**Proceedings of the ASME 2023
ICE Forward Conference
ICEF2023
October 8-11, 2023, Pittsburgh, Pennsylvania**

ICEF2023-110538

COMBUSTION PERFORMANCE AND EMISSIONS CHARACTERIZATION OF METHANE-HYDROGEN BLENDS (UP TO 50% BY VOL.) IN A SPARK-IGNITED CFR ENGINE

Jason Loprete[1,2], Rodrigo Ristow Hadlich[1,2], Amanda Sirna[1,2], Amr Shaalan[1,2], Dimitris Assanis[1,2,*]

[1]Stony Brook University, Stony Brook, NY
[2]Advanced Energy Research and Technology Center, Stony Brook, NY

ABSTRACT

Finding alternatives to carbonaceous fuels has become a prime focus to support the global goal of decarbonizing the transportation sector. Hydrogen is a promising alternative fuel given that it is lightweight, and its combustion produces zero carbon dioxide. However, the absence of widespread dedicated hydrogen infrastructure for transportation, distribution and storage greatly limit its potential as the sole source of fuel for future vehicles. Instead, hydrogen can be readily blended with existing low-carbon fuels, such as natural gas, to enhance overall transportability and additionally improve combustion characteristics of operating devices. This manuscript explores the performance and emissions output of various methane-hydrogen blends, from pure methane to 50% hydrogen, by vol., performed over a range of fuel-air equivalence ratios ranging from the lean misfire limit to ϕ =1.2. Tests were performed on a single-cylinder spark-ignited Cooperative Fuel Research (CFR) engine at a fixed compression ratio and engine speed. Even with hydrogen's lower energy density, initial results show an ability to maintain the same output as the pure methane case over several different blend ratios. A two-phase heat release was observed and attributed to the differences in methane and hydrogen's reactivity. Notably, peak cylinder temperatures and NOx emissions increased as hydrogen blend ratio increased. Lower overall CO emissions were observed with increasing hydrogen blend ratio as the combustion efficiency increased with increasing hydrogen blend percentage. CO_2 emissions were observed to decrease in proportion to the amount of carbon displacement in the fuel blend as the hydrogen blend percentage increased. Overall, using hydrogen as a combustion-enhancer for low-carbon fuels is a viable option as both combustion performance improves and emissions are reduced – all while resolving the significant infrastructure issues pure hydrogen faces.

Keywords: Hydrogen, Methane, Spark Ignition, Emissions

*Corresponding author: dimitris.assanis@stonybrook.edu

NOMENCLATURE

Greek letters:
ϕ Fuel-air equivalence ratio.

English terminology:
CA50 Crank angle position where 50% of the fuel has burned.
PFI Port fuel injection
DI Direct Injection
IMEPn Net Indicated Mean Effective Pressure
IVO Intake Valve Opening
IVC Intake Valve Closing
EVO Exhaust Valve Opening
EVC Exhaust Valve Closing
MBT Maximum Brake Torque
COV Coefficient of Variation
HRR Heat Release Rate
GWP Global Warming Potential
aTDC After Top Dead Center

INTRODUCTION

In 2020, the transportation sector represented nearly one-third of the greenhouse gas emissions in the United States [1]. Correspondingly, the transportation sector was responsible for 28% of the United State's total energy consumption in 2021, of which 90% relied on burning carbon-rich fuels [2]. As of 2021, the United States announced new climate change goals to reduce the net greenhouse gas emissions by 50% below 2005 levels by 2030 [3]. With this, finding greener alternative fuels for transportation is paramount to reducing emissions from this area and meeting closely approaching emissions target goals.

Hydrogen is one such alternative fuel due to its ability to be produced renewably using excess solar and wind power generation via water electrolysis. As a result, there is much interest in hydrogen's role as a fuel in internal combustion engines [4]. However, storage and transportation of pure hydrogen is difficult. Successful storage typically requires high-pressure vessels made from materials that cannot chemically interact with hydrogen and often results in poor volumetric energy density [5]. In addition, use of pure hydrogen in engines may be prone to backfire [6, 7], requiring additional control strategies that may prevent hydrogen from being utilized as a drop-in fuel. Combustion of pure hydrogen is also sensitive to injection timing, injection location, and nozzle design, which are found to have a strong impact on hydrogen engine performance and stability [8, 9], thus making it difficult to predict hydrogen's viability for use in existing devices. Instead, hydrogen may be able to be utilized in conjunction with existing fuels to avoid the challenges associated with pure hydrogen.

In terms of blends, several authors have investigated the role of hydrogen as an addition to existing fuels. One group performed an extensive review of the production and use of synthesis gas (syngas), a mixture of hydrogen and carbon monoxide, as a fuel across a variety of different engine architectures and combustion modes [10]. Although the properties of syngas depend on the molar ratio of hydrogen to carbon monoxide, which varies according to source and production process, fuel reforming combined with dual-fuel combustion was found to improve overall system efficiency when compared to diesel combustion. On-board reforming would remove the complexities associated with dual fuel lines, and this technique could be employed across a variety of source fuels, such as methane [11], to improve performance and reduce emissions. Some have studied the combustion performance of pure hydrogen and pure methane separately, finding that pure hydrogen combustion resulted in less cyclic and spatial variability, higher heat flux peaks, and shorter burn duration when compared to pure methane [12], which suggests blending hydrogen with methane may add stability. Others have investigated the usage of pure hydrogen mixed with gasoline and diesel [13, 14]. Increased brake thermal efficiency, shorter burn durations, and lower HC emissions at the expense of higher NOx emissions were found, but hydrogen's low volumetric efficiency was noted to cause reduced power output. In particular, one study compared the addition of hydrogen to gasoline with the addition of hydrogen to methane up to 20 % by mass at stoichiometric conditions in a spark ignition engine across a variety of engine speeds [15]. The study indicated that maximum cylinder pressure and brake mean effective pressure (BMEP) decreased with increasing hydrogen percentage and that the decrease in performance as hydrogen percentage increased was less for the methane base than for the gasoline base. The authors noted competing factors between hydrogen's high flame speed and and its low energy density, and found that hydrogen mass fractions less than 15% for gasoline and 20% for methane caused an increase in peak cylinder temperatures, which is unfavorable. Methane is thus able to incorporate a higher fraction of hydrogen than traditional fuels without incurring detrimental effects, and reduces fuel line design complexity by utilizing only gaseous fuels. Therefore, methane-hydrogen blends show potential as a primary fuel to displace traditional fossil fuels.

Transporting blends of hydrogen and methane-rich fuels has been under recent investigation. In 2021, the United States Department of Energy (DOE) began the HyBlend initiative, which focuses on addressing technical barriers of blending hydrogen into existing natural gas pipeline infrastructure [16–18]. Natural gas is typically between 75–99% methane depending on where it is sourced from [19]. Thus, the HyBlend initiative aims to enable widespread distribution of a hydrogen and methane fuel that will be available for subsequent use. Since natural gas is primarily methane, a focus on blending hydrogen with pure methane is beneficial to remove the variability associated with natural gas, thus isolating the effect of increased hydrogen percentage on overall system performance. The emissions and performance outcomes from blending hydrogen with methane are not straightforward due to competing chemical and thermal effects. For example, hydrogen has the highest laminar flame speed (over 4 times higher than pure methane at $\phi = 1$ [20]). Thus, this can lead to increased burning velocities when hydrogen is blended with methane, and in turn decrease the total burn duration. Shorter burn durations can be phased closer to TDC and produce a pressure-velocity curve that more closely approximates the ideal constant-volume cycle and thus may lead to higher engine thermal efficiency. However, this may also increase peak temperatures, which may act to increase heat transfer losses and decrease thermal efficiency. Blending hydrogen with methane increases the total H:C ratio, lowering fuel-bound carbon emissions, but may increase thermal NOx production if peak temperatures increase. Therefore, there may be some optimum blend ratio beyond which adding hydrogen is no longer beneficial.

