Effect of Hydrogen Concentration on Engine Performance, Exhaust Emissions and Operation Range of PREMIER Combustion in a Dual Fuel Gas Engine Using Methane-Hydrogen Mixtures

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ABSTRACT

A single cylinder, supercharged dual fuel gas engine with micro-pilot fuel injection is operated using methane only and methane-hydrogen mixtures. Methane only experiments were performed at various equivalence ratios and equivalence ratio of 0.56 is decided as the optimum operating condition based on performance, exhaust emissions operation stability. Methane-hydrogen experiments were performed at equivalence ratio of 0.56 and 2.6 kJ/cycle energy supply rate. Results show that indicated mean effective pressure is maintained regardless of hydrogen content of the gaseous fuel while thermal efficiency is improved and presence of hydrogen reduces cyclic variations. Increasing the fraction of hydrogen in the fuel mixture replaces hydrocarbon fuels and reduces carbon monoxide and hydrocarbon emissions. Mixtures with higher hydrogen content undergo faster heat release from flame propagation, approach knocking limit faster and are less knock resistant. 40% methane - 60% hydrogen mixture is prone to premature autoignition and superknocking, and is the critical concentration limit for methane-hydrogen mixtures.

INTRODUCTION

Diminishing petroleum reserves, strict exhaust emission standards and environmental concerns are putting more emphasis on alternative fuels. Alternative fuels are desired to have clean combustion characteristics, reliability of sources and sustainability. Hydrogen is a promising candidate as an alternative fuel: it is a green fuel that is not

involved in carbon cycle and can be produced from various sources including waste materials.

Use of hydrogen in SI engines has been suggested and investigated by various researchers as early as 1980's. It has been discovered that, on top of being suitable as the main fuel for an internal combustion engine, its rapid burning properties can make this fuel suitable as a secondary fuel for increasing thermal efficiency [1-8]. On the other hand, use of hydrogen in dual fuel gas engines is not as well investigated due to the fact that this approach requires significantly lean conditions in order to prevent knocking which limits the engine output [9-15]. On the other hand, when hydrogen is used in dual fuel gas engines, unburned hydrocarbon (HC) and carbon monoxide (CO) emissions are at negligible levels because the only source of these emissions is pilot fuel injection, but this improvement is accompanied by considerable increase in NO_x emissions [10].

Hydrogen is the main fuel component in biomass gas, also known as producer gas, alongside with CO and a small fraction of methane, which is commonly produced by processing biological wastes like wood chips. Producer gas is widely studied for use in SI engines [16-21]. Most of the studies report several problems about compatibility of producer gas with spark ignition operation. N_2 and CO_2 are common diluents in the composition, total fraction of which may exceed 60% depending on the sources. Spark ignition engines show poor performance in presence of diluents where misfiring or instability are common problems. In addition, compositional instability is at significant levels, which also has a negative impact on operation stability as well. In contrast, dual fuel CI

operation is not as sensitive to these factors. Still, dual fuel CI approach is not as well understood. While this has been studied by several research groups, the efforts are made mostly on practical use, such as insite use for electricity generation [22-26]. In order to maximize the benefits, fundamental knowledge requires further attention.

Use of producer gas in dual fuel engines have been investigated in current laboratory using micro pilot injection [27-32]. In these studies, it is found out that a fraction of gaseous fuel undergoes autoignition at the end-gas regions without knocking when the conditions are suitable. This phenomenon is later named as PREmixed Mixture Ignition in the End-gas Region (PREMIER) combustion [32]. In order to understand the underlying phenomenon better, it is necessary to understand the behavior of constituents in producer gas. So far, effect of fuel composition has been investigated by comparing producer and coke oven gas types of various compositions [31, 32]. This approach provided an overall insight, however the effect of each separate gas and the interaction between them is still not fully understood. In order to fully comprehend the behavior of components of producer gas under PREMIER combustion conditions, it is necessary to isolate a single parameter from others which might introduce uncertainty. This goal can be achieved by using only two types of gases at a time, the nature of one of which is well known.

The aim of this study is to understand the effect of hydrogen concentration on various characteristics of PREMIER combustion. In order to achieve this goal, hydrogen will be mixed into methane at various volume fractions and the results will be compared to those of 100% CH₄. Since methane is studied thoroughly and its behavior is fully understood, it can serve as an appropriate comparison basis.

