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Literature review on CNG / H2 mixtures for heavy-duty CNG vehicles

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Literature review on CNG / H_2 Mixtures for heavy-duty CNG vehicles

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ABSTRACT

Sustainably produced hydrogen (H_2) is seen as a low carbon fuel for transportation. Likewise compressed natural gas (CNG) could reduce CO2 emissions from vehicles, especially if it is produced from renewable sources such as biomethane. Being both gaseous, H_2 and CNG can easily form a mixture (H_2/CNG), and the existing gas network could potentially be used for a smooth transition from fossil natural gas to a mixture of renewable CH_4 and H_2, thereby achieving a low carbon energy supply for internal combustion engines (ICEs). Concawe commissioned DNV a literature review to assess the benefits, drawbacks and barriers of using H_2/CNG mixtures in ICEs, with a focus on heavy duty vehicles.

The report reveals that the European heavy-duty CNG vehicle market is moving from lean burn (excess air, the technology that was most used in pre-Euro V vehicles) to stoichiometric engines (current technology of choice in Euro VI vehicles). In general, the literature primarily focusses on the effect of H_2 addition to CNG in lean-burn engines, limited information being available on stoichiometric heavy-duty engines.

When 20% vol. H_2 is mixed with CNG, the literature shows:

- Engine efficiency gains between 0% and 13% in a spark-ignited lean burn engine, strongly dependent on engine parameter settings; in a stoichiometric engine, the efficiency gain is unknown;
- Greenhouse gases emissions reductions between 8% (if no efficiency gain) and 20% (13% efficiency gain)
- Vehicle driving range reductions between 24% (if no efficiency gain) and 14% (13% efficiency gain), due to the unfavourable compressibility factor of H_2 compared to CH_4.

The tolerance to H_2 content in the natural gas grid is heterogeneous in Europe, ranging between 0.1% (Belgium, UK) to up to 10% (Germany, on a case-by-case basis, depending on the grid specificities). In the whole supply chain connected to the gas grid, CNG vehicles are among the end-use applications with the lowest tolerance to H_2 addition (2% vol. max) because the steel tanks (Type 1) used in the legacy fleet may suffer from H_2 embrittlement. For this reason, the H_2 limit of 2% vol. max is fixed in the European standard EN 16723-2 for CNG as automotive fuel. However, new vehicles are no longer concerned by this issue thanks to new tank types made of non-steel composite materials. The report identifies knowledge gaps and research needs regarding the compatibility of engines and retail stations with higher rates of H_2 in CNG which concern the aftertreatment system, the spark plugs, the lubricating oil, the CNG tank and fuel lines, the injectors, the knock management and rating, the engine calibration and the sensors; the effects on engine-out emissions and combustion stability are also listed.

Moreover, the report compares the merits of use of pure H_2 both in fuel cell electric vehicles (FCEVs) and in ICEs. On the one hand, FCEVs show better fuel economy and no pollutant emissions compared to ICEs. But on the other hand, they require a very high degree of purity of H_2 at the retail station and have a higher total cost of ownership (TCO).

KEYWORDS

Compressed Natural Gas, Hydrogen, H_2/CNG, CH_4, H_2, mixture, greenhouse gas emissions, natural gas grid, low carbon pathway, heavy duty vehicle, engine efficiency, pollutant emissions, CNG tank, CNG retail station, aftertreatment, spark plug, lubricating oil, knock, sensor

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SUMMARY

The need to reduce CO_2 emission is a driving force for the use of sustainably produced hydrogen as a transportation fuel. Pure hydrogen can be used as a fuel in fuel cells or, in the near future, in internal combustion engines. Furthermore, hydrogen can be added to CNG in order to achieve a low carbon energy supply. Blending of hydrogen can be done either at the fueling station or by injecting hydrogen in the existing natural gas network. Concawe requested DNV to perform a literature review and analysis to understand the current knowledge base and assess the benefits, drawbacks and barriers of using hydrogen/CNG mixtures in internal combustion engines. Furthermore, a comparison is made between use of pure hydrogen in fuel cells and in internal combustion engines. Although the focus of this study is on CNG heavy-duty vehicles we also address the limits of hydrogen mixture for other end-use equipment that are connected to the gas grid.

1. CNG Heavy-duty vehicles

First, the literature review was performed for the impact of H_2 addition to CNG on heavy duty vehicles supplemented with interviews with relevant stakeholders. The results are summarized below.

The CNG engines present in the market are either lean-burn or stoichiometric engines, which are mainly spark ignited engines ('Otto cycle'). Recent market analyses show a shift from lean burn (excess air) to stoichiometric engines in the European heavy-duty CNG vehicles market to meet the Euro VI regulations. The engine performance of H_2/CNG blends have been studied extensively in the literature. However limited information is available on stoichiometric heavy-duty engines.

The literature review reveals that the main benefit of blending CNG with hydrogen is the reduction of engine-out CO_2, CH_4 and Non-Methane Hydrocarbons (NMHC) emissions. Furthermore, hydrogen addition can increase the fuel efficiency. However, the actual efficiency increase is strongly dependent on the engine parameter settings controlled by the Engine Control Unit (ECU), such as the air/fuel ratio and spark timing. As an example, the gain in fuel efficiency reported in the literature ranges between 0 and13% for an addition of 20 vol% hydrogen in natural gas compared to neat natural gas. An efficiency increase, however, goes together with an increase in in-cylinder peak pressure and temperature resulting in more engine wear and higher engine-out NO_x emission.

Several studies show that the presence of hydrogen in CNG increases the combustion stability (COV) which reduces the unburned hydrocarbon emissions and enables the extension of the lean flammability limit and a higher EGR ratio leading to a significant reduction of NO_x engine out emissions.

Hydrogen mixtures reduce the carbon footprint of engines in three ways. First, less CO_2 is produced per unit of energy input; second, methane slip is reduced; and third, there is a potential increase in engine efficiency (0-13%). When assuming no efficiency gain, the presence of 20 vol.% hydrogen in CH_4 will reduce the greenhouse gas emission (GHG) by about 8 vol.%, while GHG emission reduction of about 20% is obtained when assuming an efficiency gain of 13%. Hydrogen addition to CNG reduces the energy content in the CNG tank at a given pressure level. As a result, the addition of hydrogen to CNG reduces the vehicle range. When 20 vol.% hydrogen is present in natural gas, a vehicle range reduction of about 24% can be expected in the most conservative case (no fuel efficiency gain) while the vehicle range

reduction would be limited to about 14% in best case, i.e., with a 13% fuel efficiency gain.

Regarding billing, it can be noted that the addition of hydrogen to pipeline gas, will result in a change of the energy content per unit of mass (MJ/kg) with a consequent impact on the mass-based billing of CNG. Furthermore, currently installed gas analyser equipment for detecting the gas composition and determining the energy content of the fuel may experience some difficulties when hydrogen is present in CNG.

Engine components present in heavy-duty vehicles can also be impacted by the presence of hydrogen in CNG. Several demonstration projects have been completed or are in progress worldwide, whereby CNG buses have been fuelled with H_2/CNG mixtures. These field experiments revealed vehicles can handle up to 15-20 vol% of hydrogen in the CNG. For higher hydrogen content in CNG (above 15-20 vol %) recalibration of the ECU is required. During these field tests the buses were equipped with dedicated equipment that could handle up to 100% hydrogen. Engine manufacturers indicate that field tests are still needed to investigate the sensitivity of original CNG equipment to the presence of hydrogen.

Knowledge on the impact of the introduction of hydrogen in CNG on the performance and lifetime of the lubrication oil, catalyst and spark plug is currently lacking. The impact of hydrogen is better known for the fuel storage and handling systems. High-strength steel components are susceptible to hydrogen embrittlement. According to current standards CNG steel tanks can handle up to 2 vol% of hydrogen. Replacement of these tanks by, for example, Type-4 (composite) CNG tanks allows up to 100% of hydrogen, on condition that the operating pressure remains constant. However, at present it is still unknown how much hydrogen in CNG is allowed for the tank's ancillary equipment (valves, seals etc.). Depending on the materials, when hydrogen is present in CNG, materials and components of the fuel system, such as the pressure regulator, may have to be upgraded to avoid hydrogen embrittlement. It is still unknow what the maximum hydrogen tolerance is for installed CNG fuel-line components. One manufacturer indicated that former fuel-line components were compatible up to 10 vol.% of hydrogen in the blends, while for current components the maximum admissible hydrogen concentration in CNG is about 30 vol.%. Additionally, the fuel injector's performance will have to be adapted to the differences in sonic speed, density and energy density of the H_2/CNG mixture. Hydrogen addition decreases the energy flow through the fuel injectors and as a result, according to the interviewees, a recalibration of the ECU might be needed.

Hydrogen addition to CNG lowers the knock resistance and thus increases the risk of engine knocking and limits the amount of hydrogen that can be added. In the worst case, when the methane number of CNG is similar to the minimum methane number given in the engine specification, no hydrogen can be added to this CNG. Literature review reveals that almost no engine knock data is available for advanced stoichiometric CNG heavy-duty engines fueled with hydrogen/CNG. The lack of available knock data makes it difficult to determine to what extend existing methods are capable of accurately predict the knock behavior of H_2/CNG blends in stoichiometric heavy-duty engines.

Recommendations

The literature review and interviews revealed that it is still unknown in the industry if the ancillary components of the tank system (seals, valves etc.) and fuel-line components can handle H_2/CNG blends. Therefore, to maximize the hydrogen percentage in CNG for heavy-duty applications, it is necessary to investigate and subsequently develop standards on how much hydrogen these components can handle. An alternative is to replace these components with ones that are hydrogen compliant.

The literature review and interviews also revealed that to date only limited experimental data are present on the effect of hydrogen addition to CNG for heavy-duty stoichiometric vehicles. Since these engines are dominant in today's heavy-duty vehicle market, it is highly recommended to investigate the effect of hydrogen addition on the performance of stoichiometric heavy-duty CNG vehicles on the following topics and summarized in the summary table 1 below.

- The measured knock resistance and compare the results with the different methane number methods available.
- The potential fuel efficiency gain
- Hydrogen tolerance for fuel line components
- Hydrogen tolerance for fuel tanks, including tank periphery
- Effect of CNG/H_2 combustion and combustion products on lube oil degradation
- Combined catalyst and engine performance for resulting tailpipe emissions such as CH_4, NO_x, and NH_3/N_2O
- Spark plug design, selection and possible lifetime-extending operating conditions when using CNG/H_2 mixtures
- Crankcase safety when CNG/H_2 mixtures are used

Whenever CNG is stably delivered with a fixed amount of hydrogen, it is recommended to optimize the engine performance for that specific H_2/CNG fuel blend resulting in a higher fuel efficiency, lower emissions, and a stable combustion process. On the other hand, in case of a variable hydrogen content in the H_2/CNG mixture, an engine mapping should be performed to cover the full range of H_2/CNG blends supplied at the fueling station. However, as an alternative, a feed-forward gas-adaptive control system can be developed and deployed for optimum engine performance. It is recommended to assess and evaluate the techno-economic feasibility of such fuel adaptive control systems together with OEMs.

Summary Table 1: H_2 limits, knowledge gaps and recommendations for the different engine parts

Engine parts heavy-duty engine	H_2 limit	Knowledge gaps	Recommendations
Catalyst/emissions	Unknown	Lack of (research) data on total system (engine + TWC) behaviour; possible unwanted emissions (NH_3 and N_2O)	Generating practical experience with hydrogen blending using relevant heavy-duty vehicle, in joint effort with relevant OEMs
Spark plug	Unknown	Effect of hydrogen on firing end materials; heat grade selection; no consensus on potential of spark plug gap and ignition energy reduction for lifetime extension	Consult with spark plug OEMs for joined research.
Lubricating oil	Unknown	Effect of hydrogen and water in crankcase atmosphere on lube oil performances and lifetime; if needed, availability of suitable product	Consult with lube oil OEMs for joined research.
Fuel line	10 vol% for former components 30 vol% for new components	Information in this study is provided by a single manufacturer. It is unknow what the opinion is of the other manufacturers (no response).	Generating practical experience with hydrogen blending using relevant heavy-duty vehicle
CNG tank system	Steel tank: 2 vol% (UN/ECE R110) Type 4 tank: tank itself can handle full range 0-100 vol% H2 (when using identical pressure as CNG). However, the tank periphery components may be the limiting factor.	Unknown if periphery components can handle hydrogen/CNG blends and by how much hydrogen failure occurs	Development of dedicated standards for HCNG for tank periphery components. If needed replace all steel tanks (type 1) from the market Generating practical experience with hydrogen blending using relevant heavy-duty vehicle
Engine ECU/Injector	15-20 vol%, for higher percentages recalibration of the engine mapping is needed	Unknown how the ECU and stoichiometric heavy-duty engine response to >20 vol% hydrogen in CNG	Generating practical experience with hydrogen blending using relevant heavy-duty vehicle
Engine knock	Depending upon the CNG quality and engine specification (MN)	Knock sensitivity for H2/CNG blends in CNG engines Unknown how accurate the knock resistance can be predicted of H_2/CNG blends using existing MN methods for stoichiometric heavy-duty engines	Studying effect hydrogen blending on knock behaviour in a CNG engine Exploring knock mitigating measures while maintaining efficiency when H_2 is added. Studying the accuracy of existing MN methods and if needed development of a dedicated MN algorithm for relevant CNG engines

2. Hydrogen specifications in Europe

The allowed hydrogen in natural gas pipeline specifications differ from country to country and range between 0 vol% to 10 vol% as can be seen in the Table below.

Summary Table 2 Overview hydrogen gas grid specification (see Chapter 5 for more details)

Country	Max. Allowed H_2 (vol.%)
Germany	Case by case (0-10%)
Netherlands	0.5%
Spain	5%
Sweden	0.5%
Belgium	0.1%
Denmark	not specified
UK	0.1%
France	6%
Austria	4%
Italy	0.5%
Switzerland	2%
Poland	not specified

The origins of the technical specifications defined in the different countries involve the effects of hydrogen on the transmission and distribution systems, end-use equipment, and underground storage. Based on a literature review, the summary of the hydrogen tolerances by volume in natural gas is presented in the summary table below. The Table shows that gas chromatographs and correlative gas quality sensors, gas turbines, gas compressors and CNG vehicles present or installed in the fields show the lowest tolerance to hydrogen addition to natural gas.

Summary Table 3 Estimation of hydrogen tolerance by volume percentage in the natural gas grid system (see Chapter 5 for more details).

		0-0.5%	0-2%	0-5%	0-10%	0-15%	0-20%	0-25%	0-30%	0-40%	100%
Transmission	Steel pipelines	green	green	green	green	light green	light green	light green	light green	light green	light green
	Compression Centrifugal	green	green	light green	light green	orange	red	red	red	red	orange
	Compressor piston	green	green	green	light green	orange	orange	orange	orange	orange	orange
Metering equipment & components	GC's and correlative gas quality sensors	green	orange	orange	orange	orange	orange	orange	orange	orange	orange
	Valves, turbine meters, filters etc.	green	green	green	green	orange	orange	orange	orange	orange	orange
Distribution	Steel pipes	green	green	green	green	green	green	green	green	orange	orange
	Fittings	green	green	green	green	orange	orange	orange	orange	orange	orange
	Plastic pipes	green	green	green	green	green	green	green	green	green	green
	Inhouse infrastructure	green	green	green	green	green	green	green	green	orange	orange
End use	Gas engines	green	green	green	light green	light green	orange	orange	red	red	orange
	Gas Turbines	green	green	light green	light green	light green	orange	orange	red	red	orange
	Industrial burners (indirect heating)	green	green	green	green	green	green	light green	light green	orange	orange
	Industrial burners (direct heating)	green	green	green	light green	light green	light green	light green	light green	orange	orange
	Feedstock	light yellow	light yellow	light yellow	light yellow	light yellow	light yellow	light yellow	light yellow	light yellow	light yellow
	Domestic (premix) boilers	green	green	green	green	light green	light green	light green	light green	light green	orange
	Domestic hot water heaters	green	green	green	light green	orange	orange	red	red	red	orange
	Domestic Cookers	green	green	green	light green	orange	orange	red	red	red	orange
Transport	CNG vehicles	green	green	orange	orange	orange	orange	red	red	red	orange
	CNG fueling stations	green	green	white	white	white	white	white	white	white	white
Gas storage	Salt caverns	green	green	green	green	green	green	green	green	green	green
	Other under ground storage	green	green	green	orange	orange	orange	orange	orange	orange	orange

Status	**Color code**
Currently already possible	green
Adjustments may be required (case by case)	light green
Retrofit , replacement and/or additional research required	orange
Not possible	red
Depends on industrial proces (no generic statement possible)	light yellow
Unknown	white

3. *Comparison between H_2-ICE and H_2-fuel cell for heavy-duty mobility applications*

Based on a literature review and interviews with manufactures a comparison is made between the H_2-ICE and H_2-fuel cell technology. The summary of the results, including the main advantages and disadvantages of both technologies, are presented in the table below.

Summary Table 4 Comparison between H_2 ICE and H_2 fuel cell (see Chapter 7 for details)

	H_2 ICE	H_2 fuel cell (PEMFC)
Technology	Combustion	Electrochemical conversion
Fuel requirements	Less fuel quality requirements as compared to FCEV (hydrogen quality)	High purity H_2 (99.9999%, grade 5 H_2)
Max. Engine efficiency	~40%	~60%
Fuel Economy [miles per gallon equivalent]	30-40	50
Maintenance [Euro/km]	0.19-0.20*	0.48-0.53**
Total costs of ownership after 5 years of operation [€/km]	1.4-1.5	1.6-1.8
Main advantages	Relatively low cost (known technology) Low re-engineering effort Less impact of external conditions (dust, cold conditions)	Higher efficiency No tailpipe emissions Fuel cell vehicles commercially available
Main disadvantages	Potential significant NOx emissions (Mitigating measures available such as EGR, injection strategy, fuel-lean limit) Development in research phase	High cost (e.g. precious metals) Higher maintenance cost.