There is a wide range of methane-hydrogen blend ratios and equivalence ratios studied in existing literature that have resulted in an initial understanding of the effects of hydrogen addition on combustion performance and related emissions [21–29]. To summarize, most investigations noted that burn duration typically decreased as hydrogen percentage increased when spark timing was held constant, with NOx emissions increasing with increasing hydrogen percentage. This trend of increased NOx production with a larger fraction of the fuel as hydrogen is apparent in studies focusing on low-methane content ($\leq$ 20 %vol.), with the authors finding that increasing the methane content of methane-hydrogen blends reduces NOx emissions [30]. The authors also found that the equivalence ratio was the most important operating variable dictating engine performance due to its effects on combustion temperature. The equivalence ratio thus must be within certain thresholds to prevent backfire and knock, which limits the operation range. The optimum blend ratio where thermal efficiency increases without significant thermal NOx production was noted by several authors to be within the 20-30% hydrogen by volume range, with increasing hydrogen content reducing cycle-to-cycle variations and improving stability, especially under lean conditions [31–33]. Those that included an equivalence ratio sweep have noted that CO_2 and NOx emissions peak near stoichiometric operation and decrease at leaner or richer operating points. Some authors have indicated that as hydrogen percentage increases, peak cylinder pressure increases and shifts towards TDC with CA50 notably advancing [34, 35]. Thermal efficiency was found to initially increase, but eventually began to decrease as the amount of hydrogen added increased. Though the bulk of the available literature focuses on port fuel injection (PFI) engines, studies with direct injection (DI) have noted the same trends as in terms of performance and emissions as in the PFI cases [36]. In addition, advanced imaging techniques showed that the flame propagation speed and uniformity increased with increasing hydrogen percentage, with CFD studies finding larger initial spark kernels with hydrogen as opposed to without [37–39]. Though hydrogen addition has been shown to improve stability and extend lean flammability limits, some studies have noted the possibility of negative impacts on combustion above 40% hydrogen by volume, in which case backfire can occur at equivalence ratios as low as 0.66 [40]. While much research has been performed on methane-hydrogen blends, further characterization using different test specifications allow for a deeper understanding of the use of the blend in spark-ignition engines and the challenges associated with reducing emissions while maintaining performance.

This document studies the effects of increasing the percentage of hydrogen blended into methane across different equivalence ratios, ranging from the lean misfire limit to $\phi = 1.2$, in a single-cylinder spark-ignition engine. Performance metrics, such as thermal efficiency, IMEPn, and emission rates of CO_2, CO, and NOx, will be evaluated and compared to a baseline case of pure methane. These metrics will then give insight into what would be an optimal blend ratio for usage in the transportation sector in spark-ignition internal combustion engines.

METHODOLOGY

The experimental test cell has been described in detail in previous studies, capable of incorporating both gaseous and liquid fuels [41, 42]. In summary, the experimental facility utilizes a single-cylinder Cooperative Fuel Research (CFR) engine with adjustable compression ratio (6:1 to 18:1) whose experimental parameters are included in Table 1. The existing test cell was modified with additional components to make it compatible with introducing hydrogen, shown in Figure 1. These modifications include an additional mass flow controller, a flame arrestor, and a pre-mixing chamber, as well as general upgrades such as an exhaust plenum to reduce exhaust pressure fluctuations. Ignition and spark timing was controlled using a Performance Electronics PE3 Engine Control Unit (ECU). Emissions measurements were recorded using a HORIBA MEXA-7100DEGR. Engine speed was controlled using a Dyne systems DC dynamometer. The oxygen concentration in the exhaust was measured using an ECM LambdaCAN module, which was used to compute the corresponding fuel-air equivalence ratio. Data acquisition was performed using a National Instruments cDAQ. Engine parameters were monitored and adjusted using a custom LabVIEW program.

TABLE 1: SINGLE-CYLINDER CFR ENGINE SPECIFICATIONS

Parameter	Value
Bore	82.6 mm
Stroke	114.3 mm
# Valves	2
Displaced volume	611.7 cm^3
IVO	-343 °aTDC
IVC	-155 °aTDC
Compression ratio	Set to 9
Engine speed	1200 rev/min
EVO	147 °aTDC
EVC	-356 °aTDC

*Valve timings measured at 0.01 in valve lift.

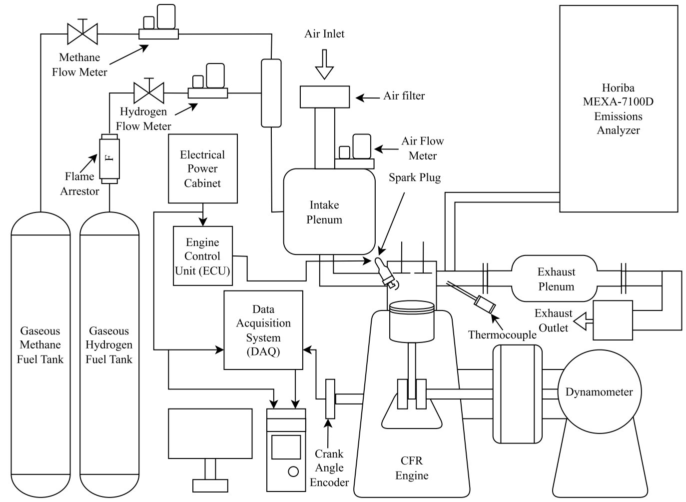

FIGURE 1: SCHEMATIC OF EXPERIMENTAL TEST FACILITY.

Before testing began, the range of equivalence ratios to study was investigated. The lower limit was selected to be the lean misfire limit, where the fuel would be reduced until unstable combustion occurred. The upper limit was selected with safety in mind to avoid unintentional detonation from excessive hydrogen concentrations in the exhaust stream. According to the U.S. Office of Energy Efficiency and Renewable Energy [43], hydrogen has a wide flammability range in air between 4 and 74% by volume, and therefore identifying the operating points which would result in a hydrogen concentration of more than 4% in the exhaust was critical to avoid these unsafe zones. For this reason, combustion simulations were modeled in CHEMKIN-PRO [44] using a 0-D constant-volume closed homogeneous reactor. The chemical kinetics model used is GRI-Mech 3.0 [45] which is an optimized model designed to predict the combustion behavior of natural gas. The domain was initialized with a mixture of air, methane, and hydrogen at prespecified pressure of 3.75 bar and temperature of 1000 K. The equivalence ratio was varied from 0.1 to 2 in steps of 0.05 and the volumetric hydrogen fraction varied from 0 to 1 in steps of 0.01, leading to a total simulation matrix of 4141 cases. The mixture was left to auto-ignite in the chamber and the final mixture composition molar ratios and temperature were recorded. This resulted in the contour map in Figure 2. An equivalence ratio below 1.2 was safe for all blends below 40% hydrogen by volume, with this point being at or beyond the safety limit for blends of 50% and higher, respectively.

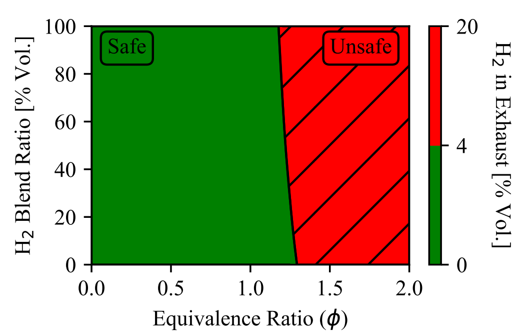

FIGURE 2: H_2 PRESENT IN EXHAUST AT DIFFERENT H_2 FUEL FRACTIONS AND EQUIVALENCE RATIOS.