EXPERIMENTAL SETUP AND TEST PROCEDURE

Diagram of experimental setup is given in Fig.1. In this study, experiments are performed in a single cylinder supercharged gas engine with 96 mm bore and 108 mm stroke and 16.4:1 compression ratio. Prior to the study, it has been noticed that engine tends to enter knocking regime very early when intake air pressure is 200 kPa, which not only narrows overall and PREMIER combustion operation range, but also causes unstable engine operation. Engine specifications are listed in Table 1. Engine peripherals include three optical sensors for detecting top dead center (TDC), cam TDC and the crank angle with 0.5 degrees sensitivity. These three signals are used as inputs of the controller and injection timing signal is generated as required with 0.5 degrees of crank angle resolution. Engine is operated at 1000 rpm constant speed.

All performance data presented in this work are indicated values and they are obtained from cylinder pressure versus crank angle data. Indicated mean effective pressure is calculated over four strokes; therefore the results presented in this work are net indicated mean effective pressures. A pressure

transducer is attached to the cylinder head in order to obtain in-cylinder pressure data. 80 consecutive cycles are recorded at each session. NO_x , carbon monoxide (CO) and unburned hydrocarbon (HC) emission are recorded using exhaust gas analyzers. Soot emissions are monitored as well; however the results were below the measuring range of the opacimeter (less than 1% in all cases) and it is safe to assume that soot emission was not detected.

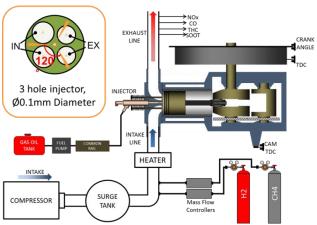


Fig.1 Diagram of experimental setup.

Table 1 Engine specifications

Table 1 Engine specifications		
Engine type	Single cylinder, supercharged,	
	4 stroke, water cooled	
Bore ×	96mm × 108mm	
Stroke		
Volume	781 cc	
Compression	16.4:1	
Ratio		
Gas oil	3 hole injector, Ø0.1mm orifices	
injection		
Injection	Common rail,	
system	electronic injection timing control	

Fuel is supplied to the engine through two routes: gas oil is delivered through the injector and gaseous fuels are mixed into intake charge. Gas oil injection pressure is 40 MPa and injection rate is kept constant at 0.8 mg/cycle in all sessions. This injection rate corresponds to below 2% of total energy supply at all cases. In order to achieve PREMIER combustion, it is essential to use an injector, sprays from which do not cover the entire combustion chamber. prerequisite is met by using a purpose built three-hole injector with orifices of 0.1mm diameter. Orifices are aligned at 120 degrees from each other. With this configuration, sprays can't reach end-gas regions and autoignition can occur when the required conditions are met. Injection timing is adjusted at each session, beginning from TDC and advanced gradually until knocking limit. Gaseous fuel delivery system consists of mass flow controllers and a programmable logic controller in order to keep equivalence ratio by suppressing the effect of instantaneous variations of intake charge flowrate. Flowrates of each gas type is governed for keeping both total equivalence ratio and fraction of each gas in the gaseous fuel mixture constant. In order to stabilize intake pressure, a surge tank is installed between the engine and compressor. Intake temperature is heated to 40°C. Experimental conditions are listed in Table 2. The reasons behind

the selected experimental conditions will be explained in detail in relevant sections.

Table 2 Experimental conditions

Table 2 Experimental conditions	
Pilot fuel injection rate	0.8 mg/cycle
Injection pressure	40 MPa
Equivalence ratio	CH ₄ : 0.52, 0.54, 0.56, 0.58, 0.60 CH ₄ -H ₂ : 0.56
Intake Pressure	CH ₄ : 200 kPa CH ₄ -H ₂ : variable
Hydrogen concentration	0 to 60% with 10% increments
Injection timing	variable