* Assuming similar maintenance costs as for LNG/CNG truck
** Stack needs to be replaced after approximately 500.000 km

1. INTRODUCTION

Increasingly stringent regulations regarding CO_2 emissions, the growing need to enhance the sustainability and to increase the security of the energy supply within the European Union are the major drivers for renewables such as biogas, wind- and solar energy. Furthermore, to comply with the climate agreements the energy mix needs to be decarbonized substantially in the near future by increasing the fraction of renewable energy in the total energy mix and by making processes and end-use equipment more energy efficient. It is well known that the production of renewable energy from solar and wind fluctuate during the day and is strongly dependent upon seasonal variation. This fluctuation in production puts a lot of pressure on the capacity of power grids. To avoid enormous investments in grid reinforcement and storage capacity in the power grid, the excess of renewable power produced can be converted to hydrogen, following the Power-to-Gas concept. Injecting hydrogen together with other renewable gases such as biogas into the existing gas grid is an effective means to avoid large investments in the infrastructure while ensuring the wide-spread use of these fuels by industrial, commercial, residential and transportation/mobility end users. An alternative solution is to use the produced hydrogen as feedstock or blend the hydrogen to natural gas at dedicated locations such as at CNG fueling stations.

The introduction of hydrogen to natural gas has an impact on the physical and chemical combustion properties of the gas delivered. As a result, hydrogen addition to natural gas can have a positive and/or negative impact to the performance of end-use equipment. Without additional mitigating measures, the addition of hydrogen to natural gas may affect engine performance such as fuel efficiency, occurrence of engine knock, catalyst lifetime and performance, wear and tear, ignitability of the mixture, life span of spark plugs and pollutant emissions (such as CO_2, NO_x and CH_4). For other equipment installed in the natural gas market the changes in combustion properties upon hydrogen addition can result in, for example, increased risk for overheating of the burner deck and flashback in domestic boilers and stoves, as well as increased NOx emission, and lower GHG emissions in (industrial) burners. Besides the changes in combustion properties of the gas, the introduction of hydrogen can also result in hydrogen embrittlement in (some) natural gas pipelines, gas compressors and compressed natural gas (CNG) tanks. The potential issues described above puts a limit on the maximum amount of hydrogen that can be added to natural gas.

This study addresses the benefits, limitations, and knowledge gaps of blending hydrogen to natural gas for CNG applications. The focus will be on compressed natural gas (CNG) heavy-duty vehicles using both conventional and alternative combustion modes such as (ultra-) lean-burn and High-Pressure Direct Injection (HPDI). In addition, the technical developments for using pure hydrogen in combustion engines are described. The pros and cons of using hydrogen in internal combustion engines are compared with using fuel cell technology.

Furthermore, this study provides an overview of the maximum hydrogen percentages allowed in the natural gas grid specification within the different EU member states. Also, insights in the sensitivity towards hydrogen blending in natural gas for the different type of end use equipment connected to the natural gas grid and for the natural gas network system itself are reported. Finally, an inventory of hydrogen detection technologies is provided.

2. IMPACT OF HYDROGEN ADDITION ON (HEAVY-DUTY) ENGINE PERFORMANCE

2.1. GENERAL OVERVIEW OF DIFFERENT GAS ENGINE TECHNOLOGIES

In the market, different gas engine technologies are available and used in different sectors such as mobility (light-, medium- and heavy-duty engines), Combined Heat and Power (stationary engines) and Marine (both short-sea and deep-sea). The different gas engine types used are presented in Figure 1. The gas engines can be divided into two different combustion principles 1) Otto cycle and 2) Diesel cycle. In the Otto cycle the fuel/air mixture is 'premixed' and ignited by an external heat source such as a spark plug or a diesel pilot injection. In the Diesel cycle, the air is compressed and subsequently fuel (e.g. diesel) is injected followed by spontaneous ignition of the fuel/air mixture. HPDI truck engines using LNG have been introduced to the market. The working principle of HPDI engines is the direct injection of the gaseous fuel (natural gas) in the cylinder during the high-pressure phase of the engine cycle, following the Diesel cycle. The gaseous fuel is injected at pressures above 300 bar, through a cryogenic pump present in the LNG tank. The start of the ignition of the injected gaseous fuel is triggered by the injection of small amount (pilot) diesel fuel. Generally, higher efficiency is obtained for the Diesel cycle in comparison to the Otto cycle. However, HPDI engines do not exist for vehicles with a CNG tank since gas compression to >300bar is inefficient and expensive.

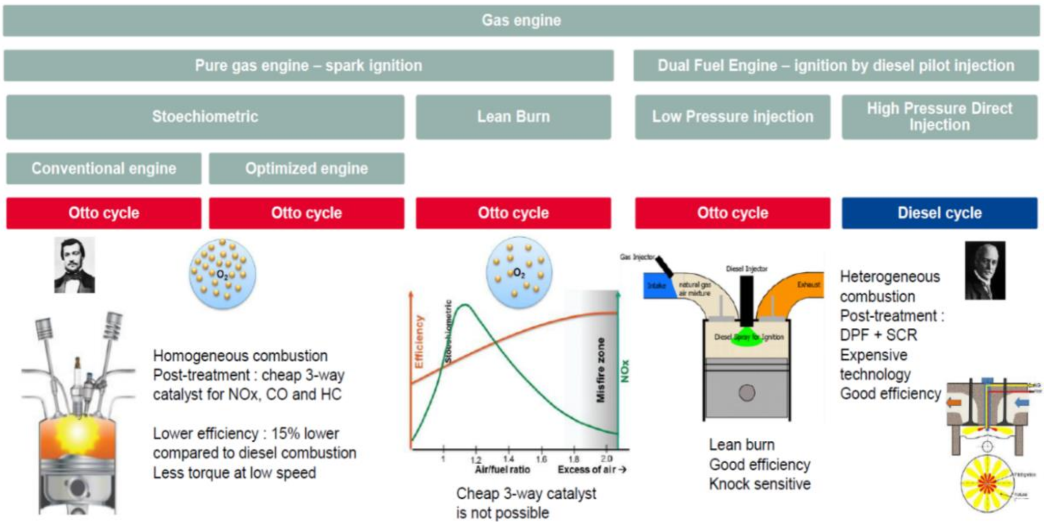

Figure 1 Overview of gas engine types, Source: Ph. China, TOTAL

The Otto cycle engines can be subdivided into stoichiometric and fuel lean (excess of air) combustion with a further characterization in terms of engine speed and specific engine load (BMEP). The lean-burn engines operate between an air-fuel ratio (λ) of $1.5 < \lambda < 1.7$ and the ultra-lean-burn engines operate at $\lambda > 2$. We note that the lambda (λ) range designated lean burn, reflects typical air-fuel-ratios used in the class of spark-ignited engines with open combustion chambers[1]. The ultra-

[1] In an open combustion chamber the fuel is injected directly into the cylinder, whereas in a prechamber is an separate auxiliary chamber connected to the main combustion chamber.

lean-burn range reflects air-fuel-ratios typically deployed in the spark-ignited, prechamber engine class and in the dual-fuel engine classes. For ultra-lean-burn spark ignited engines the prechamber is necessary to successfully ignite the very lean mixture. These ultra-lean-burn engines are typically used in the power generation sector and in the marine sector. In the mobility sector mainly stoichiometric and fuel lean spark ignited gas engines are used. The main focus of this study is on the impact of hydrogen addition to CNG on heavy-duty vehicles. Based on the literature review and response on the questionnaires we identified the major engine platforms present in these heavy-duty CNG vehicles. The results of this inventory are discussed below.

2.1.1. Inventory current engine types used in heavy-duty CNG vehicles in the EU market

An inventory of the gas engines being used and currently sold in the European market was performed to gain insight into the dominant heavy-duty engine technology in Europe. In Table 1 the heavy-duty CNG vehicles currently sold in the EU [1] is presented, see for more detail appendix A. It should be noted that vehicles with a CNG engine variant are listed.

The inventory reveals that all current HD truck (and bus) vehicles currently sold in the EU market are equipped with 4-stroke spark ignited stoichiometric engines. In the past, also vehicles with 4-stroke spark ignited lean-burn engines were sold in the EU, see Table 2 [2-7]. In contrast to the stoichiometric engine types (Table 1), the lean-burn engines do not meet the current Euro 6 emission standard and for this reason these engines will eventually be phased out when a new vehicle equipped with a stoichiometric engine is chosen for replacement. This is confirmed by our contacts with OEMs; stoichiometric engines, with or without EGR, appear to be the dominant engine technology for trucks.

Table 1 Current available heavy-duty vehicles in the EU with CNG as a fuel [1]

Engine make and type	Bus and truck series	Euro Standard Emission	Stoichiometry	Min. Methane number requirement
FPT Cursor 9	Iveco S-WAY NP CNG Truck Iveco Crealis Natural Power Bus Iveco Crossway LE Natural Power Bus Van Hool Exqui.city 24 Bus	VI	Stoichiometric without EGR	70
FPT Cursor 8	Iveco Urbanway Natural Power Bus Iveco Crealis Natural Power Bus			70
FPT Cursor 13	Long-distance and heavy-duty trucks			70
MAN E0836 LOH	Van Hool Exqui.city 18 Bus			?
Scania G280	Scania P/G CNG Version 280 Truck Scania Citywide LF CNG Bus Scania Citywide LE CNG Bus Scania Interlink LD CNG Bus		Stoichiometric with EGR	70
Mercedes M936G	Mercedes Econic NGT Truck Mercedes Citaro (G) NGT Bus Mercedes Citaro NGT hybrid Bus Mercedes Conecto (G) NGT Bus			?
Cummins Westport L9N	Isuzu Cityport CNG Bus Solaris Urbino 18 CNG Bus			?
Renault NGT9	Renault D Wide CNG Truck			?
Volvo G9K320	Volvo FE CNG Truck			?
MAN E2876 LUH	MAN Lion´s City CNG Bus			?

Table 2 Lean-burn engine types that are present in the EU market

Engine make and type	Min. Methane number requirement [2]	Euro standard emission [3-7]
Daimler M906LAG	75?	5
M447hLAG	?	4
Scania OC9 G05 305	?	5
Scania OC9 G04 270	?	5
Cummins Westport BG-230	65	5
Cummins Westport CGe280-30	65	3/4

2.2. EFFECT OF HYDROGEN ADDITION TO CNG ON THE PHYSICAL AND CHEMICAL PROPERTIES RELEVANT TO GAS ENGINES

Tables 3 and 4 show a selection of the physical and chemical combustion properties of methane/hydrogen blends. The tables show that the addition of hydrogen to CNG (CH_4) results in changes in these properties which can affect the combustion properties of internal combustion engines and the after-treatment system. For example from Table 3 it can be seen that hydrogen addition increases the adiabatic flame temperature, laminar burning velocity (S_L) and lowers the ratio of specific heats (γ) which results in changes in the in-cylinder conditions (P/T) and consequently will affect engine wear, efficiency, NO_x formation and the knock propensity. The observed decrease in minimum ignition energy with increasing hydrogen content in CNG can have an impact on the spark plug lifetime. Additionally, the broadening of the flammability limits potentially allows operation at higher excess air ratios.

Table 4 shows that the CO_2 emission decreases upon hydrogen addition and that the water content in the flue gas increases which can have an impact on the aftertreatment systems. Moreover, as can be seen in Table 4 the calorific value ($MJ/m^3(n)$) decreases with increasing hydrogen contact which will have an impact on the driving distance (mileage).

To what extent these changes in physical and chemical properties of CNG/H_2 mixtures affect engine performance is investigated in this study by performing a literature review, interviews with OEMs supplemented with illustrative calculations. The effect of hydrogen addition to CNG on the performance of CNG vehicles will be discussed in the following sub-sections.

Table 3 Combustion properties of methane/hydrogen mixtures at φ =1 (λ=1)

CH_4 (mol%)	H_2 (mol%)	S_L (cm/s)	$T_{adiabatic}$ (K)	Ratio of specific heats[1]	LFL		UFL		Quenching distance (mm)[8]	Min. Ignition energy (mJ) [9]
					Vol% in air	□	Vol% in air	□		
100	0	36.6	2228	1.3190	14.73	2.00	4.99	0.61	2.03	0.24
98	2	37.1	2228	1.3195	14.97	2.04	4.97	0.60	-	0.23
95	5	37.8	2230	1.3199	15.35	2.10	4.93	0.60	-	0.22
90	10	39.1	2232	1.3206	16.01	2.21	4.88	0.59	-	0.20
80	20	42.1	2238	1.3220	17.54	2.46	4.77	0.58	-	0.16
70	30	46.0	2244	1.3237	19.39	2.76	4.67	0.56	-	0.13
0	100	252.0	2384	1.3258	74.24	9.87	4.07	0.15	0.64	0.03

[1]calculated for a stochiometric methane/hydrogen air mixture after compression

Table 4 Data calculated at φ=1 (λ=1)

CH_4, vol.%	H_2, vol.%	CO_2, vol.% (exhaust)	H_2O, vol.% (exhaust)	H_i,[1] MJ/m^3	H_s,[1] MJ/m^3
100	0	9.48	18.78	35.9	39.8
98	2	9.43	18.86	35.4	39.3
95	5	9.35	19.00	34.6	38.5
90	10	9.20	19.22	33.4	37.1
80	20	8.88	19.75	30.8	34.4
70	30	8.49	20.38	28.3	31.7
0	100	0.00	34.10	10.8	12.8

[1]Lower heating value (Hi) and higher heating value (Hs) at P=1.015 bar and T=273.15K

The mechanical properties of metals and alloys used in pipelines, storage tanks and other components (e.g. injectors [10]) can be affected by hydrogen. This effect is known as hydrogen embrittlement or hydrogen assisted fracture which can cause the metal to loss of ductility and strength (becomes brittle) [8, 11]. The occurrence of hydrogen embrittlement depends on several factors such as the strength of the metal, temperature, pressure, hydrogen concentration, composition and nature of the metal (alloys) used, and so on [12]. For example, high strength material is known to be susceptible for hydrogen embrittlement [13]. For this reason, both regulation No 110 of the Economic Commission for Europe of the United Nations (UN/ECE, [14]) and EN 16723-2 for CNG vehicles [84] limit the hydrogen content in cylinders to 2 vol.% if the cylinders are manufactured from a high strength steel (ultimate tensile strength exceeding 950 MPA). According to Ref. [11], cylinders made of steel with a tensile strength below 950 MPA are compatible with hydrogen. According to Ref. [8], all metallic materials show a certain sensitivity towards hydrogen embrittlement, strongly depending on the stress level. Literature review reveals that copper and brass alloys, aluminium, aluminium alloys and copper beryllium can be used for hydrogen applications. Nickel, Nickel alloys and steels, titanium and titanium alloys are known to be sensitive to hydrogen embrittlement [8, 12]. The interaction between hydrogen and steel may also result in hydrogen embrittlement, depending on the chemical structure, strength, microstructure and impurities that are present as well as the heat (and welding) and mechanical treatment [8].

2.3. BASIC PRINCIPLE OF FOUR STROKE ENGINE

As described above, four-stroke engines are the main engine type are used in the European CNG vehicle market. In a four-stroke engine, the cylinder requires four strokes[2] of the cylinder to deliver one power stroke. An operating cycle (four strokes) requires two revolutions of the crankshaft

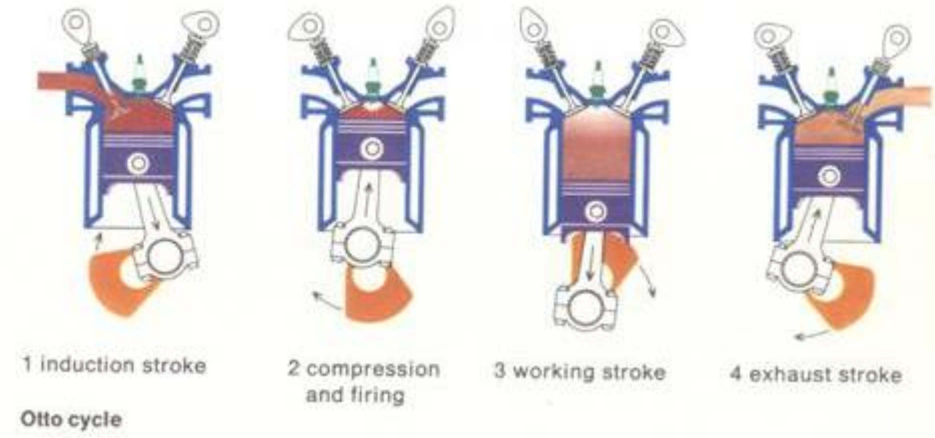

Figure 2 Four-stroke engine cycle [15]

As illustrated in the Figure above, the four strokes are;

1. **Intake stroke:** the piston moves to the bottom of the cylinder, creating volume that allows the fuel-air mixture to enter the cylinder.
2. **Compression stroke:** both intake and exhaust valves are closed. The piston moves up in the cylinder and compresses the gas mixtures.
3. **Ignition and expansion stroke**: at the end of the compression stroke the fuel-air mixtures is ignited (e.g. by a spark plug). The flame propagates through the cylinder, generating heat and additional thermal compression (increase in pressure). This high pressure and temperature gas push the piston down and force the crankshaft to rotate. For this reason, this stroke is also called the power stroke.
4. **Exhaust stroke**: As the piston moves downward, the exhaust valve opens and expels the exhaust gas.

The changes in physical and chemical properties presented in Table 1 caused changes in the in-cylinder conditions (P/T) during the compression and ignition/expansion stroke:

- mechanical compression of the unburned fuel/air mixture (compression stroke),
- ignition of the compressed fuel/air mixture and (ignition and expansion stroke),

[2] A stroke is the motion in either direction of the piston inside the cylinder.

- combustion behaviour of the fuel/air mixture (ignition and expansion stroke).

2.3.1. Effect of hydrogen addition to natural gas during compression stroke

The adiabatic compression of the unburned fuel/air mixture is dependent on the initial temperature pressure, compression ratio and the heat capacity ratio (γ) according to,

$$CR = \frac{V_2}{V_1} = \left(\frac{T_2}{T_1}\right)^{\frac{1}{\gamma-1}} = \left(\frac{P_2}{P_1}\right)^{\frac{1}{\gamma}} \quad (1)$$

At the same engine settings (CR, P_1, T_1) changes in the fuel composition results in a change in the heat capacity ratio. Consequently, the in-cylinder pressure and temperature during compression will increase upon hydrogen addition as illustrated in Figure 3. As we will discuss below this increase in pressure and temperature will have an important impact on the engine performance such as wear and tear, NO_x emission and the knock behaviour.