TABLE 2: MEASUREMENT RANGES AND UNCERTAINTIES

Sensor	Range	Accuracy[a]
Air Alicat Mass Flow Controller MCRW-500SLPM-D/5M	0–500 SLPM	± 0.8% R + 0.2 % F.S.
Methane Alicat Mass Flow Controller MCR-100SLPM-D/5M	0–250 SLPM	± 0.8% R + 0.2 % F.S.
Hydrogen Alicat Mass Flow Controller MCR-50SLPM-D/5M	0-50 SLPM	± 0.8% R + 0.2 % F.S.
Cylinder Pressure Transducer	0-250 bar	≤ ± 0.05% F.S.
Exhaust Pressure Transducer	0-5 bar	≤ ± 0.3% F.S.
Intake Pressure Transducer	0-5 bar	≤ ± 0.5% F.S.
Crank encoder	0-30,000 RPM	± 0.5 bit
Dynamometer	0-4500 RPM	N/A
LambdaCAN Module	ϕ = 0.04 to 2.5	(± 0.6% R at stoichiometric, ± 0.9% R average elsewhere)
Type K Thermocouple	-200 °C to 1250 °C	Greater of 2.2 °C or 0.75 % R

[a]R = Reading, F.S. = Full Scale.

Tests began by setting the compression ratio of the engine to 9 and adjusting the number of carbon and hydrogen atoms present in one mole of the desired fuel blend in both the LambdaCAN controller and the Horiba emissions analyzer. The engine was then motored at 1200 rev/min and supplied with pure methane until stable combustion was reached. Once combustion stabilized, hydrogen was gradually introduced until the desired blend ratio on a volume basis was achieved. Seven different blend ratios were investigated: 0, 10, 20, 25, 30, 40, and 50% Hydrogen by volume. The fuel and air flow rates were gradually adjusted to achieve the desired equivalence ratio and ensure a constant intake pressure of 75 ±0.5 kPa. This intake pressure was chosen to reflect part-load operating conditions, as much of the existing literature utilizes wide open throttle (WOT). Additionally, throttled conditions avoid excessively high pressure rise rates and prevent potential damage to the experimental setup. At each equivalence ratio and blend ratio, a spark timing sweep was performed to identify the maximum brake torque (MBT) timing. MBT timing was selected to ensure blend ratios were compared against their most efficient timings to prevent any bias in timing from impacting the comparison. Data was only collected once the coefficient of variation (COV) in the Net Indicated Mean Effective Pressure (IMEPn) fell below 5% to ensure cyclic variability was minimized. A total of 200 cycles were recorded, with emissions and fuel flow data recorded every 10 cycles due to data acquisition speed limitations. Once the raw data was collected, it was then post-processed in MATLAB to visualize performance and emissions characteristics. This post-processing relied on calculating performance parameters directly from the pressure trace, as well as estimating combustion parameters and heat losses using a modified Woschni heat transfer correlation [46]. The mean value of the parameter of interest was calculated and one standard deviation is utilized as the basis for error bars. For values that rely on ensemble-averaged quantities, the COV of IMEPn was used to estimate the standard deviation in the resulting quantities.

RESULTS AND DISCUSSION

Effects of Blend Ratio on Performance Characteristics

In order for methane-hydrogen blends to be utilized as a drop-in fuel replacement for existing systems, maintaining performance is desirable. A fair assessment of performance can be evaluated on an IMEPn basis that is calculated for the various fuel blends and comparing them back to the baseline of pure methane. As displayed in Figure 3, IMEPn for the different fuel blends differed from baseline by less than 6% for all blend ratios spanning the equivalence ratio space investigated, even as hydrogen percentage was increased. This means that up to 50% Hydrogen had little, if any, negative impacts on work output. The addition of hydrogen led to less cycle-to-cycle variations when the equivalence ratio was between 0.8 and 1.2. It is possible that this is due to the fact that the hydrogen takes up more space in the chamber than methane, and as the mixture becomes increasingly lean, adequate mixing becomes more important to maintain stable combustion. Hydrogen addition was able to extend lean operating range from $\phi \geq 0.6$ to $\phi \geq 0.4$ for blend ratios of up to 50% hydrogen. This increased lean operation may allow for reduced emissions, but the aforementioned importance of mixing becomes even more important as the lean range is extended.

In terms of combustion control, MBT spark timing was retarded as more hydrogen was introduced, as in Figure 4. This

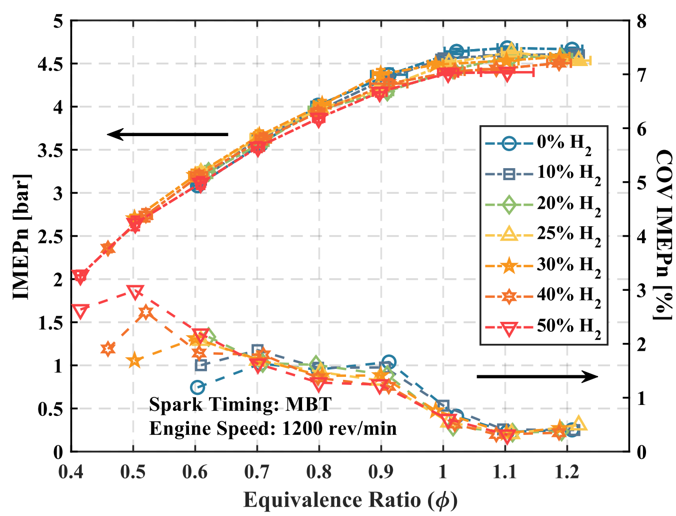

FIGURE 3: NET INDICATED MEAN EFFECTIVE PRESSURE ($IMEP_N$) AND ITS COEFFICIENT OF VARIATION (COV) AS A FUNCTION OF EQUIVALENCE RATIO ACROSS DIFFERENT BLEND RATIOS.

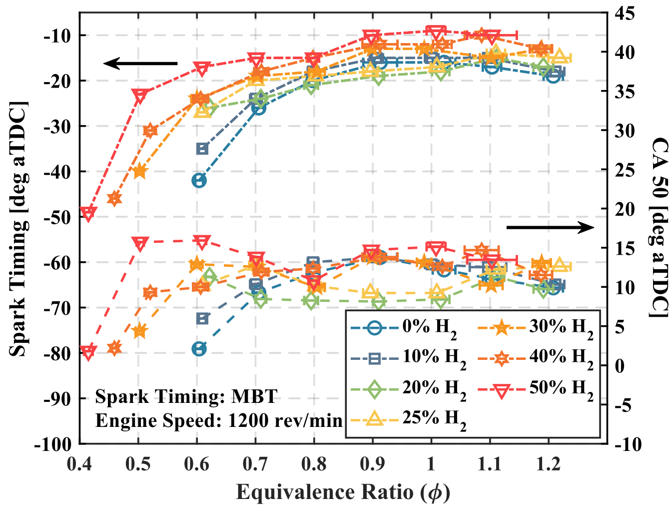

FIGURE 4: MAXIMUM BREAK TORQUE (MBT) SPARK TIMING AND CA50 AS A FUNCTION OF EQUIVALENCE RATIO ACROSS DIFFERENT BLEND RATIOS.

experimentally determined trend of MBT timing retarding, closer to TDC, with increasing H_2 fraction is expected. Hydrogen's higher flame speed results in higher burning velocities, meaning that the combustion event advances as hydrogen percentage is increased at constant spark timings. This results in a larger portion of energy release during the compression stroke effectively increasing the required compression work as work is being done against the piston trajectory. To combat this, the combustion event must be phased later in the cycle, by retarding spark timing, and minimizing the additional compression work to result in the highest possible work output. The trend suggests that as hydrogen is added, the optimal spark timing under lean conditions approaches that of stoichiometric and rich conditions. Thus, less spark advance is required to achieve stable combustion under very lean operating modes. As a result of this, the location of CA50 shifted as well, with the overall spread of CA50 values across equivalence ratio space decreasing with increasing hydrogen fraction. This allows for a constant spark timing to be applicable for a wider variety of operating modes.