DEFINITION OF PREMIER COMBUSTION

PREmixed Mixture Ignition in the End-gas Region (PREMIER) combustion is a new combustion concept which was observed during previous studies [references here]. Fig. 2 shows pressure histories and rate of heat release (ROHR) of three cycles using 100% CH₄ at equivalence ratio of 0.56. In this graph, ROHR results of the cycle with the latest injection timing, 13 degrees before top dead center (°BTDC) in this case, exhibits expected behavior of regular dual fuel combustion where the first heat release is associated with autoignition of pilot fuel. Following is the second heat release peak where heat release from flame propagation is the highest. At the later stages of this cycle, ROHR drops gradually and diminishes. In contrast, the cycle with the earliest injection timing exhibits a third heat release peak, which is clearly known to be associated with strong end-gas autoignition. This cycle is identified as a knocking cycle because of rapid pressure increase in the cylinder and the fluctuations in the pressure history, both of which are known to damage engines and are avoided in practical applications. The remaining cycle, which is marked as PREMIER, on the other hand, exhibits the third heat release peak without any signs of rapid pressure build-up or pressure fluctuations, which means that there's hardly any evidence of any harmful activity happening in the cylinder based on pressure history; hence it is safe to assume that this is not a knocking cycle. Instead, incylinder pressure is sustained for a longer duration past top dead center. Compared to the knocking cycle, third heat release peak appears later and is weaker. This helps in two ways: the fraction of fuel mixture undergoing autoignition is less, and increase in cylinder volume during expansion stroke is adequate to reduce pressure rise due to autoignition down to safe levels or cancel completely. These characteristics allow utilization of autoignition in the end-gas regions without encountering an problems. This is also accompanied by better performance, lower CO and HC emissions but deteriorated NO_x emissions.

The difference between PREMIER combustion and knocking is measurable. A common quantitative measure of knocking is known as knock intensity [33]. In this study, cylinder pressure is passed through 4kHz-20kHz band pass filter. It is observed that

knocking cycles exhibit two distinct peaks above noise level at 6.5 kHz and 14 kHz. The sum of all instantaneous pressure differences within this range is defined ask knocking intensity (KI). Knocking intensity of regular dual fuel combustion and PREMIER combustion are similar in nature. In this study, KI of 0.1 MPa is defined as noise level, and any cycle below this level is accepted as knock-free cycle.

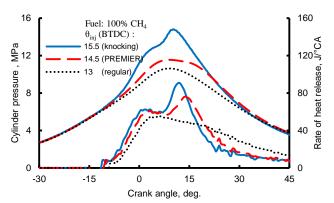


Fig.2 Comparison of knocking, PREMIER combustion and regular dual fuel combustion based on pressure histories and ROHR

RESULTS AND DISCUSSION

PRELIMINARY EXPERIMENTS

In order to understand the characteristics of hydrogen in PREMIER combustion, it is necessary to have a basis for comparison. For this purpose, preliminary experiments are performed using only methane as the gaseous fuel in the intake charge. Knock resistance of methane is known to be suitable for PREMIER combustion and its characteristics are better understood compared to hydrogen during previous studies and [references here]. Another concern of preliminary experiments is to find the optimum equivalence ratio based on efficiency, exhaust emissions and operation stability. In this part of the study, intake pressure is kept at 200 kPa while equivalence ratio is varied between 0.52 and 0.60. Of course, higher equivalence ratio also means higher engine output, but the main concern is to operate the engine with as lean mixture as possible without any stability or efficiency comprise and reasonable exhaust emissions.

Pressure history and ROHR results of methane only experiments are given in Fig.3. The results indicate that both maximum cylinder pressure and peak value of ROHR is higher for leaner operating conditions. In order to reach knocking limits, injection timing is advanced at each session; therefore start of ignition is earlier for leaner conditions and this will trigger end gas autoignition at an earlier timing. Especially the maximum values of pressures at equivalence ratio of 0.52 and 0.54 exceed well above 12 MPa and the effect of end-gas autoignition is detectable on pressure history. On the other hand, maximum pressure at equivalence ratios of 0.58 and 0.60 are at the lower end, but ROHR results show weaker heat release from end-gas autoignition compared to leaner cases. The remaining case, equivalence ratio of 0.56

displays a moderate peak cylinder pressure and heat release from the end-gas region is still similar to those of leaner operating conditions.