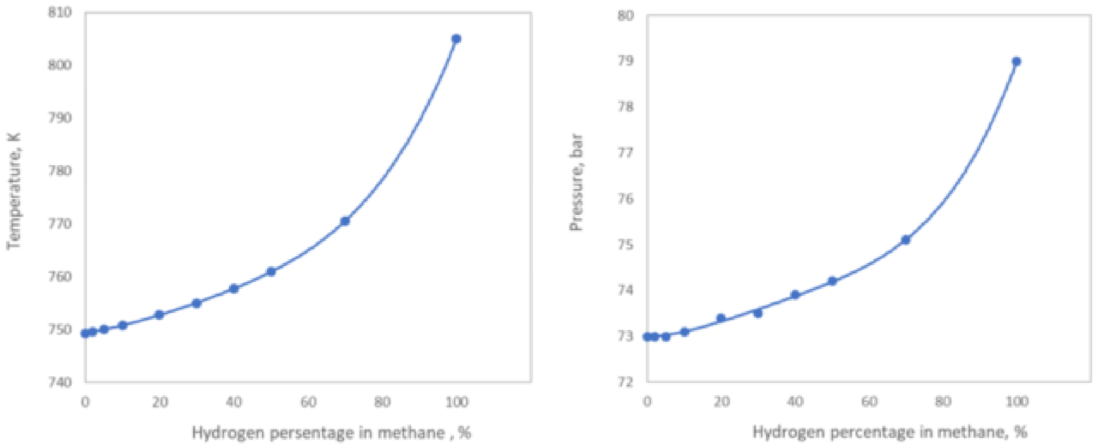

Figure 3 Temperature and pressure after compression for stoichiometric CH_4/H_2 mixtures at Pi=2.5bar, T_{intake}=308.15K and CR=12. The percentages in the figure are mole %.

2.3.2. Effect of hydrogen addition to natural gas during ignition and expansion stroke

At a certain point after the beginning of compression (and generally before top dead centre) the fuel/air mixture is ignited. To be able to ignite the mixture, the air-fuel ratio should be within the upper and lower ignition limit and enough energy should be available to ignite the fuel/air mixture (minimum ignition energy). Figure 4 shows that hydrogen addition expands the range of different fuel/air ratios where the fuel can be ignited. For example, pure hydrogen can be ignited at much higher air-fuel ratios (leaner mixture) as compared to pure methane. Moreover, the minimum ignition energy required to ignite hydrogen is much lower than needed for methane. The downside is that the mixture can be easier ignited by other sources such as hot spots which can lead to unwanted pre-ignition or knock.

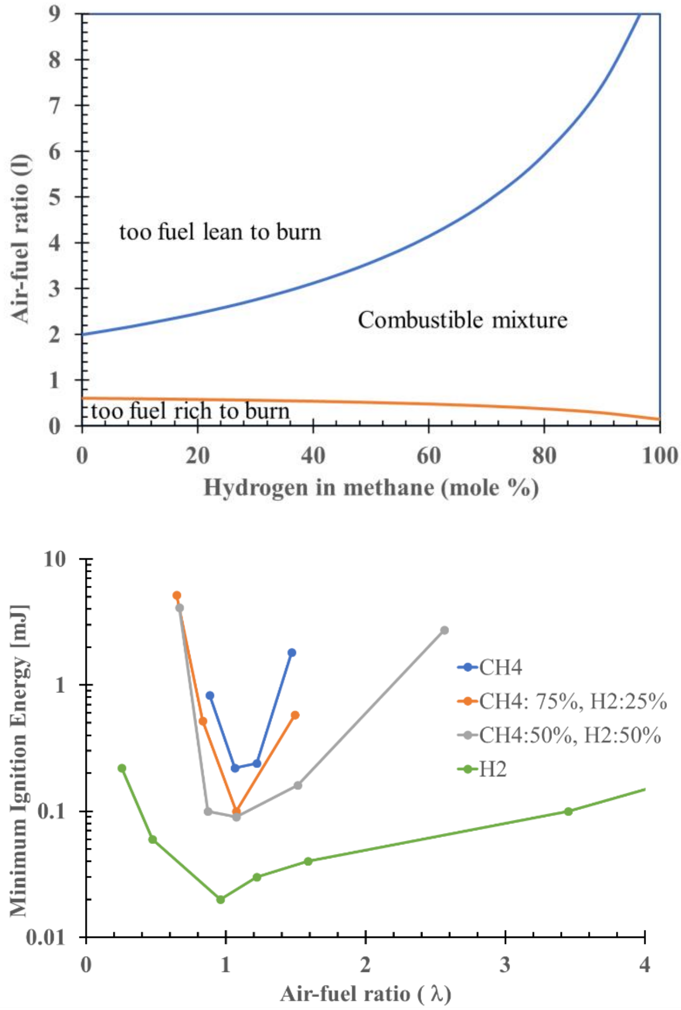

Figure 4 Lower and upper flammability range as function of hydrogen (above) and the minimum ignition energy for CH_4-H_2 mixtures at different air-fuel ratios [9, 16]. The percentages in the figure are mole %.

Once the mixture is ignited a flame propagates through the unburned fuel/air mixture. The burning velocity is the rate at which a flame propagates through the unburned gas/oxidizer mixture. This velocity is strongly dependent upon fuel and oxidizer type, air-fuel ratio and temperature of unburned fuel/oxidizer mixture. As can be seen in Figure 5, hydrogen addition significantly increases the burning velocity for both stoichiometric and fuel-lean conditions. The progressive combustion of the cylinder charge results in an extra (thermal) compression of the end gas in addition to the mechanical (piston) compression. Consequently, increasing the burning velocities due to hydrogen addition results in higher end pressure and temperature inside the cylinder as illustrated in Figure 5 (right). Furthermore, the temperature of the burned gases increases as a result of the increase in the adiabatic flame temperature upon hydrogen addition as shown in Figure 6

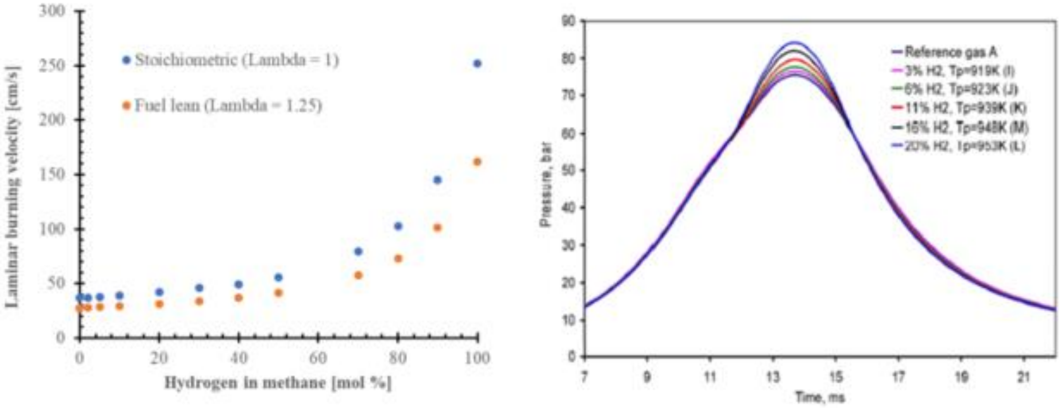

Figure 5 (Left) The effect of hydrogen addition to natural gas on the laminar burning velocity. The calculations were performed for stoichiometric (λ=1) and fuel lean (λ=1.25) conditions (1 bar, 298 K) using premix code of the CHEMKIN II package [17] and the USC II Mech chemical mechanism [18]. Right; Measured in-cylinder pressure cycle and maximum calculated temperature of the compressed unburned gas upon hydrogen addition [19].

As can be seen from the Figure above, the addition of hydrogen to natural gas increases the flame temperature (and consequently the exhaust gas temperature) for both stoichiometric as fuel-lean operating conditions. This increase in flame temperatures affects the NO_x emission as will be discussed below

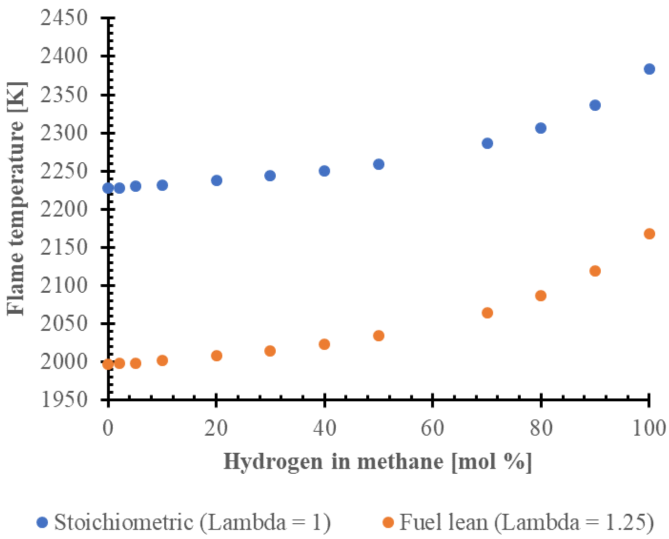

Figure 6 The effect of hydrogen addition to natural gas on the flame temperature. The calculations were performed for stoichiometric (λ=1) and fuel lean (λ=1.25) conditions (1 bar, 298 K) using TECOM [20].

To summarize the above, the addition of hydrogen to natural gas increases both flame temperature and burning velocity, while it lowers the heat capacity. The combined effect (assuming identical ignition timing) results in higher peak pressures and exhaust temperatures.

2.4. RESULTS OF THE IMPACT OF HYDROGEN ADDITION TO CNG: LITERATURE REVIEW

2.4.1. Effect on in-cylinder conditions (pressure, temperature)

Experiments [21] were performed on a six-cylinder turbocharged **lean-burn SI LNG heavy-duty engine (λ=1.18-1.27)**. As shown in Figure 7, the addition of hydrogen to CNG results in an increase in the measured peak cylinder pressure and pressure rise rate; 22 vol.% H_2 results in an approximately 20% increase in the peak pressure. Moreover, analyses show that the ignition delay time[3] and combustion duration reduces with increasing hydrogen fraction in CNG. Similar results were obtained in [22] were tests are performed in a 6-liter turbo charged engine at λ=1.3. The increase in temperature and pressure with rising hydrogen fractions in CNG is attributed to the enhanced burning velocity.

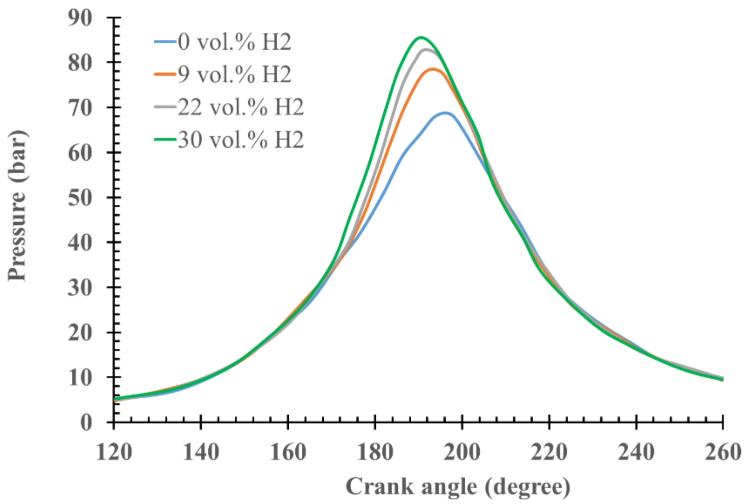

Figure 7 Measured cylinder pressures for CH_4-H_2 mixtures. The measurements were performed for a stoichiometric engine (λ=1.0). Data reprinted (adapted) from [21]

Similar combustion behaviour has been observed in lean-burn CNG engines by others (e.g. [19, 21, 23-27]. For example, measurements performed in a **lean-burn spark ignited gas engine** at fixed air factor (λ=1.55) and speed of 1500 rpm show that the in-cylinder peak pressure increases substantially while keeping constant power output, constant spark timing and constant air-fuel ratio [19]. The results show that the addition of 20 vol.% hydrogen causes a pressure rise of 9 bar (70 to 79bar; 12% increase) and a temperature rise of the unburned end-gas of about 35K. The observed increase in pressure and temperature are predominantly caused by the increase in the burning rate and the reduction in the heat capacity of the fuel-air mixture as also seen Table 3. As described in [24] the increase in peak pressure and temperature can result in increased engine wear and can affect the engine performance such as increase in the NO_x emission, engine knock and an increase in

[3] The time between spark discharge and 5-10% mass burned.

the thermal efficiency. These topics will be addressed in more detail further on in the report.

The effect of hydrogen on engine performance was studied in [24] using **a 4-stroke single cylinder research engine having port injection and an engine speed of 1400 rpm**. The experimental test conditions were chosen to study the impact on the changes in hydrogen fraction in CNG, changes in air-fuel ratio (λ=1.1 and 1.3) and the changes in spark timing on engine performance. The results show that at both λ=1.2 and λ=1.8 hydrogen addition (15, 20 and 25 vol.% H_2) results in faster combustion and as a result the peak pressure increases and shifts towards TDC with increasing hydrogen percentages. For example, at λ=1.2 the peak pressure increases by about 8% when 20 vol.% H_2 is present in CNG. However, at very lean-burn conditions (λ=1.8) the shift of the maximum peak pressure towards top dead centre (TDC) is only marginal in comparison to the results at λ=1.2 [24]. Similar results are found in [23, 28] where in [28] increasing the air-fuel ratio from λ=1 to 1.2 leads to a decrease in peak pressure values. As the H_2 percentage (up to 30 vol.% H_2) is increased, maximum peak pressure is found to be closer to the TDC.

Experimental and numerical analyses on a **multi cylinder stoichiometric heavy-duty engine**, fuelled with natural gas–hydrogen blends (0, 20 and 40 vol.% H_2) are reported in [29]. The measurements results show that when increasing the H_2 content a higher pressure gradient (pressure rise in bar per crank angle degree) is obtained due to a shorter combustion duration. Both the earliest phase of combustion (10% of the fuel is burned) and main combustion duration are reduced by the presence of hydrogen. The faster heat release upon hydrogen addition causes a faster increase in the pressure and an increase in the peak pressure [29]. The increase in peak pressure observed by the authors is about 5 bar (6% increase) and is considerably lower than found above. However, here we have to remark that during the tests with the two hydrogen blends the spark timing was more retarded which generally lower the peak pressures. [30] presents the combustion characteristics of a naturally aspirated spark ignition engine, intended for installation in vehicles, fuelled with different hydrogen and methane blends. The experimental tests were carried out in a wide range of speeds at equivalence ratios of 1, 0.8 and 0.7 (respectively corresponding to lambda 1, 1.25 and 1.43) and at full load. The ignition timing was maintained for each speed, independently of the air-fuel ratio and blend used as fuel. It was observed that hydrogen enrichment of the blend improve combustion for the ignition timing chosen. This improvement is more pronounced at low speeds, because at high speeds hydrogen effect is attenuated by the high turbulence. Also, hydrogen addition allowed the extension of the lean operation limit enabling the engine to run stable in points where methane could not be tested.

The literature review reveals that hydrogen addition to CNG result in an increase in the maximum pressure generally (and temperature) which can affect engine wear, efficiency, emissions and engine knock. Generally, 20 vol.% hydrogen in CNG increases the peak pressure between 6-20% depending on the engine settings.

2.4.2. The effect of hydrogen addition on the combustion stability (COV_{IMEP})

The major challenge of lean-burn CNG engines is to improve combustion stability. Upon increasing the air-fuel ratio (λ) the burning velocity reduces significantly up to the point that the mixture cannot ignite (outside the flammability limit, see Table 3) which results in instable combustion or in worst case misfire. As a result, the cycle-to-cycle variation increases when the mixture gets leaner. The coefficient of variations (COV_{IMEP}) is widely used to quantify the combustion stability and is expressed by a ratio between standard deviation (σ) to the mean value (μ) of indicated mean effective pressure and usually expressed in percent.

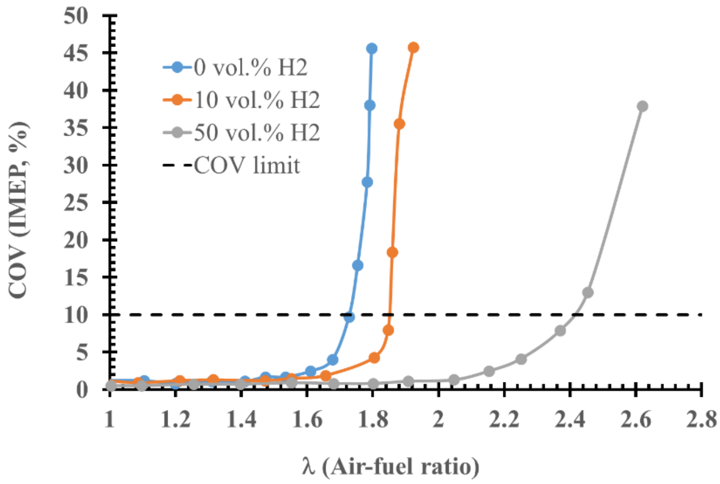

Figure 8 COV_{IMEP} versus air-fuel ratio for fuel blends of various hydrogen fraction. Data reprinted (adapted) from [31].

According to Ma [31] it is generally accepted that a COVimep above 10% is perceived by a driver as a poor running condition. As shown is Figure 8 the COV_{imep} increases with increasing air-fuel ratios [31] as also observed by others (e.g. [21, 31-33]). As a result of the improved combustion stability upon hydrogen addition engines can operate at fuel leaner conditions; for example as can be seen Figure 8 the lean limit is extended from λ=1.7 for CNG to λ= 2.4 for HCNG (50 vol.% H_2 in CNG). At idle conditions similar trends were observed [33]. As explained by Luo et al [31] the combustion stability improves with increasing hydrogen content due to the reducing the risk of misfire, enhancing of the initial flame kernel formation due to its low ignition energy and increases the burning velocity (see Table 3).