The net indicated thermal efficiency, shown in Figure 5, was at a minimum at stoichiometric conditions for all blends under typical operating ranges. The addition of hydrogen tended to lower thermal efficiency at MBT timing conditions, which is not noted in existing literature that tends to focus at WOT conditions with constant spark timing. This is likely due to increased heat transfer losses as a result of higher peak cylinder temperatures, as depicted in Figure 6. Hydrogen addition increased peak temperatures when ϕ was greater than 0.6, but reduced the peak temperature at equivalence ratios below 0.6. There are two competing effects to consider, air dilution and fuel effects stemming from the relative volume occupied by the blend constituents and their respective volumetric energy density. The air dilution effect is well understood as the amount of excess air is increasing and thus the relative fuel energy content is decreasing resulting in lower peak temperatures with decreasing equivalence ratio. The secondary fuel effect is responsible for the inflection point observed at ϕ=0.6 that inverts the trend of increased peak temperature with increasing hydrogen fuel content. To maintain constant energy content

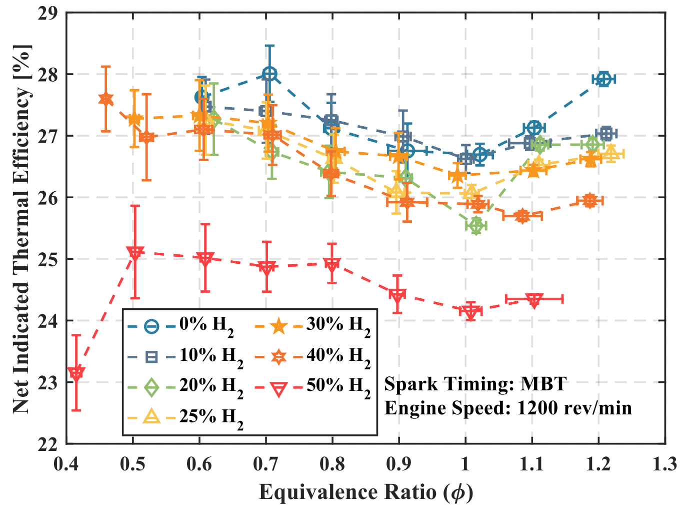

FIGURE 5: NET INDICATED THERMAL EFFICIENCY AS A FUNCTION OF EQUIVALENCE RATIO ACROSS DIFFERENT BLEND RATIOS.

per cycle, a larger fraction of the total cylinder volume is replaced with hydrogen as the hydrogen blend percentage increases. Due to the lower energy density of this larger volume, there is less volumetric energy release per unit of fuel charge volume and thus lower peak temperatures are achieved. Observing this trend closely, there is a smooth transition that continues from ϕ=1.0 all the way to the inflection point of ϕ=0.6, steadily decreasing the peak temperature addition that is contributed by the increased hydrogen content. Aside from peak temperatures, Figure 6 also shows the total burn duration at MBT shifted downward, then upward with increasing hydrogen percentage. While hydrogen addition is known to reduce burn duration, this has only been shown for constant spark timing, which was not utilized in this study. The longer burn duration at 50% hydrogen by volume and high peak temperatures appear to be responsible for the observed decrease in thermal efficiency at this blend ratio. This conjecture is further supported through the estimation of the heat transfer losses per cycle, as depicted in Figure 7, which are increased for the 50% blend case.

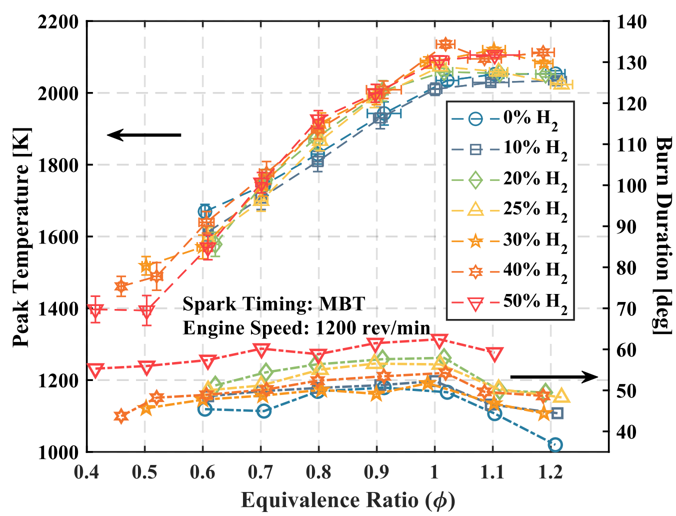

FIGURE 6: PEAK TEMPERATURE AS A FUNCTION OF EQUIVALENCE RATIO ACROSS DIFFERENT BLEND RATIOS.

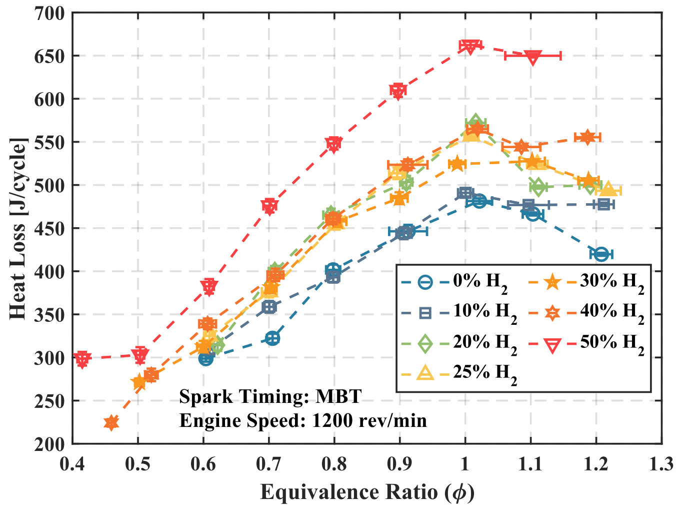

FIGURE 7: TOTAL HEAT LOSS PER CYCLE ESTIMATED FROM A MODIFIED WOSCHNI HEAT TRANSFER CORRELATION.

The combustion efficiency, depicted in Figure 8, was calculated using a correlation between the fuel flow rate, fuel heating value, and measured exhaust gas concentrations of incomplete combustion products. The combustion efficiency peaked during moderately lean operation and dropped significantly during rich operation. The combustion efficiency was largely unchanged with the addition of hydrogen at the same equivalence ratio, with differences of the order of fractions of a percent. However, the extension of the lean misfire limit shows a downward trend under very lean conditions, with combustion efficiency decreasing by several percent. While hydrogen addition under the very lean cases still acts to slightly improve combustion efficiency, the mixture is likely too dilute to sustain combustion and fully consume the fuel. Together, the combustion and thermal efficiency can be used to find the net indicated fuel conversion efficiency.

Lean operation is attractive due to its potential effects on emissions reduction. To further understand lean methane-hydrogen combustion, the apparent gross heat release rate and mass fraction burned curves for $\phi = 0.7$ are utilized and shown in Figure 9. Since MBT spark timing was utilized, the start of combustion differs for each case, hence the misalignment of the heat release rate curves. There exists a two-phase heat release profile whose initial peak becomes more pronounced with increased hydrogen percentage. This may indicate that the fuel blend is not well mixed spatially in the chamber or may be preferentially locally igniting. The heat release analysis assume a well blended fuel, so the initial observed local peak may actually be attributed to the more reactive hydrogen. If this truly indicative of poor mixing of the fuel blend, then this has implications on the usage of the fuel in existing systems and sufficient mixing criteria needs to be established. These results can be utilized to validate Wiebe function parameters for hydrogen-containing fuels [47]. Further study of lean methane-hydrogen mixtures is required to fully understand the cause of this observed two-phase heat release trend.