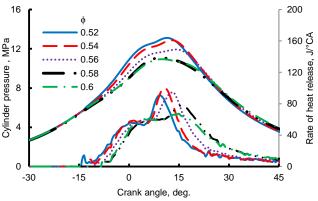


Fig.3.Pressure history and ROHR results of preliminary experiments

Indicated mean effective pressure (Pmi) and thermal efficiency (η_{th}) are two important indicators of efficiency and coefficient of variation of (COV(Pmi)) informs about operation stability. These parameters are presented in Fig.4. The values given in the Figure belong to the condition where highest efficiency value is observed, which are also at the earliest injection timing possible without knocking. Experiments with richer intake mixtures are expected to yield higher engine outputs; therefore the increase in P_{mi} in this Figure is an expected outcome. η_{th} on the other hand, is observed to show decreasing trend for richer mixtures. These two findings once again indicate that equivalence ratio of 0.56 is the optimum operating condition. COV(Pmi) is below 5% in all five cases, and are below 2% for the two leanest conditions. Since COV(P_{mi}) is not high enough in any of these cases to conclude instability, this parameter does not inform about optimum operating condition.

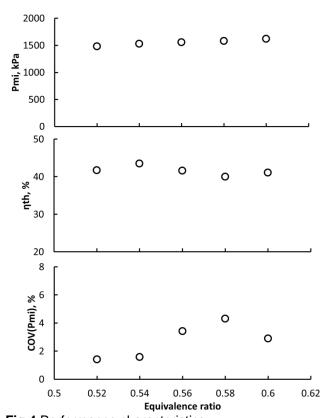


Fig.4 Performance characteristics

A final consideration for identifying optimum operating condition is exhaust emission characteristics. NOx, CO and HC emissions are given at the same operating conditions as before in Fig.5. NOx production rate is an important parameter for the engines that utilize autoignition, because cylinder temperature, hence temperature, is preserved at an elevated level for a long duration. Comparing the given conditions, NO_x production rate increases as intake mixture becomes richer. Especially NO_x emission at the richest case displays an abrupt increase, which should be avoided. Excluding the leanest operating condition, results for all the cases are similar and are between 600-700 ppm range. However CO and HC emission results don't show indicate notable difference between these operating conditions. The highest and lowest values are recorded as 693 ppm and 607 ppm for CO emissions and 2157 ppm and 1874 ppm for HC emissions with equivalence ratios of 0.54 and 0.60 respectively. While these results don't hint at any significant benefit related to operation with equivalence ratio of 0.56, they are going to be used as comparison basis on the next section of this study.

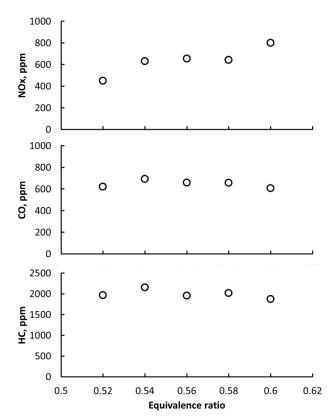
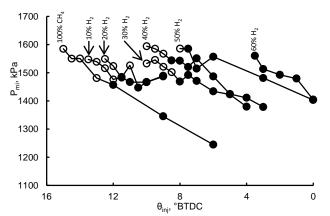


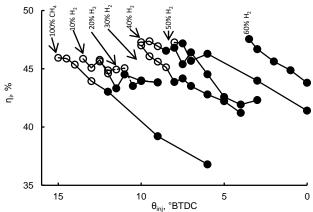
Fig.5 Exhaust emissions of 100% methane experiments

Summing up the aforementioned observations, it is found out that equivalence ratio of 0.56 is the optimum operation condition, at which 2.6 kJ energy is supplied at each cycle. This operating condition becomes the comparison basis for CH₄-H₂ mixture experiments. Due to the differences in heating values and combustion reaction stoichiometry of these two gaseous fuels, it is decided to keep energy supply rate and equivalence ratio constant while controlling intake air flowrate by changing intake pressure as required. The lowest intake pressure in this study was 193 kPa.

