burn conditions. In the plots for the homogeneous combustion modes in Figure 14, the black circles indicate the intensity of this broadband emission in individual combustion cycles, and the red lines represent the averages of these points. The intensity of the CO2* chemiluminescence was defined as the intensity at ~420 nm. The CO2* chemiluminescence clearly correlates with the OH* curve, showing that OH* is involved in CO oxidation as discussed by other researchers [38]. The maxima of the OH* and CO2* curves occurred at almost the same time (~8 - 10 CAD aTDC), a few CAD after the RoHR maximum. Like the OH* intensity, the intensity of CO2* chemiluminescence in homogeneous lean burn mode was much lower than in stoichiometric mode due to the lean conditions (larger amount of air and less injected fuel) which reduced temperature of combustion process and produced CO2 gas. It is well known that during lean burn combustion also less CO2 is produced. This should reduce CO2 emissions via the exhaust.

This work and earlier studies on combustion imaging and emission spectra [60] have shown that there is strong chemiluminescence emission intensity at higher wavelengths (~550 nm) during stratified methane combustion. The steep rise in intensity at 550 nm is characteristic of black body radiation which can be a specific indicator of soot formation. In the plot for the stratified combustion mode in Figure 14, the green circles indicate the OH* intensity (measured at 308 nm) in individual combustion cycles, and the blue line represents the average OH* intensity over 10 combustion cycles. The black circles show the soot emission intensity at 550 nm wavelength during the same combustion cycles; the red line shows the average soot intensity over 10 combustion cycles.

As mentioned above, the OH* intensity fell by a factor of two shortly after the spark plasma disappeared (at ~5 CAD aTDC) in the stratified case, and the timing of the maximum OH* intensity (~10 CAD aTDC) coincided with the maximum heat release rate. Shortly after the spark glow disappeared, the higher wavelength emissions started to increase, as shown by the red line in Figure 14. In some combustion cycles, the black body radiation was 4 times more intense than the OH* radiation. Soot formation usually occurs in fuel-rich zones in the cylinder at temperatures between 1400 and 2000 K where mixing-controlled combustion dominates.

Because OH* formation in stratified mode was reduced by the presence of localized regions with very rich air/fuel mixtures, the rate of soot oxidation was reduced and so overall soot emissions were high. There is a correlation between levels of OH* and ground state OH radical formation, so if the intensity of OH* luminescence is low, the abundance of OH is likely to also be low. This is important because OH radicals are considered to be the dominant oxidants in soot oxidation. Once a carbon has been partially
Combustion Image Analysis

Combustion images were captured through the quartz glass piston. The images were taken at the same time during the same combustion cycles as the emissions spectra. Figure 16 shows combustion images from a single combustion cycle for three different combustion modes at ~3.5 bar IMEP engine load.

The image series for all three modes start at 15 CAD bTDC and show the combustion behavior in 5 CAD timesteps until the flame front reaches the boundaries of the region visible through the quartz piston. The brightness and gain of the images were modified by several percent during post-processing to show clearer images.

At 15 CAD bTDC, flame growth around the spark area can be seen under stoichiometric and lean burn conditions. This was also the point at which OH* emission became detectable (Figure 13) and the heat release rate started increasing (Figure 7). The bluish color of the flames is mainly due to CO* emission. Generally, the CO* emission occurs at the same locations as the OH* emission.

Visible flame growth also began at around 15 CAD bTDC in stratified mode, demonstrating that the combustion reactions were initiated around this time and confirming the conclusion that OH* radicals formed by combustion are present from an early stage, alongside OH* formed by the spark discharge. The combustion images show that the flame's growth is rather asymmetric, with wrinkled boundaries. Similar results were obtained in other studies [15, 16]; the asymmetry in those cases was attributed to the turbulence caused by the piston's motion together with the slow flame speed of methane and the low rate of combustion reactions. The combustion images for the stoichiometric mode show that the flame boundaries propagated towards the cylinder walls more rapidly than in the lean burn mode. By 0 CAD, the flame in stoichiometric mode had already reached the boundary of the visible area whereas in the lean burn case, some unburned regions remained after TDC. Additionally, the blue flame was more intense in stoichiometric mode than in lean burn mode. As mentioned, the leaner mixture produced lower combustion temperatures.

The combustion images for the stratified case support the conclusions based on emission spectroscopy (Figure 11), showing that the visible spark glow persists for a very long time. The flame radius was smaller than for the homogenous modes early in the cycle (at 15 CAD bTDC), indicating a comparatively slow initiation of combustion reactions. The yellow-orange flame, whose color is mainly due to the black body radiation of soot particles, started to appear closer towards the central and lower part of the cylinder area later in the combustion process (at 5 CAD bTDC). Additionally, the blue flame in the background had a highly distorted shape. This shows that high stratification can be achieved even when using methane gas at an injection pressure of 18 bar, and that complete mixing of air and fuel cannot be achieved. Consequently, there are fuel-rich regions where soot formation occurs. Diffusion combustion typically also occurs in liquid fuels when stratified injection is used or rich mixtures are formed, which are the main sources of soot [32, 81]. The emission spectra (Figure 14, Figure 15) and combustion images clearly show, that rich zones are not oxidized to CO, the carbon will not be involved into a soot particle [79, 80]. Soot formation under stratified conditions is also favored by the combustion temperature: stratified mode produced a lower RoHR and lower combustion temperatures than the homogeneous modes. Moreover, the cylinder temperature decreased still further late in the combustion process when the piston moved down, increasing the rate of soot formation. As noted in the Introduction, OH* is a key oxidant of soot precursors and particles but its effectiveness depends on the temperature [52].