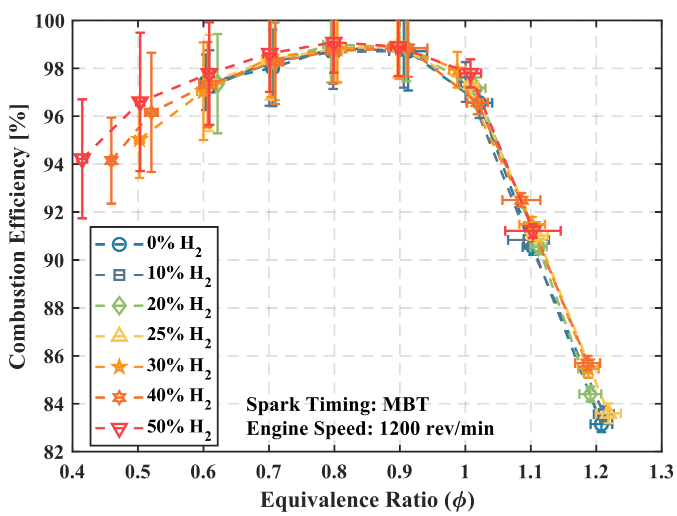

FIGURE 8: COMBUSTION EFFICIENCY AS A FUNCTION OF EQUIVALENCE RATIO ACROSS DIFFERENT BLEND RATIOS.

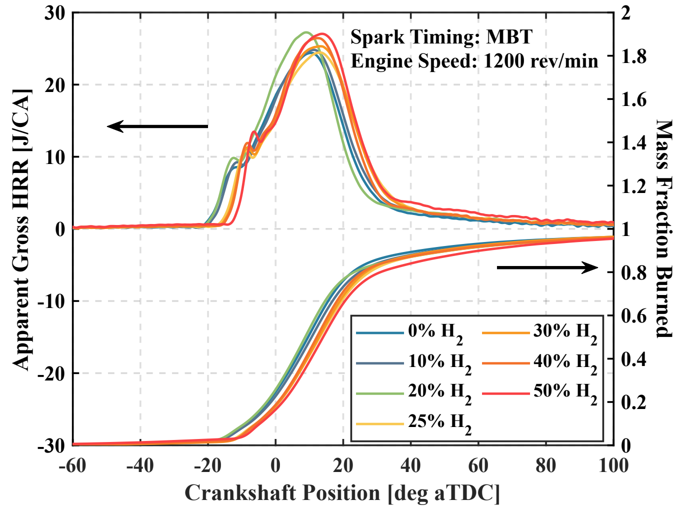

FIGURE 9: APPARENT GROSS HEAT RELEASE RATE AND MASS FRACTION BURNED CURVES ACROSS DIFFERENT BLEND RATIOS FOR ϕ = 0.7.

Effects of Blend Ratio on Emissions

Perhaps the most obvious effect of displacing hydrocarbon fuels with hydrogen is the decrease in CO and CO_2 emissions due to less fuel-bound carbon. The net indicated specific CO_2 emissions, shown in Figure 10, strictly decreases with increasing hydrogen percentage. The degree of reduction grew as operation became leaner, with a reduction of 22.5% at a hydrogen blend ratio of 50% for $\phi = 0.6$. Hydrogen addition continually decreased CO emissions by up to 20.6% at the richest case considered, but had no significant impact on stoichiometric or lean conditions. Futhermore, THC emissions broadly decreased with increasing hydrogen percentage, as shown in Figure 11. This is particularly important due to the other fuel component, methane, which has a global warming potential of 84 on a 20-yr time horizon [48]. If a large amount of methane went unburned, it would have the potential to offset any gains in reduced CO/CO_2 emissions.

Other pollutants, such as NOx, must also be considered especially as combustion temperatures increase. NOx emissions increased substantially as hydrogen percentage increased, with peak emissions at $\phi = 0.9$ being above the detectable limit of the emissions sampling system as depicted in Figure 12. This indicates an increase of at least 50% above the pure methane case,

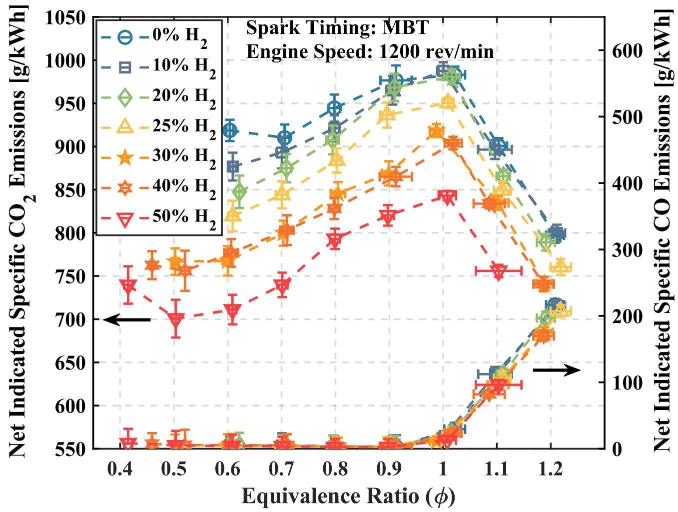

FIGURE 10: NET INDICATED SPECIFIC CO_2 EMISSIONS AND CO EMISSIONS AS A FUNCTION OF EQUIVALENCE RATIO ACROSS DIFFERENT BLEND RATIOS.

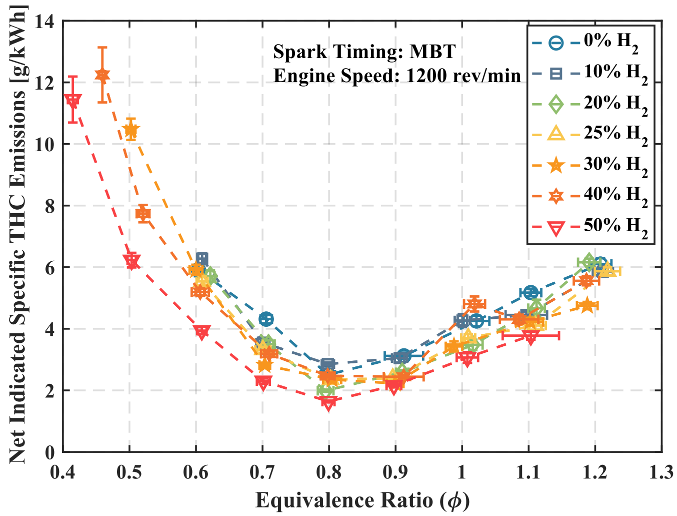

FIGURE 11: THC EMISSIONS INDEX AS A FUNCTION OF EQUIVALENCE RATIO ACROSS DIFFERENT BLEND RATIOS.

which is alarming if this operating condition is to be utilized without alterations to aftertreatment devices. This peak at slight lean conditions is a combination of elevated peak temperatures and higher nitrogen/oxygen concentrations, which encourages the thermal NOx production mechanism. Since NOx has a 20-year global warming potential (GWP) approximately 30 times that of CO_2 [49], this increase far outweighs any benefit of reduced CO_2 emissions at this operating point. However, this can be mitigated by extremely lean ($\phi \leq 0.7$) or rich operation, which results in NOx emissions at or below the baseline methane case for the highest blend ratios considered. In the extremely lean cases, although more nitrogen/oxygen is present, the additional air acts as a heat sink and reduces peak combustion temperatures. As a result, the low temperature results in lower forward reaction rates for the Zeldovich reaction mechanisms [50] and less overall NOx production. On the rich side, although the peak temperature is not significantly lower than that of the stoichiometric case, the nitrogen/oxygen concentrations are extremely low and thus less overall NOx is produced. However, the low combustion efficiency