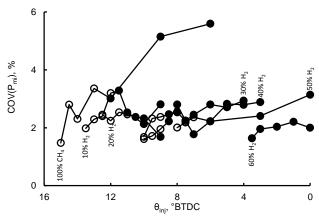
 P_{mi} , η_{th} and $COV(P_{mi})$ results of hydrogen mixtures are shown and compared to those of 100% CH₄ in Fig.6. From this figure onwards, markers with black fillings will represent regular dual fuel operation and white fillings will represent PREMIER combustion operation. Beginning with Pmi, the first noticeable behavior is that maximum values attained in 100% CH₄, 60%CH₄-40%H₂ and 50%CH₄-50%H₂ sessions are very close to each other between 1500 kPa and 1600 kPa of P_{mi} range while rest of the sessions produced poorer results. The highest P_{mi} value is recorded with 40% H₂ in the fuel mixture as 1594.1 kPa compared to 1572.6 kPa with 100% CH₄. While P_{mi} characteristics are similar, there is a noticeable increase in thermal efficiency as the fraction of hydrogen increases in the gaseous fuel. This may be confusing because total energy input is preserved but the difference between pressure histories and ROHR results explains underlying reasons, which are given in Fig.7. First of all, maximum attainable cylinder pressure drops as the fraction of hydrogen increases, which means that engine does less work during compression stroke. Second, end-gas autoignition peaks are detected at cycle timings, based on crank angle degrees, very close to each other between 13°CA and 17°CA. Furthermore, ROHR peaks of flame propagation are higher than that of pure methane and they occur later in the cycle after top dead center timing in all methane-hydrogen mixture experiments. These two ROHR characteristics also suggest that combustion of hydrogen occurs at a faster rate and probably results as better combustion efficiency, which requires further investigation. Finally, COV(P_{mi}) results suggest that addition of hydrogen reduces cyclic variations and improves operation stability. The least COV(Pmi) observed with each fuel mixture is below 2%, including pure methane, but this value never exceeded 4% with any of the hydrogen mixtures no matter how retarded injection timing was, but pure methane displays a noticeable deterioration of COV(P_{mi}) as injection timing is retarded. In contrast to all these observations, 40%CH₄-60%H₂ mixture does not follow this scenario: pressure history shows a profound increase around 15 degrees after top dead center and the maximum pressure is lower than rest of the cases. In addition, ROHR graph does not have a third heat release peak. In this case, combustion properties of hydrogen are expected to be the dominant factor.



(a) Indicated mean effective pressure



(b) Thermal Efficiency



(c) Coefficient of variation of indicated mean effective pressure

Fig.6 Comparison of performance characteristics of methane-hydrogen mixture

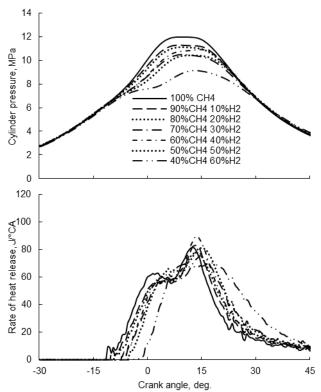
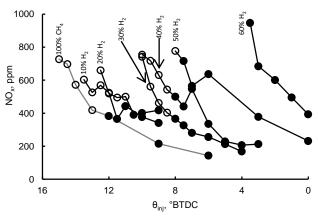


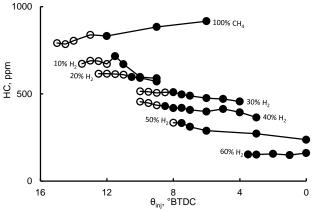
Fig.7 Pressure and rate of heat release of methane-hydrogen mixture gases at highest P_{mi} conditions

Exhaust emissions are given in Fig.8. NO_x emission results show that addition of hydrogen does not introduce any deterioration, below 50% H_2 in the

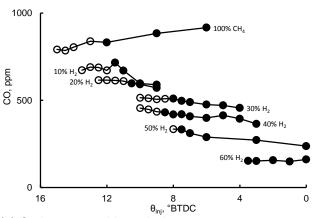
mixture limit. There is only one value that is close to 1000 ppm range which occurs with 60% H₂ in the mixture which is neither PREMIER combustion nor knocking operation. The limit between knocking and non-knocking operation is vague for this mixture due to superknocking, which is presented in Fig.11. Next, looking at CO and HC emissions, it can be seen that both show a systematic decrease as the fraction of hydrogen in the mixture is increased. It is obvious that substituting hydrocarbon fuels with hydrogen is the reason for this outcome; However, the difference between the behaviors of 100% CH₄ and mixtures is interesting: with earlier injection timings, these two emissions noticeably improve with pure methane but stay almost constant for CH₄-H₂ mixtures, which shows that adding hydrogen into the mixture is beneficial for decreasing these two emissions.