Based on the literature review we conclude that hydrogen addition improves the combustion stability reducing the risk of misfire and allows to extend the fuel lean limit.

2.4.3. Effect on efficiency

In this literature review it was investigated to what extent hydrogen addition affects the efficiency of conventional CNG engine platforms and of other combustion modes. From theory one can expect an increase in the Otto cycle efficiency, see equation 2 with increasing percentages hydrogen in CNG as a result of the increase in specific heat ratio (see Table 3). Furthermore, the increase in thermal compression due to the increase in the burning velocity (as shown above), along with a combustion closer to isochoric conditions are expected to increase the cycle efficiency.

$$\eta = 1 - \left(\frac{1}{CR}\right)^{\gamma-1} \qquad (2)$$

Experiments performed on a six-cylinder turbocharged lean-burn spark ignited LNG heavy-duty engine (λ=1.18-1.27) operated at a constant spark timing (28° CA BTDC) shows that with the continuous increase of hydrogen contents in the blends, the brake and indicated efficiencies[4] first rise and then reduce for hydrogen concentrations above 20 vol.% [21]. The authors attribute the increase in efficiency with hydrogen addition due to a faster heat release and a shorter burning duration. Above 20% hydrogen addition the heat release is too advanced and the higher temperature results in more heat loss and consequently a drop in efficiency [21]. This is probably the result of an engine calibration (in particular spark timing) which is not well fitted to higher hydrogen ratios, which would probably have required more retarded spark timing. The authors state that for a fixed compression ratio (13.6) hydrogen addition (up to 20 vol.%) improves the brake thermal efficiency by 1.8%. In the fuel lean engine studied in [28, 34, 35] the effective thermal efficiency of the fuel increases with increasing amount of hydrogen in the fuel. However, at high load no change in the efficiency was observed [34]. In contrast to [21], in [36] the authors observe that with increasing air-fuel ratio an increase in the BTE (break thermal efficiency) was noticed through an enhancement of the hydrogen fraction in the fuel blends of more than 20 vol.%. However, when the λ<1.4, hydrogen addition was not beneficial to improve the efficiency according the authors [36].

In [30] an increase in the indicated thermal efficiency (ITE) with the presence of 10 vol.% hydrogen in CNG is achieved at lean equivalence ratios and at stoichiometric conditions. For example, at stoichiometric conditions about 1.0% efficiency gain is achieved while at lean conditions (λ=1.25) about 4% efficiency gain is observed at 2500 rpm. At stoichiometric conditions the presence of 30 vol.% and 50 vol.% hydrogen in CNG results only in a moderate increase in the efficiency or even a reduced efficiency is observed depending upon the engine speed.

In [23, 36, 37] the authors observed that the BTE improves with hydrogen addition. In [23] the effect of hydrogen addition at different fuel-to-air ratios was studied. It was observed that an increase in the hydrogen percentage in the HCNG causes an increase in the efficiency of the used engine at all studied fuel-to-air ratios. For example, at stoichiometric conditions and a BMEP of 5.3 bar hydrogen addition leads to a BTE increase from 25.7% BTE for CNG to 27.2% (10 vol.% H_2), 28.1% (20 vol.% H_2) and 28.5% (30 vol.% H_2) BTE. The authors explain the observed BTE increase as a result of the higher flame speeds with HCNG, which makes the combustion duration shorter and results in the comparatively earlier start of combustion (SOC). A decrease of more than 25% in the fuel consumption rate (kW) was achieved in HCNG idle operations compared to CNG [33], as a result of improved stability (COV). In [25] two fuel lean (Euro 3) buses for urban transit service were fuelled with HCNG blends with different percentage of hydrogen (5 vol.%, 10 vol.%, 15 vol.% and 20 vol.% hydrogen). The results show that the fuel efficiency increases with increasing amounts of hydrogen in CNG. For example, when using 20 vol.% hydrogen a fuel efficiency gain of about 13% was observed.

Experiments on stoichiometric heavy-duty engine in [29] highlighted a faster combustion without an efficiency improvement when adding 20 vol.% and 40 vol.% hydrogen to natural gas. Analyses based on numerical show that more complete combustion takes place when adding hydrogen to natural gas of which is related to a lower “quenching distance” [29], thus the flame is typically extinguished at a

[4] The brake thermal efficiency is defined as the ratio of the amount of power available at the crankshaft (brake power) to the fuel energy. The indicated thermal efficiency is the ratio of the work produced (power exerted on the cylinder) to the fuel energy. The brake thermal efficiency is lower than the indicated thermal efficiency since it also includes friction losses (e.g. between cylinder and the walls).

lower distance from the wall, at H_2 increasing, giving higher heat losses. This higher heat loss explains the negligible effect of the observed faster combustion on engine global efficiency [29]. However, here we have to note that during the tests described in [29] the spark timing was more advanced (i.e. occurring earlier) when using hydrogen in CNG. Advancing the spark timing affects the thermal compression. We expect that when keeping the spark timing constant (or when retarding the spark timing to obtain a constant combustion phasing) with increasing hydrogen addition the efficiency will increase.

At DNV, the effect of hydrogen addition on the combustion performance using a high-speed medium BMEP lean-burn spark ignited gas engine was investigated. The tests were performed using two different air-fuel ratio (AFR) control modes, speed-density AFR control and lambda-sensor AFR control respectively, reflecting the two most commonly used air-fuel ratio control strategies used in CHP engines. When using the lambda sensor control system, the AFR and power output is kept constant when adding hydrogen to natural gas. As a result of the increase in combustion speed more fuel is consumed near TDC resulting in an increase in the maximum pressure (and temperature) as can be seen in Figure 7. As a result, the shaft efficiency increases by about 2.5 %-points when 20 mol% hydrogen is added to natural gas. When using the speed density control system, the power output and the intake manifold pressure is kept constant. Consequently, when adding hydrogen to natural gas the lambda is increased. The increase in lambda compensates the increase in burning velocity upon hydrogen addition (increasing lambda results lowers the burning velocity, see Figure 5). The increase in efficiency is about 1 %-point when adding 20 mol% hydrogen to natural gas.

Table 5 Lean-burn high speed medium BMEP engine using two different lambda control systems. Measurements are performed at constant speed and power (Fixed engine speed: 1500 rpm and power output: 220kW)

Natural gas (mol%)	Hydrogen (mol%)	Lambda sensor AFR control		Speed density AFR control	
		Lambda	Shaft eff. %	Lambda	Shaft eff. %
100.0	0.0	1.657	37.60	1.666	37.64
97.7	2.3	1.657	37.83	1.677	38.01
94.5	5.5	1.657	38.33	1.692	38.24
90.2	9.8	1.659	38.87	1.710	38.55
85.1	14.9	1.659	40.27	1.734	38.36
80.4	19.6	1.661	40.41	1.752	38.65

The literature shows that hydrogen addition potentially can significantly improve the efficiency. However, the increase in BTE strongly depends upon the variation of engine parameter settings that is controlled by the ECU such the air factor, spark timing etc.. In the literature described above, often multiparameter variations are applied in different engine types which makes it difficult to draw quantitative conclusions with regards to the effect of hydrogen addition on the thermal efficiency. However, based on the literature review generally it is observed an efficiency gain between roughly 0-4%-points and fuel efficiency gain of about 0-13% for 20 vol.% hydrogen addition. The efficiency gain is found to be beneficial for mainly lean-burn engines since hydrogen addition generally improves combustion stability (e.g. misfire) and increases combustion rate. However, the effect of hydrogen on the efficiency gain can be

neutral/unfavourable depending on the spark timing settings and heat wall losses.

Give the limited data found for heavy-duty stoichiometric vehicles it is strongly recommended to experimentally determine the fuel efficiency gain when adding hydrogen to CNG using a heavy-duty stoichiometric truck engine.

2.4.4. Effect on NO_x, CH_4, CO and unburned hydrocarbon emissions

2.4.4.1. Engine-out emissions (Pre-catalyst)

In this paragraph the impact of hydrogen addition to CNG on the pollutant formation is investigated based on a literature review. The pollutants studied in the literature are NO_x, C_xH_y, CO and CO_2 emission. Current regulations for heavy-duty vehicles in Europe must meet the Euro 6 regulations presented in Table 6. As discussed above, the stringent Euro 6 emission regulations resulted in gradual phasing out of lean-burn engines which currently have difficulties to comply with emission regulations. Stoichiometric engines (with- and without EGR) are equipped with a three-way-catalyst after treatment system that can control the NO_x, CO and hydrocarbon (HC) emission. Lean-burn engines are equipped with (only) a Selective Catalytic Reduction (SCR) after treatment system to lower the NO_x emission. In this Chapter, we focus both on the effect of hydrogen addition on the engine-out and tailpipe catalyst emissions (CO, HC, CH_4 and NO_x). The findings are summarized below

Table 6 Euro 6 regulations using the WHTC cycle

	g/kWh
CH_4	0.5
NMHC[1]	0.16
NO_x	0.46
CO	4.0
PM	0.01

[1]non methane hydrocarbons

The addition of hydrogen changes the H/C ratio of the fuel which decreases the engine-out unburned hydrocarbon and CO emission. Additionally, hydrogen addition increases the flame temperature, flame speed, improves the combustion stability and reduces the quenching distance[5] leading to a more complete combustion of the fuel [23, 24, 31, 38, 39, 81]. This decrease in emission upon hydrogen addition has been observed for both stoichiometric [40, 29, 32, 41, 81] and lean-burn engines [22-25, 28, 33, 34-35, 39, 42-43, 81]. The reduction in emission is seen to be dependent upon the amount of hydrogen added to the fuel. For example, Park et al. [27] observe approximately 41% reduction for the hydrocarbon when 30 vol% H_2 is present in CNG, whereas Lather et al. [32] observe a reduction up to 15% for hydrocarbon emission when 10 vol.% hydrogen is added to the fuel. In [23] the presence of 30 vol.% hydrogen reduces the CH_4 emission by about 50% at an air-fuel ratio of λ=1.3 as shown in Figure 10.

Experiments performed at different air-to-fuel ratios in different engines (described in e.g. [24-25, 28, 34, 44]) show that the addition of hydrogen to natural gas results in an increase of the upstream NO_x emission as a result of increased combustion temperature. As an illustration, Figure 9 shows that the NO_x increases linearly with

[5] The flame travels longer distance before being extinguished when the quenching distance is shorter.

increasing hydrogen percentage in natural gas at constant engine conditions (power, spark timing and lambda); the presence of 15% H_2 in natural gas increases the NO_x emission by around a factor two. The quantitative increase of the NO_x emission upon hydrogen addition depends strongly upon the engine settings and conditions. For experiments performed in a lean-burn heavy-duty engine show that the NO_x increases by around 35-40% when 30 vol.% hydrogen is present in the fuel [44].

This increase in NO_x can be controlled by applying NO_x mitigating strategies such Exhaust Gas Recirculation (EGR), changing the ignition timing and using after-treatment systems such as SCR (lean-burn engines) and, for stoichiometric engines, a three-way catalyst. Here we remark that in [29] no differences in NO_x emission was found with (20 and 40 vol.% H_2) and without hydrogen in CNG downstream the catalyst due to the high conversion efficiency of the used three-way catalyst (TWC). Similar results for a stoichiometric engine were also observed by [45]. Interviews with leading truck manufactures do not expect substantial increasing post-catalyst NO_x emission, however it was suggested that the presence of hydrogen potentially can result in higher NH_3 emission.

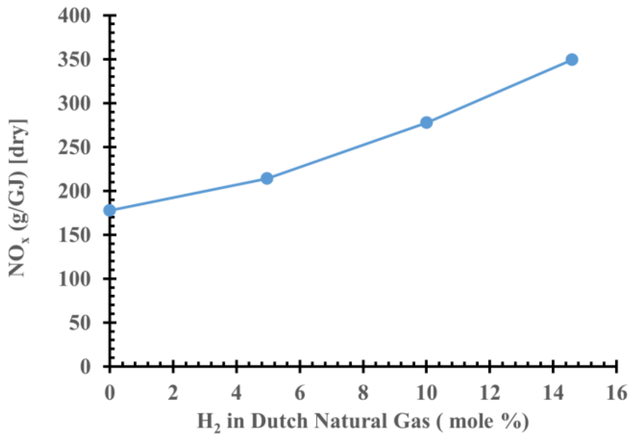

Figure 9 NO_x measurements[6] in hydrogen/natural gas mixtures performed an in lean-burn spark ignited engine at 1500 rpm at a fixed power, lambda, and spark timing [46].

The literature review reveals that hydrogen addition to CNG results in an increase in the engine out NO_x emissions and a decrease in the engine out CO, CH_4 and C_xH_y emissions. The quantitative changes in pollutant emission were found to depend upon the engine settings and conditions.

Given the limited data for modern stoichiometric heavy-duty engines found in the literature it is strongly recommended to perform a study on the pollutant emission using different H_2/CNG blends in this type of engines.

2.4.4.2. NO_x mitigating measures (engine-out)

The strategies to reduce NO_x when hydrogen is present in natural gas are summarized below and can also be used as input when, for example, new dedicated HCNG engines will be designed. Several NO_x mitigating strategies have been studied [21, 23, 24, 39]. In reference [24] it was shown that changing the spark timing from 30 to 20 °CA BTDC reduces the NO_x emission about 10% while maintaining a relatively

[6] NOx in dry exhaust; increasing H_2 content reduces the amount of dry exhaust as exhaust gases get wetter.

higher thermal efficiency (because retarding spark timing to compensate for the faster combustion helps keeping an optimal combustion phasing) under fixed engine load and equivalence ratio. Similar trends in NO_x emission were observed in [21, 39]. Another effective strategy to reduce the NO_x emission is to increase the air-fuel ratio as seen in Figure 10 and also studied and observed by others (e.g. [25, 39, 44, 81]). For fuel lean operating engines further increasing the amount of excess air is an effective strategy since it decreases the peak temperature and consequently the NO_x emission as shown in Figure 10. However, when using CNG the increased excess air results in an increase in the COV (more instable combustion) which results with an increase in CH_4, CO and HC emissions [44] as can be seen in Figure 10 (Right) and ultimately can result in misfire.

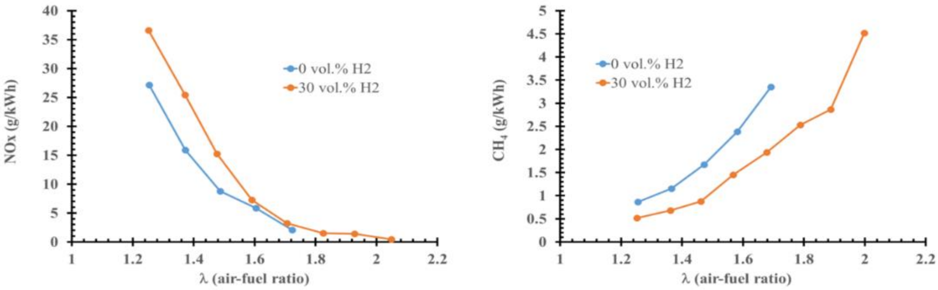

Figure 10 NO_x emission (Left) and CH_4 emission (Right) in relation to air-fuel ratio for each fuel. Data reprinted (adapted) from [44].

Addition of hydrogen enables to run the engine under fuel-leaner conditions resulting in a lower NO_x emission as shown in Figure 10. In general, the addition of hydrogen at fuel-leaner conditions results in a reduction of CO, CH_4 and hydrocarbon emissions as compared to the situation when no hydrogen is present in the fuel as also can be seen in Figure 13 [44]. For example, Deng et al [39] studied the effect of CO and hydrocarbon emission upon hydrogen addition (up to 50 vol.%) when operating under extended fuel operating limit as shown in Figure 11. The results for CO and hydrocarbon emission indicate that when the air-fuel ratio exceeds 1.7 more hydrogen addition resulted in less CO and hydrocarbon emission. This was attributed to the ability of hydrogen to improve the combustion (smaller COV, see above). Similar results were also obtained for CH_4 emission, see e.g. [39, 44], as also shown in Figure 10.

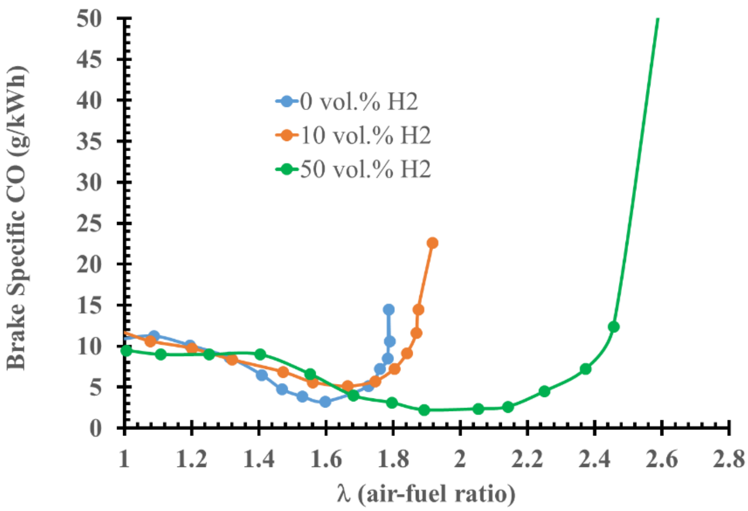

Figure 11 Measured brake specific CO emission at different air-fuel ratios (λ) at 0, 10, 30 and 50vol % H_2 present in CNG. Data reprinted (adapted) from [39]

Another common strategy to reduce the NO_x is applying Exhaust Gas Recirculation (EGR). When applying EGR the fresh fuel/air mixture is diluted with some amount of inert exhaust gases. These inert gases absorb the heat generated during combustion and lower the flame temperature resulting in a decrease in the thermal NO_x formation and thus in less NO_x emission levels. The disadvantage of using EGR is that the combustion instability increases when the EGR ratio increases which results in an incomplete combustion causing an increase in the HC emission (see for example Figure 12). Generally, EGR is applied for stoichiometric engine since lean-burn engines already operate often near their limit of stable combustion.