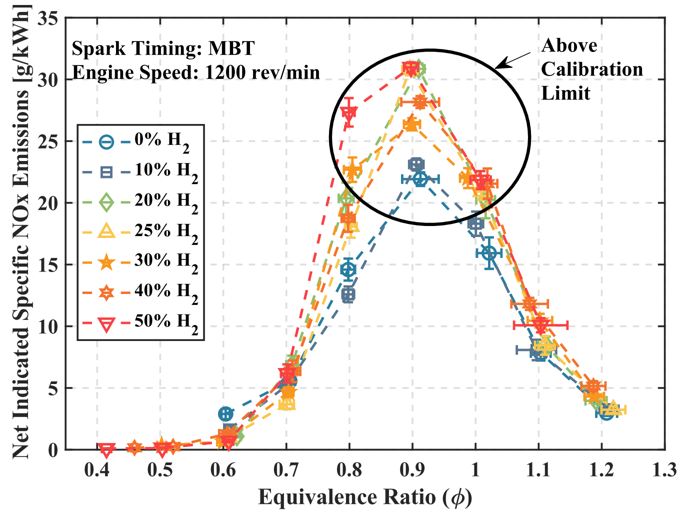

FIGURE 12: NET INDICATED SPECIFIC NOX EMISSIONS AS A FUNCTION OF EQUIVALENCE RATIO ACROSS DIFFERENT BLEND RATIOS.

under rich conditions results in low fuel conversion efficiencies, which makes it difficult to justify rich operation as the definitive solution to high NOx emissions.

Performance vs Emissions Trade-Offs

The addition of hydrogen to methane tends to maintain IMEPn across all equivalence ratios investigated. MBT spark timing across equivalence ratio space differs less and less with increasing hydrogen percentage, with the span of CA50 locations reduced as well. However, NOx emissions peak at $\phi = 0.9$. Due to the higher GWP of NOx compared to CO_2, hydrogen addition above $\phi = 0.7$ and below $\phi = 1.1$ is not recommended without further after treatment technologies or advanced combustion strategies to reduce peak temperatures. This is best displayed by summing the total specific emissions on an equivalent CO_2 emissions basis, as shown in Figure 13. This equivalent basis utilizes the GWP of NOx and CH4 on a 20-yr timescale to estimate the emissions impact of hydrogen addition. From this, it is clear that $\phi \leq 0.7$ results in increasing total emissions reductions with increasing blend ratio. Fuel conversion efficiency is minimized under rich conditions and continually decreases with increasing hydrogen percentage, and so rich operation with hydrogen is not recommended either. Instead, hydrogen addition can be deployed as a means to extend lean-burn operation at throttled conditions. For $\phi \leq 0.7$, the maintained IMEPn, high fuel conversion efficiency, and reduced CO_2, CO, and NOx emissions for all blend ratios relative to pure methane make hydrogen addition an attractive pathway for both improving combustion performance and reducing emissions. With this operating range, hydrogen addition is increasingly beneficial up to 40% by volume.

If higher load is required, $\phi \geq 1.2$ may be suitable as the next-best operating range due to lowered NOx emissions, but is still less favorable due to the near 20% drop in combustion efficiency and dangers associated with hydrogen in the exhaust. Further blend ratios above 30 % should be explored only under very lean conditions to determine optimal combustion strategies, as at near-stoichiometric conditions there is no discernible benefit from the data discussed beyond reducing cycle-to-cycle variations. If sto-

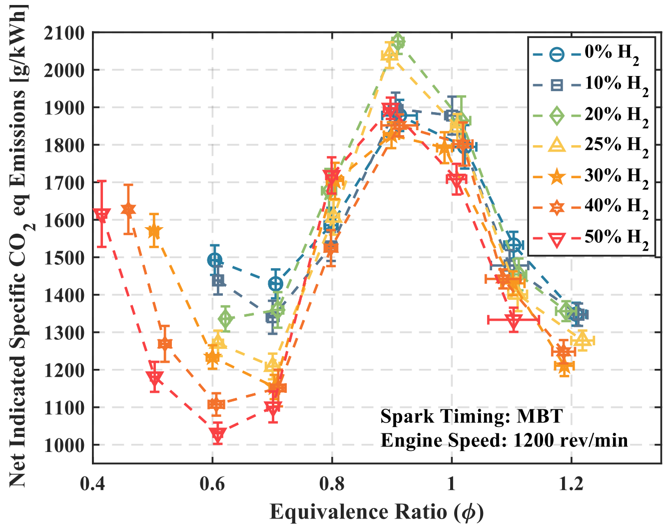

FIGURE 13: EQUIVALENT CO_2 EMISSIONS CONSIDERING CO_2 AND THE GWP OF NOX AND CH4 EMISSIONS AS A FUNCTION OF EQUIVALENCE RATIO ACROSS DIFFERENT BLEND RATIOS.

ichiometric conditions must be maintained for optimal three-way catalyst operation, additional control strategies are required to lower peak temperatures and reduce NOx production. These can be in the form of water injection, as has been performed for other low-temperature combustion fuel blends [51]. However, this may result in further decreases in thermal efficiency and requires further study in the context of methane-hydrogen combustion. Overall, the results indicate that the optimal hydrogen percentage under throttled conditions for spark ignition vehicles is up to 40% by volume, as it results in almost the same emissions reductions without facing the thermal efficiency penalties of the 50% blend.

CONCLUSIONS

A single-cylinder CFR engine was used to determine the effect of increasing hydrogen percentage on the combustion and emissions performance of methane-hydrogen blends. In summary, the key findings of this investigation include:

- Engine load, observed as IMEPn, was maintained to within 6% from baseline across all equivalence ratios investigated even as hydrogen percentage increased to 50% by vol.
- Peak NOx emissions occurred at $\phi = 0.9$ for all blend ratios utilized, peaking at above the detectable limit of instrumentation and indicating an increase of over 50% above the baseline pure methane case.
- Operation between $\phi = 0.6$ - 0.7 or at $\phi \geq 1.2$ is determined to be optimal for emissions reductions on a total net indicated specific emissions output, as operating around $\phi = 0.9$ conditions produced prohibitvly increased NOx emissions.
- Hydrogen addition, at MBT spark timing, resulted in lower engine thermal efficiency and higher heat transfer losses caused by the higher peak cylinder temperatures.
- MBT timing was retarded with increasing hydrogen content, but optimal timings remained closely grouped.
- Two-phase combustion was observed, indicating the potential importance of mixing for methane-hydrogen blends as the hydrogen percentage increases.

ACKNOWLEDGMENTS

This work was enabled by funding received from the Alliance for Sustainable Energy, LLC, Managing and Operating Contractor for the National Renewable Energy Laboratory for the U.S. Department of Energy under Subcontract No. NHQ-9-82305-02, as well as financial support provided by the U.S. Office of Naval Research under Award No. N00014-22-1-2001.

REFERENCES

[1] U.S. Environmental Protection Agency. "Sources of Greenhouse Gas Emissions ." (2022). URL https://www.epa.gov/ghgemissions/sources-greenhouse-gas-emissions.

[2] U.S. Energy Information Administration. "Energy use for transportation." (2022). URL https://www.eia.gov/energyexplained/use-of-energy/transportation.php.

[3] Shouse, Kate C., Lattanzio, Richard K., Legget, Jane A., Procita, Kezee, Ramseur, Jonathan L., Clark, Corrie E., Croft, Genevieve K., Miller, Rena S. and Westphal, Michael I. "U.S. Climate Change Policy." Technical report no. Congressional Research Service (CRS). 2021. URL https://crsreports.congress.gov/product/pdf/R/R46947.

[4] Onorati, A, Payri, R, Vaglieco, BM, Agarwal, AK, Bae, C, Bruneaux, G, Canakci, M, Gavaises, M, Günthner, M, Hasse, C, Kokjohn, S, Kong, S-C, Moriyoshi, Y, Novella, R, Pesyridis, A, Reitz, R, Ryan, T, Wagner, R and Zhao, H. "The role of hydrogen for future internal combustion engines." *International Journal of Engine Research* Vol. 23 No. 4 (2022): pp. 529–540. DOI 10.1177/14680874221081947.