(a) NO_x emissions

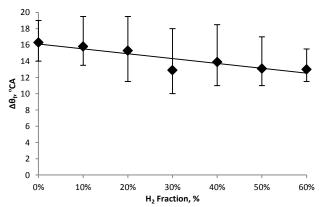


(b) Unburned Hydrocarbon emissions

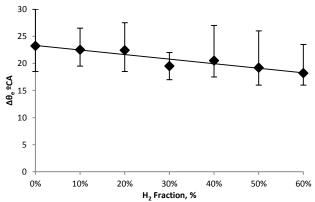


(c) Carbon monoxide emissions

Fig.8 Comparison of exhaust emissions of methanehydrogen mixtures



(a) Duration from injection to flame propagation heat release peak

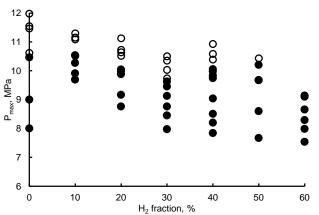


(b) Duration from injection to end-gas autoignition heat release peak

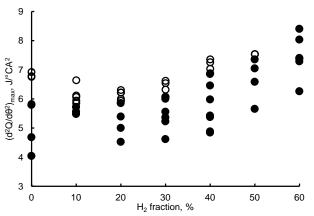
Fig.9 Durations from injection to heat release peaks

It is shown in Fig.6 and Fig.8 that knocking operation occurs with earlier injection timings when the fraction of hydrogen is higher in the mixture. In addition, Fig.7 showed that end-gas autoignition peaks occur around regardless of mixture composition, excluding the most hydrogen rich mixture. It is also observed that the peak value of both flame propagation and end-gas autoignition heat release occurs earlier when hydrogen concentration is increased. This behavior is given in Fig.9. In this figure, $\Delta\theta_f$ refers to the duration in degrees of crank angle from pilot fuel injection to the time that second heat release peak is observed, and $\Delta\theta_e$ refers to the time from pilot fuel injection to the time that end-gas autoignition heat release peak is observed. The error bars attached to the data points indicate the longest and shortest durations of these two parameters instead of standard deviations. These observations indicate that heat release due to propagating flames is faster when more hydrogen is available in the mixture. Also, faster flame propagation helps end-gas autoignition happen faster. In order to explain this phenomenon further, maximum cylinder pressures, the fastest increase rate of ROHR and highest pressure rise rates ((dP/dθ)_{max}) are given in Fig.10. First of all, maximum attainable in-cylinder pressures are lower for the fuel mixtures with higher hydrogen content. It may be discussed whether decreasing intake pressure for higher hydrogen concentration might have significant contribution in this outcome; the difference of intake pressures between 100%CH₄ and 40%CH₄-60%H₂

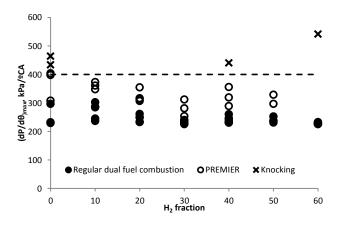
whereas the difference between maximum attainable cylinder pressures is 2.83 MPa. Second, the first derivative of ROHR shows significant increase as fraction of hydrogen increases in the mixture. Please note that these values were observed while ROHR is reaching at the second peak, therefore is related to flame propagation rather than autoignition, so it is an indicator of how quickly end-gas autoignition be satisfied. Combining can observations on these two parameters inform that increasing the hydrogen content in the mixture increases speed of flame propagation; it is wellknown that ignition delay times of fuels become shorter when the temperature is increased, which means that cylinder pressure needs to be kept lower in order to maintain end-gas mixture temperatures at a lower level, so that the mass of end-gas region that undergoes autoignition is not large enough to cause knocking. Finally, maximum pressure rise rates show that, while the values recorded during PREMIER combustion are higher than those of regular dual fuel combustion, they are still below 400 kPa/°CA. In addition, methane-hydrogen mixtures have lower maximum pressure rise rates than pure methane. Any value above this pressure rise rate is observed to display knocking intensities higher than 0.1 MPa; therefore they are marked as knocking operation. which are shown as cross marks in the figure. It is reported in the literature that knocking is observed when pressure rise rate is above a certain value [34. 35]. This limit is observed to be 400 kPa/°CA in current study, which is in agreement with the conclusion of the study that has similar operating conditions [34].