In [41] the performance of a stoichiometric operating engines with exhaust gas recirculation (EGR) was investigated experimentally using hydrogen-natural-gas blends. Optimal efficiencies were achieved with an EGR rate of 5-10 per cent; the NO_x emissions can be decreased by about 80% using 25% EGR dilution, but the combustion variation increases simultaneously. With hydrogen enrichment, the combustion variation (COV) can be controlled under 5 %, while the NO_x, hydrocarbon, and carbon monoxide emissions are kept at a low level (see also Figure below). Similar results have been observed in [81].

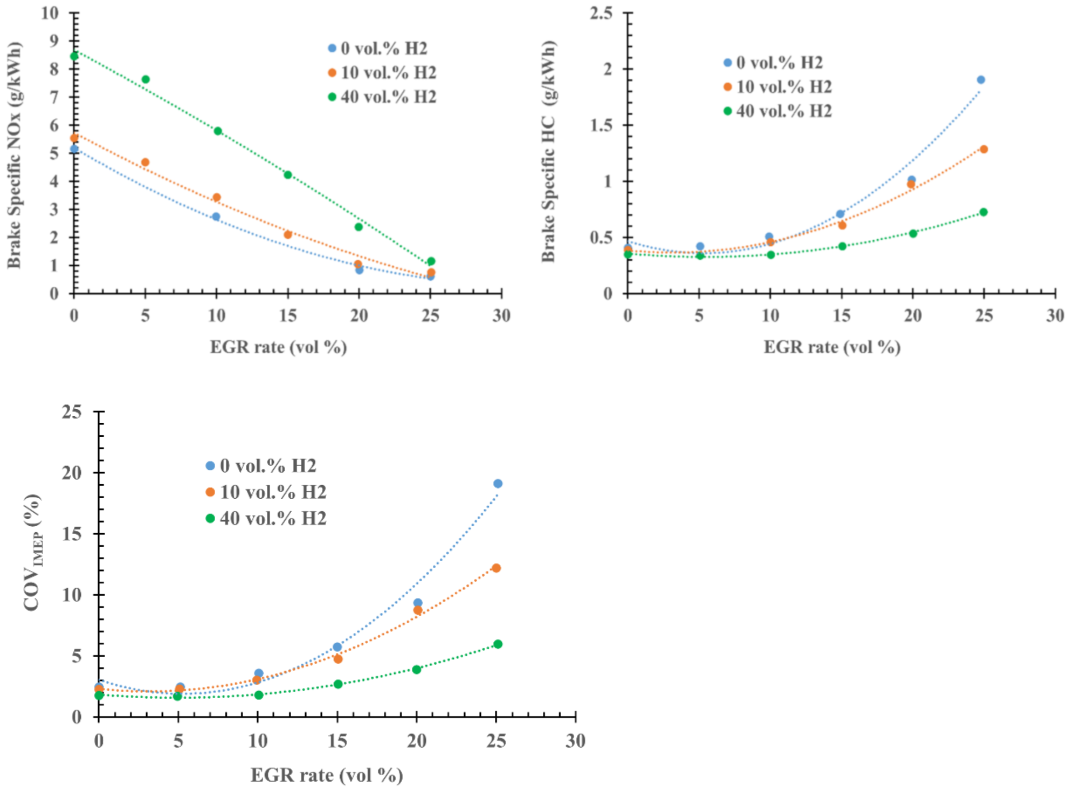

Figure 12 Measured pre-catalyst COV and NO- and HC emission at different ERG ratios and hydrogen fractions in CNG. Data reprinted (adapted) from [41].

Experiments on a Volvo TD100 Spark Ignited (SI) engine fuelled by NG and 25 vol.% HCNG (i.e. 25 vol.% H_2 in CNG) [45] were performed at lean-burn and stoichiometric conditions using EGR and TWC. The effective lower NO_x emission was found for the EGR condition; it shows near linear decreasing trend with increasing EGR percentage for 25 vol.% hydrogen enrichment with increasing BMEP, which is shown in Figure. 13. It has been concluded in [45] that the addition of hydrogen to natural gas reduce the difference between the emission levels for lean-burn operation and stoichiometric operation with EGR and a three-way catalyst. However, it was not possible to get close to the very low NO_x and HC emissions in the lean-burn operating engine as found at stoichiometric condition using a TWC.

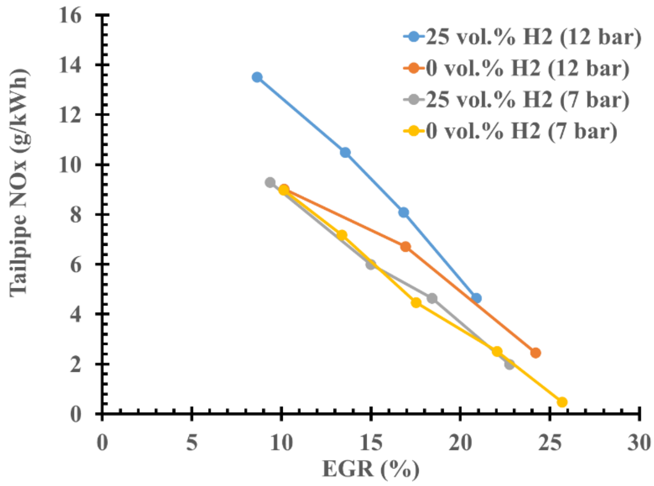

Figure 13 NO_x emission before catalyst under lean-burn conditions, EGR. Pressure values refer to BMEP levels. Data reprinted (adapted) from [45]

Several other studied such as in [47–49] found that the combustion stability was more efficient and greatly extended when hydrogen is present in CNG which allows EGR utilization to achieve substantial reduction of the NO_x emission. These findings suggest that when CNG always contain a fixed amount of hydrogen engines can be optimized for a specific H_2/CNG fuel blend. For example, in [48] a the 4-cylinder, 1.0 L, CR of 13.5, naturally aspirated Volkswagen Polo gasoline SI engine was optimized for HCNG mixture and high EGR rates in the part load condition. The experimental study showed that 15 vol.% HCNG (i.e. 15 vol.% H_2 in CNG) fuel blend can improve 3% thermal efficiency of the engine with optimum EGR and ignition timing. The engine's NOx emission reduces by 45%, THC improved by 7% and no change has encountered in the CO emission.

As discussed above, the presence of a fixed amount of hydrogen in CNG together with optimization of the engine settings offers opportunities to meet the increasingly stringent emission regulation for both stoichiometric and lean-burn engines.

2.4.4.3. Effect on aftertreatment systems

As discussed above, it is generally assumed that hydrogen mixture in natural gas will have an impact on the engine-out emissions, typically towards higher NO_x and lower CO, CO_2 and hydrocarbon emissions as compared to pure natural gas. As also addressed above, there are multiple engine control strategies, for both stoichiometric and lean-burn engines, to counteract or enhance a given impact on given engine-out emissions in response to hydrogen mixture. While possibly differing in their impact on emissions both qualitatively and quantitatively, all relevant measures have in common that they will also impact other key engine performances such as power output, fuel efficiency, combustion stability, component mechanical and/or thermal load and so on. In effect in practise, the margins for engine OEMs for recalibration of a given engine designed for operation on pure natural gas to accommodate hydrogen in the fuel mix for the purpose of optimizing engine-out emissions are limited, as other critical engine performance requirements will need to be assessed too. This means that structural changes for the worse in the engine-out emissions when introducing hydrogen in the fuel matrix of Natural Gas Vehicles (NGVs), must be balanced by the aftertreatment system.

With stoichiometric combustion rather than lean-burn combustion being the norm in modern (HD) NGVs, the question is how Three-Way-Catalysts (TWC), as being the dominant aftertreatment system for stoichiometric engines, behave with the changes in the engine-out emissions, and with possible changes in the operating conditions, associated with hydrogen mixture in natural gas. Unfortunately, most of the relevant (public) research seems to focus on engine-out emissions rather than taking a total system approach on tailpipe emissions also involving typical aftertreatment systems.

A leading supplier of (automotive) aftertreatment systems answer suggests that the introduction of hydrogen in the fuel mix would reduce the engine-out CO and CO_2 emissions, while increasing the NO_x and H_2O emissions, and likely also the exhaust gas flow rate. Unfortunately, no feedback was given regarding the anticipated effect on the conversion efficiency of TWCs in response to these changes in operating conditions. It was however claimed that catalyst formulations currently used in NGVs, possibly with minor modifications, would be fit for purpose. Regarding the lifetime of the TWC, it was suggested that the possible higher exhaust gas temperatures associated with hydrogen combustion, could have a negative impact.

Another major supplier of (automotive) aftertreatment systems shared some findings from a recent performed technology review and simulation of HCNG engines. In the case of NGVs without hardware modifications, they expect low impact on critical engine performances and TWC operating conditions for up to 20% hydrogen in natural gas, assuming sensible engine recalibration. Again, their feedback did not include information on the expected TWC performances, other than suggesting lower CO and hydrocarbon emissions. More impact was expected in the case of hardware modifications towards increasing the EGR rate taking advantage of the positive effect on combustion stability associated with hydrogen admixture. Unfortunately, no feedback on the expected tailpipe emissions here either.

In its bi-fuel Hydrogen 7 car, BMW [50] adopted a two-stage strategy for emission control in hydrogen-mode: ultra-lean-burn combustion at low loads, and slightly fuel-rich operation at high load. At low loads, the lean-burn combustion was sufficient to keep the engine-out NO_x emissions very low, not requiring aftertreatment. For adequate NO_x abatement at high load however, a TWC was required to comply with emission regulations. For this purpose, the engine was run slightly fuel-rich making use of the associated H_2 slip to efficiently reduce NO_x in the TWC. Interestingly, BMW said the TWC coating was slightly modified as compared to the standard configuration for gasoline operation, delivering over 99.9% NO_x reduction.

A recent study [51] confirms the high NO_x conversion efficiency to be obtained in a TWC on account of hydrogen slip in a hydrogen engine running slightly fuel rich. At least, this suggests that hydrogen slip can be an effective replacement for the typically decreasing fractions of CO and unburned hydrocarbons in the engine-out exhaust gas when admixing hydrogen to natural gas for the purpose of reducing NO and NO_2 in a TWC. While positive, it is unsure whether there will be enough hydrogen in the exhaust gas, combined with lower CO- and unburned hydrocarbon fractions, to reduce the expected higher engine-out NO_x emissions under all operating conditions.

Related to the anticipated hydrogen-slip involved with hydrogen mixture in natural gas, heavy-duty truck OEMs shared a concern for possible increased tailpipe emissions of NH_3 (and N_2O) since hydrogen is part of the reaction path in the formation of NH_3 (and N_2O) in TWCs under given operating conditions [52].

A topic apparently not yet addressed is the possible effect of the higher water (vapor) fraction in the exhaust gas associated with hydrogen combustion on the oxidation reactions in a TWC designed for natural gas combustion. From traditional oxidation catalysts for hydrocarbon abatement it is known that water inhibits the hydrocarbon conversion rate and deactivates the catalyst over time. As shown in Table 4, the water vapor in the exhaust increases with almost a factor two when changing from CNG to H_2. However, when 20 vol% H_2 is present in CNG the water vapor in the exhaust increases from 18.8 vol% to 19.8 vol%. %. To what extend this increase in water vapor and possible increase in exhaust gas temperature for stoichiometric heavy-duty vehicles affects the catalyst performances is yet unknown. Research is needed to assess if this increase will affect the catalyst performance.

In conclusion, exhaust gas aftertreatment systems, and in particular TWCs as being the dominant technology, are expected to be impacted by the introduction of hydrogen in the fuel mix for NGVs in terms of changes in their operating conditions and in the upstream exhaust gas composition matrix. Two major suppliers of (automotive) aftertreatment systems, with one of them suggesting that TWCs currently in use in NGVs would be fit for purpose for hydrogen mixture in natural gas, did/could not provide hard evidence to support such claims. Apparently, research related to a total system approach on tailpipe emissions also involving typical aftertreatment systems is few or is just being started. This, and a concern regarding possible unwanted emissions (NH_3 and N_2O) requires to have careful consideration of the performance of CNG vehicle aftertreatment systems when mixing hydrogen into natural gas. It is advised to consult with aftertreatment system OEMs and engine/truck OEMs for a coordinated possible research program investigating this topic.

2.4.5. Effect on engine knock

The knock resistance of gaseous fuels (CNG, LNG, H_2, and so on) is characterized by a methane number, which is like the octane number used to qualify gasoline. The engine knock is caused by autoignition of unburned fuel mixture in the cylinder before the mixture is completely consumed by the propagating flame. Mild engine knock increases fuel consumption and pollutant emissions, while severe knock can physically damage the engine, and should thus be avoided.

Pure hydrogen is very prone to engine knock in comparison to other fuels. The Figure below shows the experimental knock limited equivalence ratio as function of the compression ratio for hydrogen, Gasoline (octane number of 92) and methane [53]. The experiments were performed on a single cylinder, four stroke, spark ignition, CFR engine while operating unthrottled under atmospheric pressure conditions. The engine is of variable compression ratio (from 4:1 to 16:1) and spark timing. The knock limited equivalence ratio was determined by gradually varying the fuel-air mixture composition from lean towards stoichiometric until the onset of knock was first encountered, while keeping all other operating parameters constant [53].

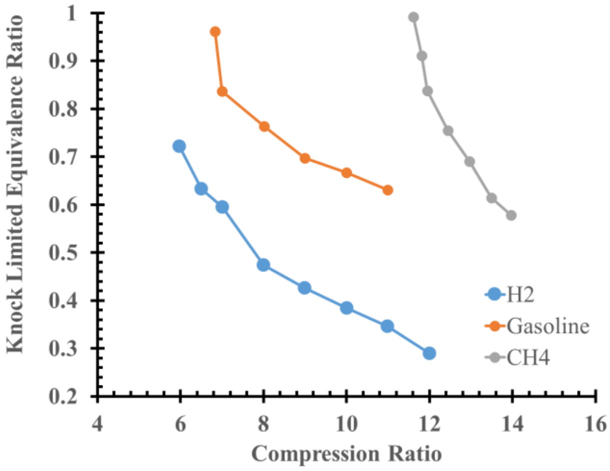

Figure 14 Knock resistance (KLER) for different fuels and compression ratios. Data reprinted (adapted) from [53].

Hydrogen only permits knock-free operation at very fuel lean conditions (low equivalence ratios). On the other hand, methane is less sensitive to engine knock, see Figure 14, since it allows knock-free operation at a wider range of air-fuel ratios (e.g. up to stoichiometric for a compression ratio of 12). As will be discussed below, for this reason the methane number is often scaled from 0 (hydrogen, most sensitive to engine knock) to 100 (methane, less sensitive to engine knock).

Engine knock occurs depends upon the gas composition and in-cylinder conditions such as pressure and temperature. In Figures 15 and 16 the operating conditions (pressure and temperature) are shown which were measured for three different engine platforms when running on LNG. As can be seen from these Figures, the operating conditions for the dual-fuel marine engine are significantly different than those for the stoichiometric truck engine and fuel-lean CHP engine [2]. As a result, each engine platform shows a different sensitivity towards engine knock.

Figure 16 shows the temperature dependence of the autoignition delay time, which is an indicator of the occurrence of engine knock: shorter values under the same conditions increase the change of the occurrence of engine knock. Furthermore, the results show that the variation in the combined effect of composition (mainly ethane and hydrogen) with temperature implies that different temperature regimes in different engines can yield quantitatively, and even qualitatively, different knocking behaviour in the unburned fuel mixture, the so-called end gas.

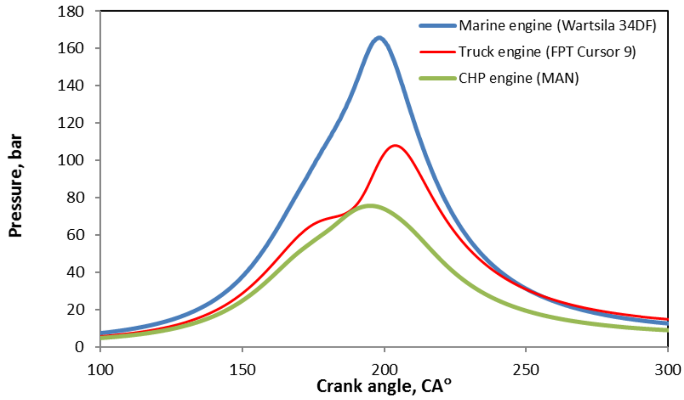

Figure 15 Measured pressure profiles for LNG in the marine engine (W34DF), the truck engine (Cursor 9) and the CHP engine (MAN). Data reprinted (adapted) from [2].

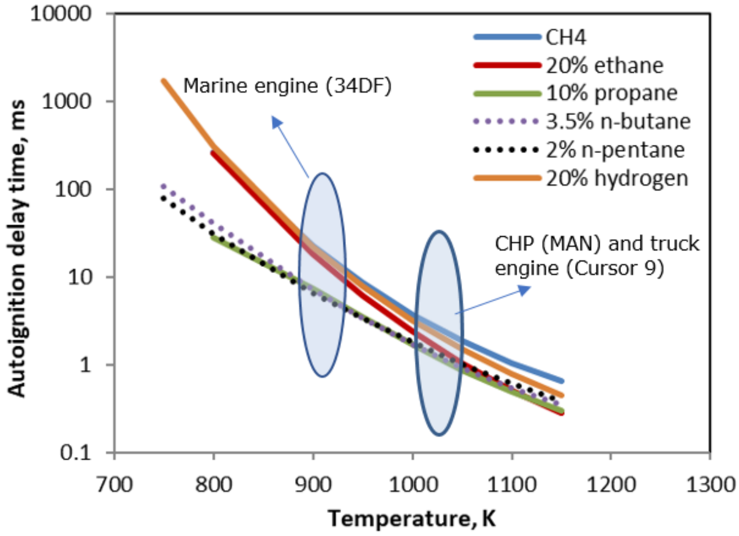

Figure 16 Calculated autoignition delay times as function of temperature. The percentages in the figure are mol%. Data reprinted (adapted) from [2]. The blue circles represent typical maximum operating temperatures of the compressed unburned end-gas for a marine engine (34DF) and a CHP/ Heavy-Duty truck engine.