[5] Abdalla, Abdalla M., Hossain, Shahzad, Nisfindy, Ozzan B., Azad, Atia T., Dawood, Mohamed and Azad, Abul K. "Hydrogen production, storage, transportation and key challenges with applications: A review." *Energy Conversion and Management* Vol. 165 (2018): pp. 602–627. DOI 10.1016/j.enconman.2018.03.088.

[6] Dhyani, Vipin and Subramanian, K.A. "Fundamental characterization of backfire in a hydrogen fuelled spark ignition engine using CFD and experiments." *International Journal of Hydrogen Energy* Vol. 44 No. 60 (2019): pp. 32254–32270. DOI 10.1016/j.ijhydene.2019.10.077.

[7] Eicheldinger, Stefan, Karmann, Stephan, Prager, Maximilian and Wachtmeister, Georg. "Optical screening investigations of backfire in a large bore medium speed hydrogen engine." *International Journal of Engine Research* Vol. 23 No. 5 (2022): pp. 893–906. DOI 10.1177/14680874211053171.

[8] Younkins, Matthew, Boyer, Brad and Wooldridge, Margaret. "Hydrogen DI Dual Zone Combustion System." *SAE International Journal of Engines* Vol. 6 No. 1 (2013): pp. 2013–01. DOI 10.4271/2013-01-0230.

[9] Lee, Sanguk, Kim, Gyeonggon and Bae, Choongsik. "Effect of injection and ignition timing on a hydrogen-lean stratified charge combustion engine." *International Journal of*

Engine Research Vol. 23 No. 5 (2022): pp. 816–829. DOI 10.1177/14680874211034682.

[10] Paykani, Amin, Chehrmonavari, Hamed, Tsolakis, Athanasios, Alger, Terry, Northrop, William F. and Reitz, Rolf D. "Synthesis gas as a fuel for internal combustion engines in transportation." *Progress in Energy and Combustion Science* Vol. 90 (2022): p. 100995. DOI 10.1016/j.pecs.2022.100995.

[11] Mitchell, Robert and Kocsis, Michael. "Performance Evaluation of Dedicated EGR on a 12 L Natural Gas Engine." *SAE Technical Paper*. 2019. DOI 10.4271/2019-01-1143.

[12] Demuynck, J., Raes, N., Zuliani, M., De Paepe, M., Sierens, R. and Verhelst, S. "Local heat flux measurements in a hydrogen and methane spark ignition engine with a thermopile sensor." *International Journal of Hydrogen Energy* Vol. 34 No. 24 (2009): pp. 9857–9868. DOI 10.1016/j.ijhydene.2009.10.035.

[13] Apostolescu, Nicolae and Chiriac, Radu. "A Study of Combustion of Hydrogen-Enriched Gasoline in a Spark Ignition Engine." 1996. DOI 10.4271/960603.

[14] Akal, Dinçer, Öztuna, Semiha and Büyükakın, Mustafa Kemalettin. "A review of hydrogen usage in internal combustion engines (gasoline-Lpg-diesel) from combustion performance aspect." *International Journal of Hydrogen Energy* Vol. 45 No. 60 (2020): pp. 35257–35268. DOI 10.1016/j.ijhydene.2020.02.001.

[15] Kamil, Mohammed and Rahman, M.M. "Performance prediction of spark-ignition engine running on gasoline-hydrogen and methane-hydrogen blends." *Applied Energy* Vol. 158 (2015): pp. 556–567. DOI 10.1016/j.apenergy.2015.08.041.

[16] U.S. Department of Energy. "HyBlend: Opportunities for Hydrogen Blending in Natural Gas Pipelines ." (2021). URL https://www.energy.gov/eere/fuelcells/hyblend-opportunities-hydrogen-blending-natural-gas-pipelines.

[17] Erdener, Burcin Cakir, Sergi, Brian, Guerra, Omar J., Lazaro Chueca, Aurelio, Pambour, Kwabena, Brancucci, Carlo and Hodge, Bri-Mathias. "A review of technical and regulatory limits for hydrogen blending in natural gas pipelines." *International Journal of Hydrogen Energy* Vol. 48 No. 14 (2023): pp. 5595–5617. DOI 10.1016/j.ijhydene.2022.10.254.

[18] Mahajan, Devinder, Tan, Kun, Venkatesh, T., Kileti, Pradheep and Clayton, Clive R. "Hydrogen Blending in Gas Pipeline Networks—A Review." *Energies* Vol. 15 No. 10 (2022): p. 3582. DOI 10.3390/en15103582.

[19] SIMON, ANDREW L. "Natural Gas." *Energy Resources*. Elsevier (1975): pp. 67–73. DOI 10.1016/B978-0-08-018750-1.50011-6.

[20] Schefer, Robert W., White, Christopher and Keller, Jay. "Lean Hydrogen Combustion." *Lean Combustion*. Elsevier (2008): pp. 213–VIII. DOI 10.1016/B978-012370619-5.50009-1.

[21] Karim, G.A., Wierzba, I. and Al-Alousi, Y. "Methane-hydrogen mixtures as fuels." *International Journal of Hydrogen Energy* Vol. 21 No. 7 (1996): pp. 625–631. DOI 10.1016/0360-3199(95)00134-4.

[22] AKANSU, S, KAHRAMAN, N and CEPER, B. "Experimental study on a spark ignition engine fuelled by methane–hydrogen mixtures." *International Journal of Hydrogen Energy* Vol. 32 No. 17 (2007): pp. 4279–4284. DOI 10.1016/j.ijhydene.2007.05.034.

[23] Moreno, F., Muñoz, M., Arroyo, J., Magén, O., Monné, C. and Suelves, I. "Efficiency and emissions in a vehicle spark ignition engine fueled with hydrogen and methane blends." *International Journal of Hydrogen Energy* Vol. 37 No. 15 (2012): pp. 11495–11503. DOI 10.1016/j.ijhydene.2012.04.012.

[24] Bundele, Hiresh, Kurien, Caneon, Varma, Penmatsa Sandeep and Mittal, Mayank. "Experimental and computational study on the enhancement of engine characteristics by hydrogen enrichment in a biogas fuelled spark ignition engine." *International Journal of Hydrogen Energy* Vol. 47 No. 71 (2022): pp. 30671–30686. DOI 10.1016/j.ijhydene.2022.07.029.

[25] Ortiz-Imedio, R., Ortiz, A., Urroz, J.C., Diéguez, P.M., Gorri, D., Gandía, L.M. and Ortiz, I. "Comparative performance of coke oven gas, hydrogen and methane in a spark ignition engine." *International Journal of Hydrogen Energy* Vol. 46 No. 33 (2021): pp. 17572–17586. DOI 10.1016/j.ijhydene.2019.12.165.

[26] Ortiz-Imedio, Rafael, Ortiz, Alfredo and Ortiz, Inmaculada. "Comprehensive analysis of the combustion of low carbon fuels (hydrogen, methane and coke oven gas) in a spark ignition engine through CFD modeling." *Energy Conversion and Management* Vol. 251 (2022): p. 114918. DOI 10.1016/j.enconman.2021.114918.

[27] Soltic, Patrik and Hilfiker, Thomas. "Efficiency and raw emission benefits from hydrogen addition to methane in a Prechamber–Equipped engine." *International Journal of Hydrogen Energy* Vol. 45 No. 43 (2020): pp. 23638–23652. DOI 10.1016/j.ijhydene.2020.06.123.

[28] KAHRAMAN, N, CEPER, B, AKANSU, S and AYDIN, K. "Investigation of combustion characteristics and emissions in a spark-ignition engine fuelled with natural gas–hydrogen blends." *International Journal of Hydrogen Energy* Vol. 34 No. 2 (2009): pp. 1026–1034. DOI 10.1016/j.ijhydene.2008.10.075.