(a) Maximum in-cylinder pressure



(b) Maximum of rate of change of ROHR



(c) Maximum of pressure rise rates

Fig.10 Maximum in-cylinder pressures and maximum rate of change of ROHR

Operation range of PREMIER combustion is defined as the interval of injection timing with which the third heat release peak is achievable. PREMIER combustion operation range is given in Fig.11. When the engine is operated with pure methane, PREMIER combustion is observed within all operation conditions injection timings between 13°BTDC and 15°BTDC. This range narrows down to 1.5°CA range for fuel mixtures of 10% to 30% hydrogen content, 1°CA for mixture of 40% H₂ content and down to a single operating condition at 8°BTDC for 50%. Injection timings are also retarded when fraction of hydrogen is higher. Eventually, **PREMIER** combustion could not be achieved when hydrogen concentration is 60% or more in the gaseous fuel mixture and this is named as critical hydrogen concentration. Critical hydrogen concentration of 60% is important, because the regular dual fuel operation turns to knocking abruptly, and on top of that, superknocking was observed without any indication of underlying reason, and this observation is given in fig. 12. As it can be seen in the figure, heat release occurs prematurely, prior to pilot fuel injection, which is commonly named as superknocking. Usually engine operation is expected to turn from knock-free operation to light knocking operation while the strength of knocking is expected to intensify as injection timing is further advanced; however 40%CH₄-60%H₂ mixture did not exhibit the expected behavior. As a matter of fact, knocking happened before injection timing, and also engine was operating at a temperature below the stable operation temperature; therefore hot spots inside the cylinder, the amount of leftover exhaust gas inside the cylinder due to cyclic variations or a combination of these two with other minor factors are can be suggested as underlying reasons. Nevertheless, this case could not be investigated further because the maximum pressure reached during the experiments was well above recommended limits for the experimental engine. Further analyses need to be conducted in future studies.

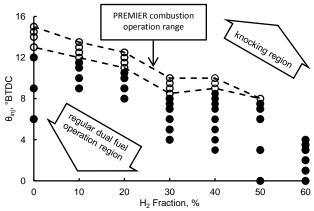


Fig.11 Operation range of PREMIER combustion with Methane-Hydrogen mixtures

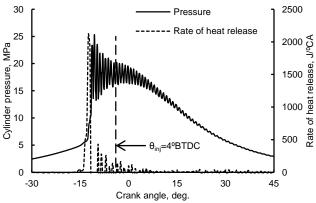


Fig.12 Superknocking observed during 40%CH₄-60%H₂ mixture experiments

CONCLUSIONS

In this study, a gas engine is operated using pure methane and various methane-hydrogen mixtures in a dual fuel gas engine. The results showed that

- 1. Addition of hydrogen into methane improves engine performance. Maximum attainable indicated mean effective pressure does not change significantly with addition of hydrogen, but thermal efficiency is improved. This result is more emphasized at PREMIER combustion points. This is because combustion of hydrogen is faster, so heat release is happening faster as well at both flame propagation and end-gas autoignition situations. In addition, combustion is closer to completeness compared to pure methane.
- 2. Presence of hydrogen in the fuel mixture reduces cyclic variations when end-gas autoignition is weak or inexistent, but has no distinguishable benefit when PREMIER combustion is achieved.
- Addition of hydrogen improves carbon monoxide and unburned hydrocarbon emissions systematically. This is because of reduced amount of hydrocarbon based fuel in the cylinder mixture. In addition, NOx emissions are not affected by hydrogen concentration.
- 4. Knocking resistance of hydrogen mixtures are strongly influenced by flame propagation speed and associated heat release. Mixtures with higher hydrogen concentrations require more retarded pilot fuel injection but both second heat release peak and end-gas autoignition heat release peak

- are observed to happen faster when hydrogen concentration is increased.
- 5. Operation range of PREMIER combustion becomes narrower when hydrogen concentration in methane-hydrogen mixture is increased. At a certain upper hydrogen concentration limit, engine can't be operated with PREMIER combustion because engine operation mode switches from regular dual fuel combustion to light knocking without displaying PREMIER combustion behavior. This limit is observed to be 60% hydrogen in methane-hydrogen mixture under given conditions.
- 6. Superknocking restricts the maximum fraction of hydrogen that can be used in methane-hydrogen mixtures. This outcome is due to deteriorating knock resistance of methane-hydrogen mixtures with increasing hydrogen composition. This coincides with the hydrogen concentration limit at which PREMIER combustion becomes unachievable.

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