2.4.5.1. Example of experimental knock data

Only limited engine knock measurement data are available in the literature for natural gas /hydrogen mixtures. In [56] the variations in the knock limited spark timing (KLST) was measured with increasing fractions hydrogen in methane. The results presented in Figure 17 (Left) show that the KLST decreases almost linearly with the hydrogen percentage in methane up to about 60 vol.% hydrogen in methane. The linear decrease in the knock resistance reflects the nearly linear reduction in the combustion duration, while affecting the reactivity of the end gas mixture itself only little as also observed in [19, 54]. In [53] knock measurements were performed by changing the compression ratio at fixed ignition timing (KLCR). These measurements also show that the knock resistance decreases with increasing fractions hydrogen in the fuel.

In Figure 17 (Right) the knock resistance measurements in a lean-burn medium BMEP high speed engine for various hydrogen/Dutch natural gas mixtures is presented [55]. Like the results found in [56], the measured knock resistance (KLST) was found to linearly decrease with increasing hydrogen percentages in Dutch natural gas (DNG). From the results found in the literature we conclude that the addition of hydrogen lowers the knock resistance however the quantitative effect of hydrogen strongly depends upon the in-cylinder conditions (λ, P and T).

For stoichiometric heavy-duty vehicles, no relevant information of the influence of hydrogen on the knock resistance was found in literature. Interviews with OEMs revealed that in field demo's the maximum hydrogen concentration added while ensuring no knock issues is about 20 vol.% hydrogen in CNG. However, the OEMs remark that these experiments were performed with so called 'lean' gases having high methane numbers. In case hydrogen is added to CNG that has a low methane number (close to knock limit of the engine) the amount that can be added is strongly limited.

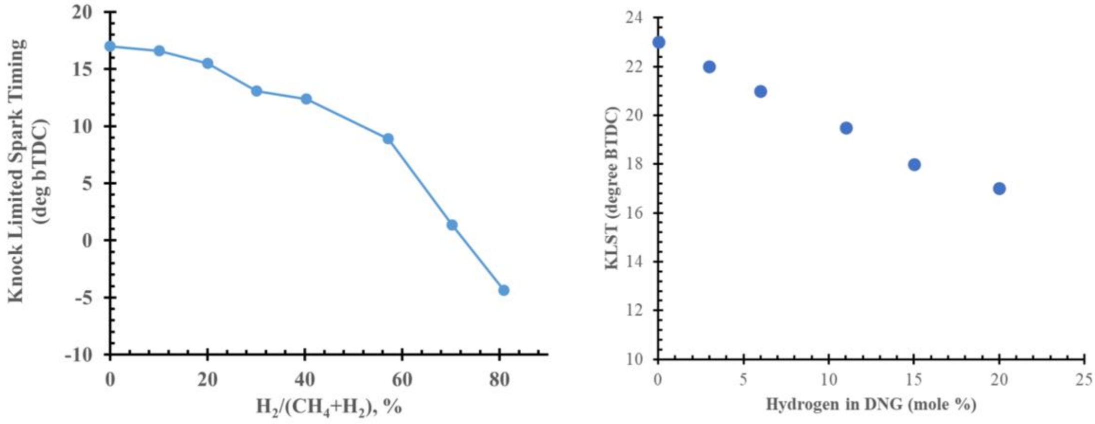

Figure 17 (Left) Measured knock resistance expressed in knock limited spark timing (KLST) for different volume percentages hydrogen in natural gas. Right: Measured KLST data for different H_2/Dutch natural gas blends measured in a spark ignited fuel lean-burn (λ=1.55) high speed gas engine (1500rpm). Data reprinted (adapted) from [56] and [55].

2.4.5.2. Overview of methane number calculation methods

Several methods have been developed to classify gaseous fuels for their knock sensitivity such as AVL [57], MWM [58–56], CARB [61-62], GRI [61-62], Cummins [63], Waukesha Knock Index method (WKI) [64], Wärtsilä (WMN) [65] and PKI MN [58, 66]. Each method calculates the methane number using the fuel composition as input. As can be seen in Table 7, each method was validated and optimized for a specific engine platform. This means that the methane number methods can give different outcomes for the same fuel composition, which results in confusion for end-users and fuel suppliers in the gas value chain. The detailed description of the different methods available is given in Appendix B. In Table 7, an overview of the methane number methods available in the market is given.

In Figure 18 the effect of hydrogen addition to CNG[7] on the methane number calculated using the different methane number methods is presented below. Here we note that the CARB/GRI method is not included since the calculated methane numbers increases with increasing hydrogen percentages in the fuel which is contrary to the experimental observations presented in Figures 14 and 17.

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[7] The CNG composition was 94.9 mole% CH_4, 3 mole% C_2H_6, 1 mole% C_3H_8, 0.5 mole% n-C_4H_{10}, 0.5 mole% N_2 and 0.1 mole% CO_2

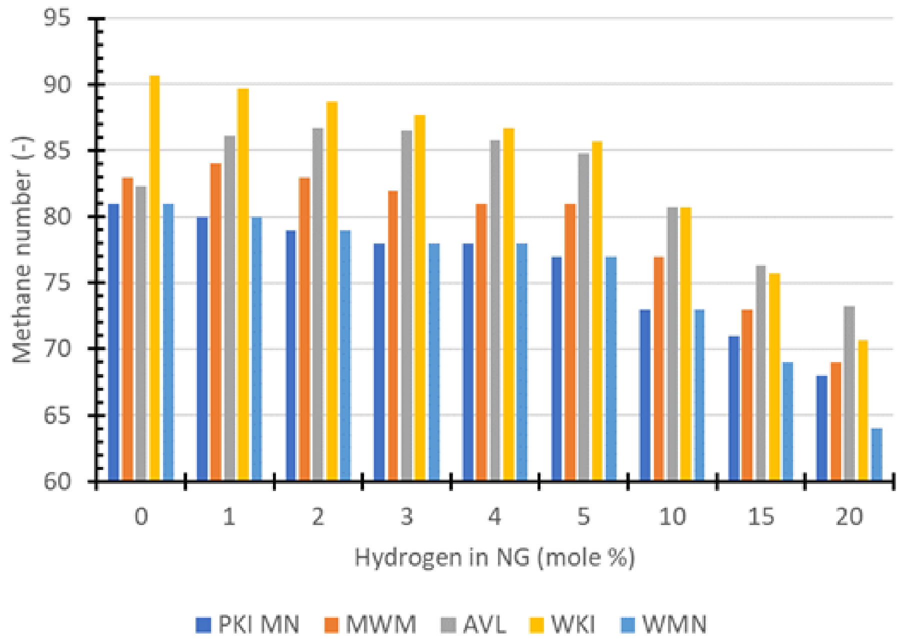

Figure 18 Calculated methane numbers using different methods (AVL, MWM, PKI MN, WKI and WMW) for CNG/H_2 mixtures.

The results presented in Figure 18 show that the calculations using the PKI MN, WKI, MWM and the AVL MN method give similar results for a typical CNG composition (0 mole% H_2) with the maximum observed deviation being about 2 Methane Number points for these gases. However, the WKI method shows a large deviation of about 10 Methane Number points for pure CNG. The knock resistance calculated for the various CNG/H_2 blends presented in Figure 18 show substantial qualitative and quantitative differences when using the different Methane Number methods. The WKI, PKI MN and the WMN methods show a decrease in the Methane Number with increasing fractions hydrogen in CNG and thus a decrease in the knock resistance as also observed in the measurements (see e.g. Figure 17). In contrast, the knock resistance calculated using the AVL methane number show a substantial increase from 82 to 87 Methane Number points when adding small percentages of hydrogen to CNG (0-3 mole%). For hydrogen percentages above 3 mole% in CNG the AVL MN decreases with increasing hydrogen content. The MWM method shows up to 5 mole% hydrogen in CNG only a marginal reduction in the Methane Number and up to 1 mole% H_2 in CNG even an increase in the MWM methane number. For percentages above 5 mole% a substantial decrease in the Methane Number is observed.

Table 7 Overview of methane number methods

Method	H_2 range (mole %)	Experimental basis	Pros	Cons
AVL	0-100%	**Stoichiometric** CFR engine (1960s)	• Available for the full hydrogen range (0-100%) • Most **popular** method in engine manufacturing industry	• Modern engine technology differs from test engine used • No discrimination between isomers of higher hydrocarbons • Small H_2 content shows **increase** in methane number (up to 3%) which is in contrast with experimental results • Triangles, complex fitting routine; not easy to implement • Propriety software
MWM	0-100%	Method is based on AVL data and adapted for a "modern gas **lean-burn** engine"	• Available for the **full** hydrogen range (0-100%) • Publicly available • Algorithm available in ISO 23306 standard (LNG as marine fuel)	• No discrimination between isomers of higher hydrocarbons • Triangles, complex fitting routine; **not easy** to implement • No discrimination between isomers of higher hydrocarbons
CARB/GRI	H/C ratio > 2.5	CFR test engine. Correlations between the motor octane number (MON) and methane number	• Publicly available via Website	• No discrimination between isomers of higher hydrocarbons • Hydrogen addition shows **increase** of methane number which is in contrast with experimental results • Algorithm **not** publicly available
Waukesha knock index (WKI)	0-100% (?)	Stationary engine	• Publicly **available** via Website • Available for the full hydrogen range (0-100%) • Algorithm publicly available	• **Large** (relative) difference in MN compared with other methods for natural gas
Cummins Methane Number (CMN)	up to 0.03%	Unknown	• Publicly available	• **Not suitable** for (high) hydrogen content in natural gas • Access only via website
Wärtsilä Methane Number (WMN)	0-30%	Ultra-lean-burn marine engine (□~2)	• Available for (limited) hydrogen percentage • Publicly available (website)	• **Limited** hydrogen percentage • No offline version
PKI Methane Number	0-20%	Lean-burn CHP engine (□=1.66)	• Limited hydrogen percentage up to 20% H_2 in CNG • **Open access** • **Simple** to implement polynomial equation • **Easy** to expand the range of hydrogen percentage in CNG • ISO 23306 standard (LNG as marine fuel)	• **Limited** hydrogen percentage

2.4.5.3. Comparison methane number calculations with measurements

Literature review revealed that the amount of experimental gas engine knock data is very scarce and only limited measurement data are available in the literature that can be used to test (qualitatively) the accuracy of the above mentioned methane number methods for natural gas/hydrogen blends [55-56]. As an illustration we compared here the measured knock resistance (KLST) in a lean-burn gas engine [55] and calculate for the corresponding gas mixtures the methane numbers using the different methods (see Appendix C for gas mixtures, KLST and methane number values). The results of the comparison presented in Figure 19 show that the methane numbers calculated with the WKI MN, PKI and the WMN methods linearly decrease with increasing hydrogen percentages in the fuel as also observed in the measurements. In contrast, as also shown above the AVL MN show an increase in the Methane Number up to 3% hydrogen and the MWM Methane Number methods show only limited sensitivity when relatively small fractions hydrogen (<3 mole% H_2) are present in natural gas.

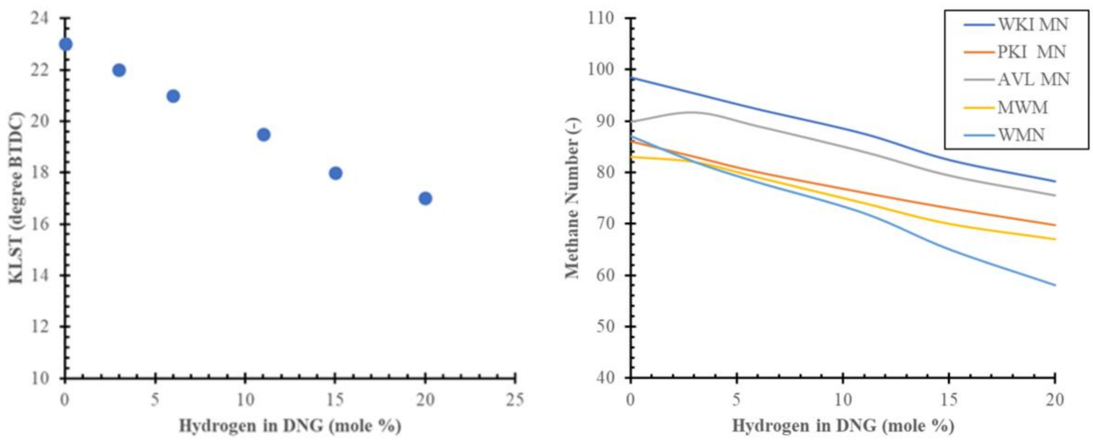

Figure 19 (Left) Measure knock resistance expressed in knock limited spark timing (KLST) for different fractions hydrogen in natural gas in fuel lean-burn (λ=1.55) high speed medium BMEP gas engine (1500rpm) [55]. Right: Calculated methane number using different methane number method available in the literature.

For stoichiometric heavy-duty vehicle engines, to our knowledge no CNG/H_2 knock data are available. However, in [2] the knock resistance of different LNG compositions was tested on a stoichiometric CNG truck engine (FPT cursor 9). The measured knock resistance (Knock Limited Spark Timing, [2]) was compared with PKI, AVL and MWM calculated methane numbers, see for details Appendix C. The results show that both the AVL and PKI MN method show good agreement with the knock measurements while the MWM method show large differences which is not surprisingly since the MWM method is optimized for fuel lean engines

The literature review shows that hydrogen addition lowers the knock resistance and thus increases the risk of the occurrence of engine knock. The validity of existing methods to accurately predict the knock behavior of CNG/H_2 blends in stoichiometric heavy-duty CNG engines is currently unknown. For this reason, it is highly recommended to experimentally investigate the knock behavior of CNG/H_2 mixtures in CNG engines and compare the results with the outcome of the above discussed methane number methods. This offers an opportunity to

select the methane number method that unambiguously calculate the impact of hydrogen addition to natural gas on the occurrence of engine knock.

2.4.6. Effect on spark plug

The effect of hydrogen mixture in natural gas on the performances of the ignition system and more specifically the spark plugs in NGVs, includes several (interrelated) topics.

A first topic concerns the design of the spark plug. As discussed, hydrogen mixture in natural gas tends to make engines more prone to pre-ignition/backfire induced from hotspots in the combustion chamber. One such possible hotspot is the spark plug. In traditional, so-called J-type, spark plugs, the centre and ground electrode(s) protrude into the combustion chamber. The electrodes assume a certain temperature profile depending on heat input from combustion and heat rejection to the base of the spark plug i.e. heat sink. Spark plugs come in different heat grades i.e. different heat rejection rates for the purpose of 'controlling' spark plug temperature for the specific engine application. If chosen too hot, the spark plug will wear more rapidly and induce the risk of pre-ignition, if selected too cold, the spark plug will foul from depositing. Given the lower ignition energy and the higher (peak) combustion temperatures associated with hydrogen combustion as compared to natural gas, it is widely suggested to select cold-rated spark plugs [8, 67-68] to avoid pre-ignition in 100% hydrogen-fuelled engines.

BMW [69], in 2006, issued a patent for a J-type spark plug specifically designed for hydrogen engines. Its design comprised of two rounded rather than sharp-edged mass electrodes for reduced surface/volume ratio of the electrodes. This design was claimed to improve heat rejection, and hence to reduce the risk of pre-ignition. BMW did not use said spark plug design in its bi-fuel engine for the BMW Hydrogen 7 car [50]. Instead, they used a so-called surface-ignition type spark plug. In this type of spark plug, there is no protruding ground electrode(s), thus avoiding the ground electrode hotspot issue. We remark that most, if not all, spark plugs used in NGVs are J-type spark plugs.

It should be noted that selecting cold-rated spark plugs can itself introduce a risk of pre-ignition as the electrode temperatures can be too low to effectively burn deposits off, with the deposits acting as (glowing) hotspots.

A second topic concerns the materials used in the firing end of the spark plug. Modern spark plugs in high-performance engines typically use platinum in the ground electrode, and sometimes also iridium in the centre electrode, for better wear/erosion resistance i.e. extended spark plug life. A renowned HD truck OEM confirmed the use of such spark plugs in their LNG/CNG vehicles. References [8, 67] mention previous work indicating that platinum in spark plugs for 100% hydrogen-fuelled engines could induce pre-ignition on account of its catalytic activity for oxidizing hydrogen in the presence of air. A leading spark plug OEM also noted this as a possible risk, referring to [69]. In addition, they point to iridium possibly posing a similar risk.

A global supplier of ignition systems further mentioned hydrogen embrittlement as possibly relevant for the integrity of spark plugs. Hydrogen-induced cracks in the firing end of spark plugs not only pose the risk of mechanical degradation but can also introduce hotspots from electrode sections for which cracks hamper proper heat rejection.

The lifetime of spark plugs is a key factor in service intervals for NGVs. It is sometimes suggested that spark plug lifetime could benefit from hydrogen mixture in natural gas on account of the favourable ignition properties of hydrogen i.e. low ignition energy and wide ignition range. A higher ignition energy could enhance the erosion of the spark plug reducing its lifetime. Figure 4 gives proof to the beneficial effect of hydrogen mixture in methane on the minimum ignition energy.

This could mean lower spark energy would be sufficient to retain stable ignition of the hydrogen-containing cylinder charge as compared to a cylinder charge with 100% natural gas as fuel. Lower spark energy would mean lower erosion/wear of the spark plug electrodes and hence a longer service interval.

Another way is that the favourable ignition properties of hydrogen could be used to decrease the (initial) spark plug gap without compromising stable ignition and related engine performances. A smaller spark plug gap means by definition lower spark voltage, and also lower erosion/wear of the spark plug electrodes and possibly more gap margin, with both effects being beneficial for spark plug life.