[29] Sofianopoulos, A., Assanis, D. N. and Mamalis, S. "Effects of Hydrogen Addition on Automotive Lean-Burn Natural Gas Engines: Critical Review." *Journal of Energy Engineering* Vol. 142 No. 2 (2016). DOI 10.1061/(ASCE)EY.1943-7897.0000319.

[30] Diéguez, P.M., Urroz, J.C., Marcelino-Sádaba, S., Pérez-Ezcurdia, A., Benito-Amurrio, M., Sáinz, D. and Gandía, L.M. "Experimental study of the performance and emission characteristics of an adapted commercial four-cylinder spark ignition engine running on hydrogen–methane mixtures." *Applied Energy* Vol. 113 (2014): pp. 1068–1076. DOI 10.1016/j.apenergy.2013.08.063.

[31] Kosmadakis, G.M., Rakopoulos, D.C. and Rakopoulos, C.D. "Assessing the cyclic-variability of spark-ignition engine running on methane-hydrogen blends with high hydrogen contents of up to 50%." *International Journal of Hy-*

drogen Energy Vol. 46 No. 34 (2021): pp. 17955–17968. DOI 10.1016/j.ijhydene.2021.02.158.

[32] Molina, Santiago, Novella, Ricardo, Gomez-Soriano, Josep and Olcina-Girona, Miguel. "Experimental Evaluation of Methane-Hydrogen Mixtures for Enabling Stable Lean Combustion in Spark-Ignition Engines for Automotive Applications." 2022. DOI 10.4271/2022-01-0471.

[33] Molina, S., Ruiz, S., Gomez-Soriano, J. and Olcina-Girona, M. "Impact of hydrogen substitution for stable lean operation on spark ignition engines fueled by compressed natural gas." *Results in Engineering* Vol. 17 (2023): p. 100799. DOI 10.1016/j.rineng.2022.100799.

[34] Duan, Xiongbo, Li, Yangyang, Liu, Jingping, Guo, Genmiao, Fu, Jianqin, Zhang, Quanchang, Zhang, Shiheng and Liu, Weiqiang. "Experimental study the effects of various compression ratios and spark timing on performance and emission of a lean-burn heavy-duty spark ignition engine fueled with methane gas and hydrogen blends." *Energy* Vol. 169 (2019): pp. 558–571. DOI 10.1016/j.energy.2018.12.029.

[35] Liu, Jingping, Duan, Xiongbo, Yuan, Zhipeng, Liu, Qi and Tang, Qijun. "Experimental study on the performance, combustion and emission characteristics of a high compression ratio heavy-duty spark-ignition engine fuelled with liquefied methane gas and hydrogen blend." *Applied Thermal Engineering* Vol. 124 (2017): pp. 585–594. DOI 10.1016/j.applthermaleng.2017.06.067.

[36] Di Iorio, Silvana, Sementa, Paolo and Vaglieco, Bianca Maria. "Analysis of combustion of methane and hydrogen–methane blends in small DI SI (direct injection spark ignition) engine using advanced diagnostics." *Energy* Vol. 108 (2016): pp. 99–107. DOI 10.1016/j.energy.2015.09.012.

[37] Di Iorio, Silvana, Sementa, Paolo and Vaglieco, Bianca Maria. "Experimental investigation on the combustion process in a spark ignition optically accessible engine fueled with methane/hydrogen blends." *International Journal of Hydrogen Energy* Vol. 39 No. 18 (2014): pp. 9809–9823. DOI 10.1016/j.ijhydene.2014.04.065.

[38] Merola, Simona Silvia, Di Iorio, Silvana, Irimescu, Adrian, Sementa, Paolo and Vaglieco, Bianca Maria. "Spectroscopic characterization of energy transfer and thermal conditions of the flame kernel in a spark ignition engine fueled with methane and hydrogen." *International Journal of Hydrogen Energy* Vol. 42 No. 18 (2017): pp. 13276–13288. DOI 10.1016/j.ijhydene.2017.03.219.

[39] Sun, Xingyu, Liu, Hengli, Duan, Xiongbo, Guo, Hao, Li, Yangyang, Qiao, Junhao, Liu, Qi and Liu, Jingping. "Effect of hydrogen enrichment on the flame propagation, emissions formation and energy balance of the natural gas spark ignition engine." *Fuel* Vol. 307 (2022): p. 121843. DOI 10.1016/j.fuel.2021.121843.

[40] Karmann, Stephan, Eicheldinger, Stefan, Prager, Maximilian and Wachtmeister, Georg. "Optical and thermodynamic investigations of a methane and hydrogen blend fueled large bore engine." *International Journal of Engine Research* Vol. 23 No. 5 (2022): pp. 846–864. DOI 10.1177/14680874211066735.

[41] Ran, Zhongnan, Longtin, Jon and Assanis, Dimitris. "Investigating anode off-gas under spark-ignition combustion for SOFC-ICE hybrid systems." *International Journal of Engine Research* Vol. 23 No. 5 (2022): pp. 830–845. DOI 10.1177/14680874211016987.

[42] Nikiforakis, Ioannis, Ran, Zhongnan, Sprengel, Michael, Brackett, John, Babbitt, Guy and Assanis, Dimitris. "Investigating realistic anode off-gas combustion in SOFC/ICE hybrid systems: mini review and experimental evaluation." *International Journal of Engine Research* Vol. 23 No. 5 (2022): pp. 876–892. DOI 10.1177/14680874211058324.

[43] U.S. Office of Energy Efficiency and Renewable Energy. "Hydrogen Safety." URL https://www1.eere.energy.gov/hydrogenandfuelcells/pdfs/h2_safety_fsheet.pdf.

[44] "Reaction Design." *ANSYS Chemkin Theory Manual*. ANSYS, San Diego (2015).

[45] Smith, Gregory P., Golden, David M., Frenklach, Michael, Moriarty, Nigel W., Eiteneer, Boris, Goldenberg, Mikhail, Bowman, C. Thomas, Hanson, Ronald K., Song, Soonho, Gardiner, William C., Lissianski, Vitali V. and Qin, Zhiwei. "GRI-Mech 3.0." URL http://www.me.berkeley.edu/gri_mech/.

[46] Chang, Junseok, Güralp, Orgun, Filipi, Zoran, Assanis, Dennis N., Kuo, Tang-Wei, Najt, Paul and Rask, Rod. "New Heat Transfer Correlation for an HCCI Engine Derived from Measurements of Instantaneous Surface Heat Flux." 2004. DOI 10.4271/2004-01-2996.

[47] Yang, Ruinan, Ran, Zhongan and Assanis, Dimitris. "Estimation of Wiebe Function Parameters for Syngas and Anode Off-Gas Combustion in Spark-Ignition Engines." *Journal of Engineering for Gas Turbines and Power* Vol. 145 No. 7 (2023). DOI 10.1115/1.4056856.

[48] IEA. "Methane Tracker 2021." (2021). URL https://www.iea.org/reports/methane-tracker-2021/methane-and-climate-change.

[49] Lammel, Gerhard and Graßl, Hartmut. "Greenhouse effect of NOX." *Environmental Science and Pollution Research* Vol. 2 No. 1 (1995): pp. 40–45. DOI 10.1007/BF02987512.

[50] LAVOIE, GEORGE A., HEYWOOD, JOHN B. and KECK, JAMES C. "Experimental and Theoretical Study of Nitric Oxide Formation in Internal Combustion Engines." *Combustion Science and Technology* Vol. 1 No. 4 (1970): pp. 313–326. DOI 10.1080/00102206908952211.

[51] Hountalas, D. T., Mavropoulos, G. C., Zannis, T. C. and Mamalis, S. D. "Use of Water Emulsion and Intake Water Injection as NOx Reduction Techniques for Heavy Duty Diesel Engines." *SAE 2006 World Congress Exhibition*. 2006. SAE International. DOI 10.4271/2006-01-1414.