Although the intensity of the black body emissions was decreasing up to 60 CAD aTDC (Figure 13), detectable soot luminescence continued until much later in the combustion process in certain areas of the combustion chamber. Soot luminosity was even detected at very late timings (>90 CAD aTDC) (Figure 15) in some stratified combustion cycles when emissions spectra were acquired in the lower part of the combustion chamber (below the spark plug, see Figure 2). The combustion images confirmed that soot formation continues even at very late timings in some combustion cycles. In keeping with this result, other study [49] found that soot luminosity could be detected late in the expansion stroke, at ~120 CAD aTDC.

Soot luminescence during the late stages of stratified combustion is undesirable because temperatures at this stage are low and oxidation is slow [81]. Additionally, the soot intensity in the region below the spark plug was high in many of the observed combustion cycles.

However, the intensity of black body radiation in some combustion cycles (both near the spark plug and below it) was markedly lower than in others during the early stages of mixing controlled combustion (from 0 to 20 CAD aTDC). This early decrease in soot luminosity could be explained by a rapid turbulent mixing with surrounding hot lean regions, which led to rapid soot oxidation and early complete burnout [49]. Overall, one would still expect the particulate content in the exhaust gas from an engine with a gas DI system to be lower than that for a gasoline DI engine because of complexity of gasoline's molecular structure. As mentioned before, soot formation is very dependent on the fuel's H/C ratio. Accordingly, studies on CNG DI engines showed that the particle number concentration can be lower than in comparable gasoline DI engines [32].

![FIGURE 15](image-url) OH* and soot emission spectra below the spark area in stratified combustion mode at -3.5 bar IMEP.
uniformly distributed in the cylinder: they are mainly found on the exhaust valve side, possibly because of in-cylinder motion and turbulence.

Emission spectroscopy and previous work [60] showed that a well-mixed blue flame was present even late in the combustion process (after 5 CAD aTDC) (Figure 16), and at the same time there was lots of soot formation at different cylinder locations when the stratified combustion was applied in the methane direct injection system. However, combustion images and emission spectra indicated that homogeneous combustion generates well-mixed blue flames with none of the soot formation zones seen in the stratified case.

Conclusions

An experimental study comparing three different combustion modes (homogenous stoichiometric, homogeneous lean burn and stratified) using methane with an 18 bar direct injection system yielded the following conclusions:

- As expected, lean fuel/air mixtures showed the lowest fuel consumption. Because of reduced pumping losses and lower heat losses, the ISFC in stratified mode was ~31% lower than that for stoichiometric combustion at the lowest load (~2.5 bar) and ~9% lower at the highest load (~5 bar). Lower fuel consumption in lean modes would be expected to reduce CO₂ emissions in the exhaust gas.

- The CoVIMEP was highest in the homogeneous lean burn and stratified combustion modes. Lean A/F mixtures resulted in lower flame propagation speeds and RoHR values, and also increased the cycle-to-cycle variation in the combustion process. Notably, high variation in the early stages of flame development increased the variation in cylinder peak pressure. The physical properties of methane such as its low inertia and poor penetration, as well as injector characteristics (such as variation in the quantity of fuel injected between shots) could also contribute to differences in combustion stability between modes.

- Emission spectroscopy revealed that CN* and OH* emission peaks can be observed at their characteristic wavelengths during methane combustion in different modes. The intensity and duration of CN* emission depended on the spark glow time and the nature of the ignition system. The OH* peak, as an indicator of hot-burning gas, was a relatively good marker of the heat release rate in all combustion modes: the OH* emission intensity correlated with the RoHR and the CO₂* intensity in homogenous cases.

- Stable stratified combustion could only be achieved using an improved ignition system. The limited mixing time resulted in the emergence of locally fuel rich regions that enabled soot formation, which peaked around the time of maximum RoHR. Black-body emissions from the soot were also detected in the emission spectra and combustion images acquired late in the combustion process when the temperature was too low for efficient oxidation, which would result in engine-out soot emissions. Strategies to improve air/fuel mixing to reduce soot formation and enhance its oxidation will therefore be required.

- Combustion images confirmed the emission spectroscopy results, showing that the flame had more wrinkled boundaries and propagated more slowly towards the cylinder walls with leaner mixtures because of slower combustion reactions. The flame shape in the stratified case was more distorted due to an uneven air/fuel mixing and turbulence in the cylinder, and soot formation was confined to localized areas.

References


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Definitions/Abbreviations

A/F - Air/fuel
aTDC - After top dead center
bTDC - Before top dead center
C* - Diatomic carbon
CAD - Crank angle degree
<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Definition</th>
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<tr>
<td>CH*</td>
<td>Hydrocarbon</td>
</tr>
<tr>
<td>CH₄</td>
<td>Methane</td>
</tr>
<tr>
<td>CN*</td>
<td>Cyano radical</td>
</tr>
<tr>
<td>CNG</td>
<td>Compressed natural gas</td>
</tr>
<tr>
<td>CO</td>
<td>Carbon monoxide</td>
</tr>
<tr>
<td>CO₂</td>
<td>Carbon dioxide</td>
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<td>CR</td>
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<td>Hydrogen and carbon atom ratio</td>
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<td>Indicated mean effective pressure</td>
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<td>ISFC</td>
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