Ghent University [8, 70] claimed that, in dedicated hydrogen engines, a smaller (initial) spark plug gap is required to reduce the otherwise required increased spark voltage as a consequence of the lower ion concentration between the electrodes in hydrogen-containing cylinder charges. They further advised that a too small gap could induce engine cold-start issues from water condensation at the spark plug electrodes. The aforementioned spark plug supplier and a second, large ignition system supplier both supported the suggestion of reducing the spark plug gap in the case of hydrogen as engine fuel. The spark plug OEM believed that have a beneficial effect on the spark plug lifetime without providing evidence, while the ignition system supplier, having just started related R&D, said to have not seen evidence (yet) to suggest changes in spark plug erosion characteristics when using hydrogen mixture in natural gas.

TIAX [71] investigated the potential of reducing the ignition/spark energy when mixing hydrogen in natural gas fuel for the purpose of an expected associated reduction in spark plug wear. Using a lean-burn test engine operating at a lambda of 2.0 i.e. 100% air excess, they could reduce the ignition energy by 0, 7, 16, 22 and 27% for hydrogen fractions of 23, 27, 27, 32 and 38% vol in natural gas respectively, apparently without compromising engine-out NO_x emissions and combustion stability. These findings, without considerations regarding the experimental approach taken and analysis performed, do support the idea that hydrogen mixture in natural gas could extent spark plug lifetime through a reduction in ignition energy. We note that a reduction in spark plug gap as discussed above could further increase lifetime, although a reduction in spark plug gap could limit the amount of reduction in spark energy allowed to sustain stable engine performances, and vice versa.

The first mentioned ignition system supplier, being involved in projects with hydrogen engines, suggested a possible requirement for higher ignition energy following the introduction of hydrogen as a fuel. This requirement would come from the leaner air-fuel mixture required to counteract an otherwise increase in engine-out NO_x emissions and more knock-prone combustion when introducing hydrogen. While stoichiometric rather than lean-burn combustion is the norm in modern NGVs, this supplier expected a similar effect in stoichiometric engines with EGR requiring a higher EGR fraction and hence a higher ignition energy to counteract the said NO_x and knock effects when introducing hydrogen in the fuel blend.

The same ignition system OEM further pointed out that active control of the ignition energy will require capacitive-type ignition systems rather than the (cheaper) inductive-type ignition systems typically used in light-duty and medium-duty natural gas vehicles. Modern HD NGVs most probably already typically use capacitive-type ignition systems capable of controlling the ignition energy.

In conclusion, at least for 100% hydrogen engines, it is generally assumed that the heat grade of and materials used in spark plugs need careful consideration. Regarding possible (initial) spark plug gap and ignition energy reduction and their effect on spark plug lifetime when introducing hydrogen in the fuel mix of NGVs, there is less consensus. Further, dedicated research should address the following issues:

- **Do precious metals in spark plugs increase a risk for pre-ignition when mixing hydrogen in natural gas, and if so, from what hydrogen fraction and under what operating conditions?**
- **Do typical spark plug firing end materials suffer from hydrogen embrittlement?**
- **Regarding the heat grade of spark plugs, is there a balance to be found between avoiding too high electrode temperatures and too high deposit rates?**
- **Can the spark plug gap and/or ignition energy be reduced when mixing hydrogen in natural gas, and if so, from what hydrogen fraction, and, to what extent is the spark plug lifetime affected?**

We note that the aforementioned spark plug OEM has expressed an interest in join performing research on the effects of hydrogen mixture in natural gas on spark plug integrity and performances.

2.4.7. Effect on CNG tank system

Based on interviews and literature review [72-73], the following four different tanks for storing CNG are available.

- **Type 1**: All metal construction, generally made of steel. One of the least expensive tank designs, but also one of the heaviest. Interviews reveal that this type is not sold anymore in Europe.
- **Type 2**: Thinner metal wall as compared to type 1 tanks; the core is wrapped with a fibre-reinforced polymer to provide reinforcement of the cylinders. Slightly lighter than type 1 tank.
- **Type 3**: Aluminium core wrapped with carbon fibre composite, much lighter than types 1 and 2. Interviews with manufacturers reveal that this type has a restriction in the length/diameter ratio by the material properties of aluminium and hence are often bulky. This type of tank is not widely used.
- **Type 4**: Polymer membrane with carbon fibre or carbon/glass fibre wrapped around. This tank is the lightest tank available on the market and has no restriction in length/diameter ratio. New CNG vehicles are equipped with this type of tank. Type 4 tanks are also used for storing pure hydrogen.

An issue with steel tank is the possible occurrence of hydrogen embrittlement, which is the case for type 1 tanks. Although these tank types are not sold anymore, it could be possible that current vehicles have this tank type. As mentioned above, the regulation 110 of the Economic Commission for Europe of the United Nations (UN/ECE, [14]) put a stringent limit of 2% by volume for steel tanks with a tensile strength exceeding 950 MPa [11, 74-75], which is often the case because these materials allow thinner walls and hence, lighter tanks [76]. Interviews and literature review reveal that this limit is seen as conservative and possibly higher percentages (10-12 vol.% H_2) for steel tanks are possible. However, further research is needed to investigate this.

Interview with OEMs reveal that the type 4 tanks for pure hydrogen and the ones for CNG are constructed from the same material. However, to be able to withstand the much higher pressure at which pure hydrogen is stored (typically 350-700 bar) different sealing are used. It is not expected that replacement of CNG by CNG/H_2 or H_2 while maintaining the maximum operating pressure of 250 bar for CNG will cause any issues with the CNG tank itself. However, it is yet unknown if the periphery (such as valves, sealing materials, etc.) can safely handle H_2/CNG blends. The complete tank system must be checked whether it is able to handle hydrogen percentages in CNG and/or store the gas at a different pressure. Manufactures of tank systems indicate that new standards for H_2/CNG blends are necessary. As an alternative it was suggested to use the same standards (certification) as used for pure hydrogen components. The disadvantage of this route is that these hydrogen standards are more stringent than those for CNG which may result in more expensive materials/equipment to meet the requirements (e.g. [82-83]). However, the advantage is that no new standards must be developed for H_2/CNG blends.

The regulation 110 (UN/ECE) puts a limit of 2 vol.% H_2 for most steel tanks (type 1, > 950 MPa). Type 4 tank can handle both CNG and hydrogen. The type 4 tank itself, should not have any problems with mixtures of H_2 and CNG provided that the maximum operating pressure is not changed. However, it is unknown if the periphery (valves, sealing material and so on) can handle mixtures of hydrogen and CNG. It is recommended to further investigate this.

2.5. FUEL LINE

According to OEMs the materials and parts used fuel line system such as the pressure regulator, connecting pipe and fuel rail with injectors may have to be upgraded to avoid hydrogen embrittlement when hydrogen is present in CNG depending on the materials. Although pressure regulators and fuel injectors are available on the market that already are compliant with pure hydrogen, feedback from interviews revealed that not much is known about the effect of hydrogen at high pressure for the existing CNG components currently present in the fuel line. For example, experiments performed by one of fuel line supplier demonstrated that former components were compatible up to 10 vol.% hydrogen in the blends. For the current components sold on the market the same fuel line supplier indicated a maximum allowable fraction of hydrogen of 30 vol.% in CNG. An alternative approach suggested by one of the manufacturers is to replace the components by ones that are compliant with pure hydrogen. However, these components are often more expensive than the CNG versions.

Furthermore, the performance of the injectors will have to be revised to be compliant with different sonic speed, density and energy density when hydrogen is added to CNG. As an illustration the energy flow through the injector is calculated with increasing hydrogen content in CNG by considering the changes in energy density and sonic speed. The results presented in Figure 20 show that the energy flow decreases with increasing hydrogen percentages in CNG up to about 80 mole% hydrogen in CNG, mainly driven by the change in energy density; it increases beyond 80 mole% hydrogen in CNG, mainly driven by the quadratic increase in sonic speed. Interviews with OEMs indicate that for hydrogen percentages above 15-20 mole% that recalibration of engine mapping is needed.

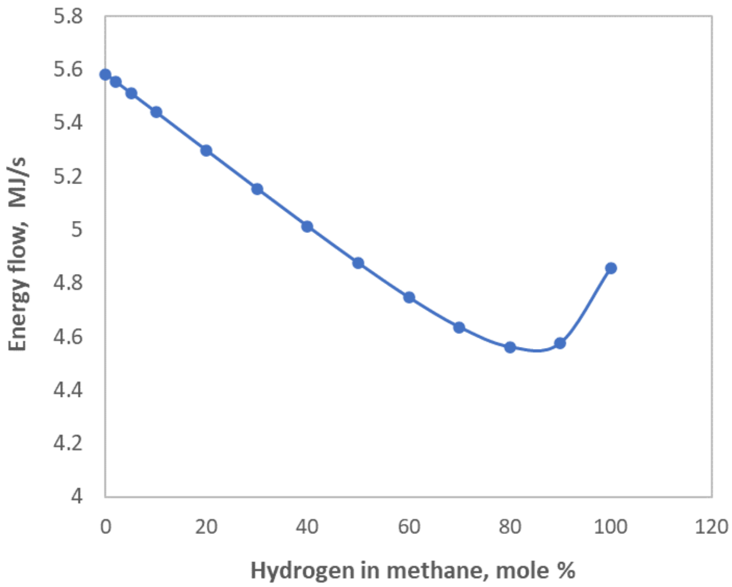

Figure 20 Calculated energy flow through fuel injector for varying hydrogen volume percentages in methane (calculated at 15˚C and 200 bar).

2.6. OTHER ISSUES

2.6.1. Crankcase ventilation/safety

In modern turbo-charged engines operating above approx. 50% engine load, there is a positive pressure difference between the cylinder and crankcase during the entire working cycle. The same is true at loads under around 50%, except for the intake phase of the cycle. This pressure difference causes a fraction of the cylinder charge to leak past the piston rings into the crankcase, known as blow-by. In premixed engines, i.e. engines in which the fuel gas and combustion air are mixed before entering the cylinder, this blow-by comprises of both fresh fuel/air mixture and combustion products. In addition, above around 50% engine load there is a mixture of fresh air and combustion products leaking from the turbocharger into the crankcase via the turbocharger lubrication oil return circuit. Effectively, the crankcase atmosphere contains varying fractions of unburned fuel gas (and air and combustion products), depending on engine load and engine wear primarily. In the case of hydrogen mixture in natural gas, the crankcase atmosphere will thus also contain hydrogen in addition to natural gas (or hydrogen only in the case of 100% hydrogen fuel). The higher the hydrogen fraction in the fuel gas, the higher the hydrogen fraction in the crankcase atmosphere.

Research by DNV on a lean-burn gas engine has shown hydrogen fractions in the crankcase atmosphere of up to 3% by volume for a fuel gas containing 35% hydrogen [77]. Ghent University reported hydrogen fractions exceeding the analyser upper range limit of 5% by volume for an automotive-type lean-burn engine running at 100% hydrogen [70].

Modern CNG vehicle engines have crankcase breather systems in which this blow-by stream, after being separated from lubricating oil, is piped back into the low-pressure section of the air intake system of the engine. The blow-by stream is subsequently burned, thus preventing fuel loss (and related direct emissions). However, hydrogen in the crankcase atmosphere could pose an increased risk of crankcase explosions, given the favourable ignition properties of hydrogen as compared to natural gas.

Recently, CIMAC addressed the issue of increased risk of crankcase explosions in marine-type gas and dual-fuel engines as compared to traditional oil-fuelled marine engines in a position paper [78] for the purpose of supporting (safety) rule-making by IACS. It was concluded that the introduction of fuel gas in the crankcase did not increase crankcase explosion probability or explosion severity. We note that the CIMAC analysis did not involve hydrogen as fuel gas component. We further note that marine safety rules already command mitigating measures such as crankcase explosion release valves and hot-spot detection.

BMW, in its bi-fuel engine for the BMW Hydrogen 7 car, installed a 'shut-off valve' in the crankcase breather system to prevent crankcase explosions induced from backfire in the intake system [50].

In conclusion, crankcase ventilation/safety in relation to hydrogen combustion commands consideration, as exemplified by the safety measures incorporated in BMW's hydrogen car. However, detailed information on this issue, and especially for engines with (variable) mixture of hydrogen in natural gas, seems partially available from the field. Given the physical and reputational damage involved with possible crankcase explosion incidents in NGVs, it is advised to further investigate the risk (and if needed mitigation measures) of crankcase explosions associated with hydrogen mixture in CNG vehicle-type engines. Research questions to be addressed are:

- **What hydrogen fractions to expect in CNG vehicle-type engine crankcases?**
- **What parameters affect this hydrogen fraction: fuel composition/hydrogen fraction, engine operating conditions, engine wear & tear, crankcase breather system design …**
- **Does hydrogen mixture affect the blow-by rate (low density versus low quenching distance effect on piston top-land crevice leak rate, …)?**
- **For the resulting hydrogen fractions, what ignition energy/component temperatures could induce crankcase explosions, and with what severity?**
- **If required, what are the mitigating measures?**

2.6.2. Lubricating oil

As discussed in Section 3.6.1, hydrogen and combustion products enter the engine crankcase as a result of blow-by. This means that hydrogen and combustion products can interact with the lubricating oil, possibly affecting the properties and lifetime of the lubricating oil.

In relation to its Hydrogen 7 car, BMW emphasized its efforts in minimizing blow-by to avoid adverse effects from a high hydrogen and water content in the crankcase [50]. Hydrogen, through its combustion product water as part of the blow-by stream, increases the water content in the crankcase. Depending on the engine temperature, i.e. cold starts and engine cool down versus hot engine running, the water will appear in the crankcase in gaseous and/or liquid state. Either way, the lubricating oil will need to accommodate the higher water content, without suffering loss of performance. One possible issue is the lubricating oil and water forming an emulsion, which can adversely affect oil pump performance thereby impacting the thermal load and lifetime of critical engine components. Ghent University and Argonne National Laboratory [61] refer to work citing the advised use of lubricating oils with improved demulsifying properties when using hydrogen as engine fuel.

In addition to water affecting the lubricating oil, it is also reported that hydrogen in the crankcase impacts critical performances of the lubricating oil from chemical interactions. Ghent University [70] compared the properties of a given lubricating oil used in experimental work on a hydrogen engine with those of the fresh oil. They saw a decrease in lubricating properties as indicated by a significant decrease in the concentration of lubrication and wear-resistance promoting additives. They also observed changes in the viscosity (index) of the lubricating oil, considered to effect higher friction during engine cold start and poor lubrication at normal engine operating temperatures. It was advised to apply motor oils dedicated for use in hydrogen engines (although those were not available at the time). In line with the above advice, BMW [50] stated that the engine oil quality selection had to be optimised for the hydrogen operating mode of their bi-fuel hydrogen car engine.

The selection of lubricating oil for hydrogen-burning engines should further consider the ash-forming properties of the oil. While some ash formation and -deposition is considered beneficial for the lifetime of critical engine components such as valves and valve seats, such depositions can also form hot-spots in the combustion chamber inducing pre-ignition or backfire in the intake system when burning hydrogen-containing fuels.

In conclusion, the introduction of hydrogen in the fuel mix for NGVs raises the question whether the engine lubricating oils in use in today's NGVs are fit for purpose. As with the issue of crankcase safety, this topic is addressed in relevant publications, but not into high detail. With possible positions ranging between simply selecting dedicated lube oils, if available, or accepting reduced engine oil change intervals and/or reduced lifetime of critical engine components, it is advised to consult lubricating oil suppliers on this topic. Questions to be answered are:

- **Does hydrogen in the crankcase atmosphere (adversely) affect lubricating oil performances, and if so, from what fraction?**
- **Does the increased water fraction in the crankcase atmosphere (adversely) affect lubricating oil performances, and if so, from what fraction?**
- **If required, are fit-for-purpose lubricating oil alternatives available? And if not (readily) available, what impact on the lifetime of the lubricating oil and/or critical engine parts to expect?**

2.7. FUEL ADAPTIVE FEED FORWARD ENGINE CONTROL

As shown above the presence of hydrogen in CNG can cause combustion performance issues such as engine knock and increased NO_x emissions. The introduction of hydrogen outside the fuel specification of the engine requires that engine manufacturers ordinarily must either derate the engine or restrict the range of fuels that can be supplied to the engine. Restricting the range of fuels results in either a limitation of the supply options for the end user or increased processing cost for the fuel supplier, or in (structural) reduction in engine performance. A better solution for both fuel suppliers and end users is the real-time adjustment of the engine settings based on the measured composition of the fuel that enters the engine. The advantage of such a feed-forward fuel-adaptive engine control system [79] is that the engine only will be adjusted from its optimal setting (maximum power and efficiency) when the methane number is higher or NO_x emission is lower than specified.

As discussed, one of the engine setting to mitigate the risk of the occurrence of knock and to reduce the NO_x emission is retarding the spark timing. As an illustration we used the engine model to simulate the NO emission (Fig.21, left) and knock resistance (Fig. 21, right) while retarding the spark ignition [80]. The simulation results show that the NO emission reduces substantially when retarding the spark timing. For example, retarding the spark timing from 14 to 10 °CA BTDC the NO emission of the H_2/DNG (20% H_2) is reduced to values similar to that of pure DNG (14 °CA BTDC). Furthermore, retarding the spark timing mitigates the risk of engine knock occurrence as illustrated in Figure 21 (right). For example, no knock is observed when retarding the spark timing from 14 to 10 °CA BTDC.

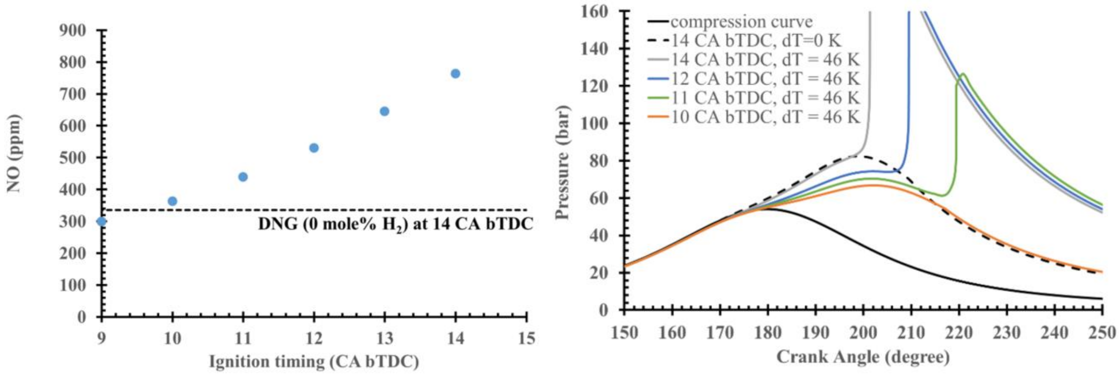

Figure 21 Simulated effect of varying the spark timing on the NO emission (Left) and the knock resistance (Right) for 20% mole H_2 in DNG. “dT” relates to the relative increase in intake temperature compared to the reference dT = 0K. This artefact was used to trigger knock in the simulation base case, which would otherwise not occur.

In [79] such a fuel adaptive control system was tested for LNG gases and the schematic of the test is presented in Figure 22. The results show that by applying the fuel adaptive control system the engine is used for a broad range of fuel compositions and a maximum of 6% fuel savings was observed. It is strongly recommended to study the system for H_2 containing gases and include NO_x control strategy for heavy-duty engines. One of the challenges is to select a fast response, cheap and robust gas quality analyser. An inventory of such analysers will be discussed in Chapter 5.

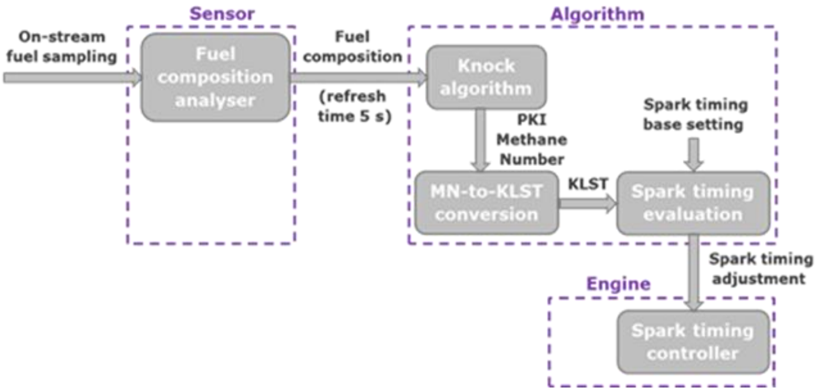

Figure 22 Schematic of a feed forward fuel adaptive control system [80].

2.8. EFFECT HYDROGEN ADDITION VEHICLE RANGE, GREENHOUSE GAS EMISSION AND BILLING

An important parameter for (truck) drivers is the distance travelled by the vehicle on a single tank of fuel (vehicle range). Changes in the CNG quality can cause changes in the energy density of CNG which subsequently impacts the vehicle range. As shown in Figure 23 the addition of hydrogen reduces the energy density[8] of the CNG present in the fuel tank at 200 bar as a result of the change caloric value of the fuel (see Table 4) and the change in compressibility of the fuel. Furthermore, the fuel quality can also impact the efficiency and thus the amount of fuel consumed.

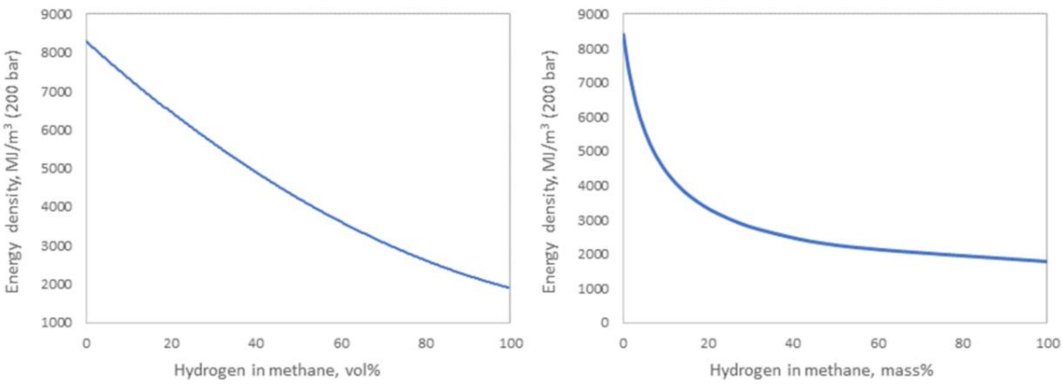

Figure 23 Energy density in the tank at a pressure of 200 bar for different CH_4/H_2 blends by volume and mass (see also Appendix E).

As illustrated in Figure 24, one of the benefits of the blending CNG with hydrogen is the reduction of CO_2 per unit energy of the fuel as a result of the change in C/H ratio. As discussed above, hydrogen addition can improve the energy efficiency which can results in a further reduction of the CO_2 emission. Furthermore, the methane slip also reduces when hydrogen is added to CNG (see paragraph 3.4.4).

[8] The energy density in the CNG tank is calculated using the lower heating value (Hi) shown in Table 4 and the compressibility factor (z) at 200 bars.

The combined effect of CH_4 and CO_2 reduction with increasing percentages hydrogen in CNG results in a reduction of the total greenhouse gas emission.

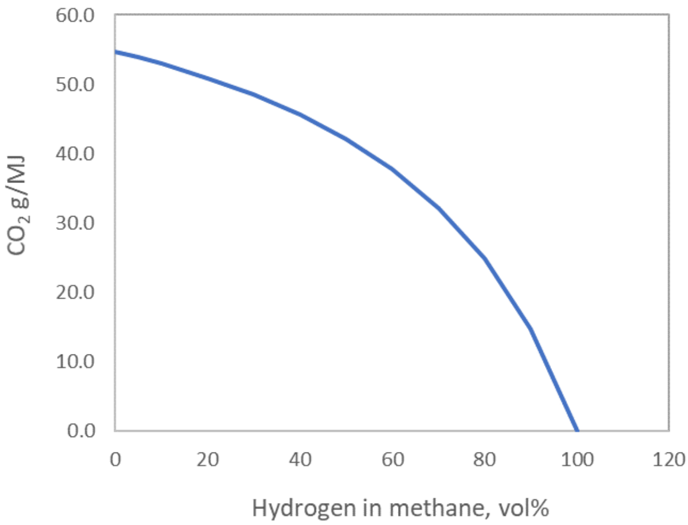

Figure 24 Hydrogen percentage in methane as function as the CO_2 emission

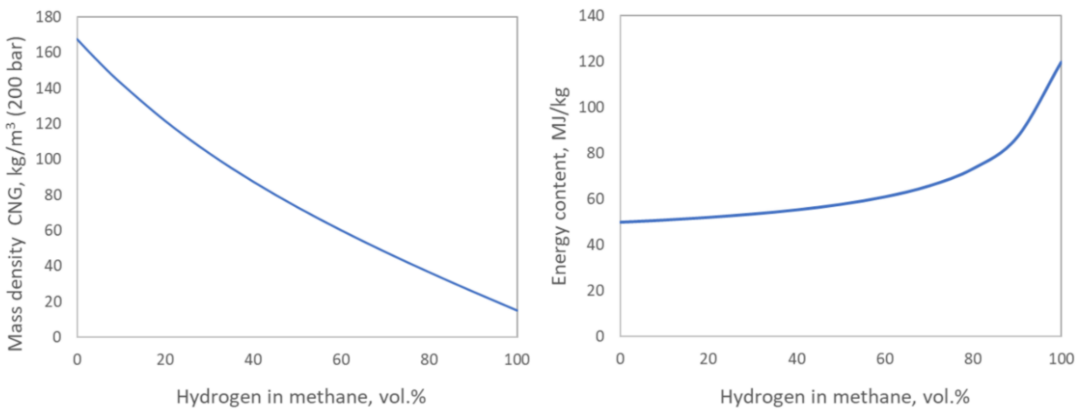

Figure 25 (Left) Mass density of various H_2/CNG blend at 200 bars. Right: Energy content expressed MJ/kg for different H_2/CNG blends.

The billing of CNG at fuelling stations is mass based. However, when hydrogen is added to CNG both the mass density (Figure 25, Left) and the energy density (Figure 23) of the fuelled CNG changes substantially. Consequently, the energy content fuelled, expressed in MJ/kg changes with increasing hydrogen percentages present in CNG as shown in Figure 25, right. From this we conclude that the energy content per kg increases with increasing amounts of hydrogen.

2.8.1. Case study

To estimate to what extend the addition of hydrogen will impact vehicle range, greenhouse gas emissions and billing, an illustrative case study is presented using as input the findings from the literature review described above. The case study is performed for a reference gas of pure methane and for a gas containing 20 vol.%

hydrogen in methane. In the case study we make two assumptions: 1) as a conservative assumption we assume that the addition of hydrogen will not affect the fuel consumption, 2) we assume, based on the findings described in the literature (see above) that the fuel consumption decreases by 13% (best case) when adding 20 vol. % hydrogen to CNG [25]. Furthermore, we assume based on the finding in the literature that the methane emission will be reduced with 30% and a driving distance of 635 km/m^3(n) for CNG.

Table 8 Input calculations case study using the reference gas CH_4 and H_2/CH_4 (20 vol.% H_2)

	Conservative case 1	Best case 2
Tank volume, m^3	1575	1575
Pressure, bar	200	200
Fuel price, euro/kg	1	1
Fuel price (Hi), Euro/MJ	0.020	0.020
Fuel price (Hs), Euro/MJ	0.018	0.018
Methane slip (CNG as fuel), g/kWh	0.5	0.5
CH_4 emission reduction using H_2/CNG (20 vol% H_2)	30%	30%
Fuel savings using H_2/CNG (20 vol% H_2)	0%	13%

<u>Vehicle range (mileage):</u>

As shown in Figure 26 the driving distance decreases from 1000 km to 757 km when 20 vol.% hydrogen is present in CNG (CH_4) assuming no impact on fuel efficiency. The literature study reveals that in best case about 13% fuel efficiency can be expected when 20% hydrogen is present in CNG which extends the driving range to 855 km.

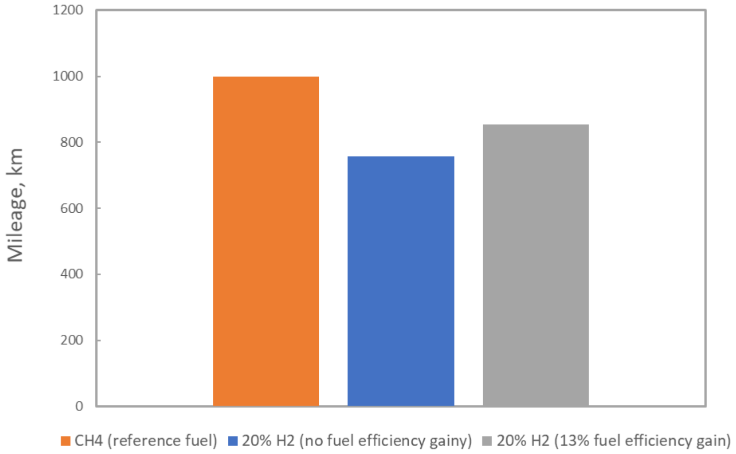

Figure 26 Vehicle range (mileage) for pure methane and 20 vol.% hydrogen assuming 1) similar fuel efficiency 2) 13% increase in fuel efficiency

From this case study we conclude that in the most conservative case (no fuel efficiency gain) a vehicle range reduction of about 24% can be expected. In best case (13% fuel efficiency gain) a vehicle range reduction of about 14% is expected when 20 vol.% hydrogen is present in natural gas.

Greenhouse gas emission (GHG)

The addition of hydrogen to natural gas will reduce the carbon content and thus the GHG emission measured at the tailpipe. When assuming no change fuel efficiency upon hydrogen addition to CNG, the CO_2 emission will decrease with about 7% when 20 vol.% hydrogen is added to methane. From gas engines it is known that the methane emission also contributes to the greenhouse gas (GHG) emission. Based on the Euro 6 requirements we assume that heavy-duty vehicles have a methane emission of 0.5 g/kWh which equals to about 14 g/kWh CO_2e[9]. According to the literature review the methane emission (engine out) is reduced with increasing hydrogen percentages in the fuel. Although the literature review reveals that the methane emission reduction depends upon the engine setting. In this case study, we assume that the presence of 20 vol.% hydrogen in CNG reduces the methane emission with 30% [44]. The effect of the presence of 20 vol% hydrogen in methane is presented in Figure 27. When assuming no efficiency gain the presence of 20 vol.% hydrogen in methane will reduce the GHG by about 8% and a GHG emission reduction of about 20% is obtained when assuming a fuel efficiency gain of 13% [25].

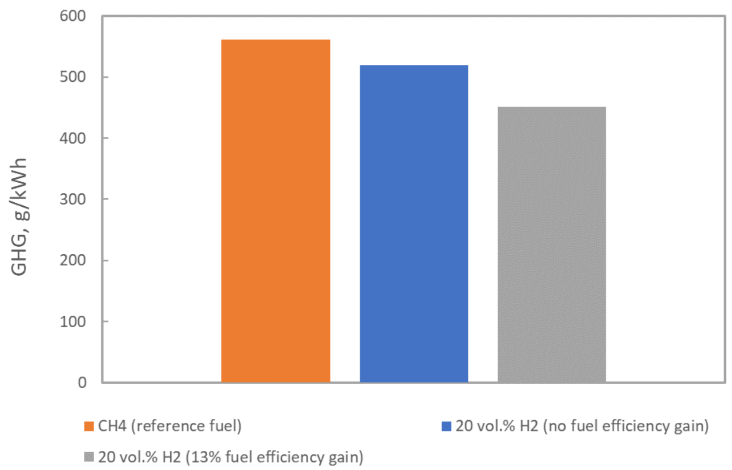

Figure 27 GHG for pure methane and 20 vol.% hydrogen assuming 1) similar fuel efficiency 2) 13% increase in fuel efficiency

2.8.2. Billing

Currently, billing of CNG is based on the amount of mass (kg). When hydrogen is added to pipeline gas the ratio MJ/kg (see Figure 25) will change which consequently will impact the billing of CNG. As an illustration the fuel price expressed in Euro/kg is presented in Figure 28. As reference the fuel price of CNG is shown in the orange bar based on the assumptions present in Table 8. When assuming that the presence of 20 vol.% hydrogen does not have an impact on the fuel efficiency and the accuracy of the mass flow meters installed at the fuelling station the fuel price drops from 0.264 Euro/km (CNG) to 0.253 Euro/km (HCNG, 20 vol.% H_2). This 4% price difference is the result of the change in MJ/kg when hydrogen is added to CNG. The price difference increases with increasing amount of hydrogen in the fuel as shown in appendix B. When assuming (best case) a fuel efficiency of 13% when 20 vol.% hydrogen the fuel price will drop to 0.22 Euro/km.

[9] Assuming that methane has a 28 times stronger effect on the climate than CO2

As an alternative the CNG market might consider to base the billing by using the energy content of the mixtures. However, this requires the determination of the energy content of the CNG at the filling station.

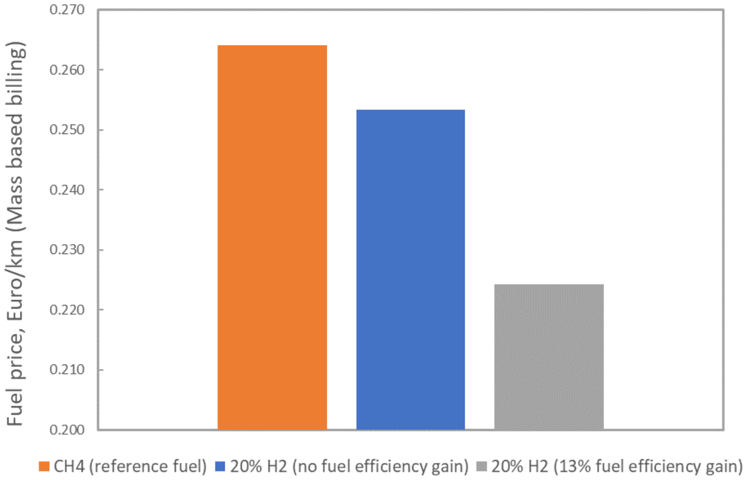

Figure 28 Fuel price (km/Euro) for CNG and 20 vol.% hydrogen when using mass-based billing assuming 1) similar fuel efficiency 2) 13% increase in fuel efficiency

2.9. SUMMARY & RECOMMENDATIONS LITERATURE REVIEW

Summary

- *Market analyses* shows a trend towards applying stoichiometric engines in the European heavy-duty CNG vehicles.
- *In-cylinder conditions:* Hydrogen addition to CNG result in an increase in the maximum pressure generally (and temperature) which can affect engine wear, efficiency, emissions and engine knock. Generally, 20 vol.% hydrogen in CNG increases the peak pressure between 6-20% depending on the engine settings.
- *Combustion stability:* Hydrogen addition improves the combustion stability reducing the risk of misfire and allows to extend the fuel lean limit.
- *Efficiency:* Hydrogen addition has the potential to significantly improve the efficiency and hence result in fuel saving. However, the increase in efficiency strongly depends upon the variation of engine parameter settings that is controlled by the ECU such as the air factor, spark timing etc. In the literature described above often multi-parameter variation are applied in different engine types which makes it difficult to draw quantitative conclusions with regards to the effect of hydrogen addition on the thermal efficiency. However generally we observe an efficiency gain between roughly 0-4 point-% and fuel saving gain of about 0-13% for 20 vol.% hydrogen addition.
- *Emissions (pre-catalyst):* Hydrogen addition to CNG results in an increase in the engine out NO_x emission and a decrease in the engine out CO, CH_4 and C_xH_y emissions. The quantitative changes in pollutant emission were found to depend upon the engine settings and conditions. The presence of a fixed amount of

hydrogen in CNG together with optimization of the engine settings offers opportunities to meet the increasingly stringent emission regulation for both stoichiometric and lean-burn engines.

- *Vehicle range (mileage):* The addition of hydrogen to CNG reduces the vehicle range. From this case study it is concluded that in the most conservative case (no fuel efficiency gain) a vehicle range reduction of about 24% can be expected. In best case (13% fuel efficiency gain) a vehicle range reduction of about 14% is expected when 20 vol.% hydrogen is present in natural gas.
- *Greenhouse gas emission* (GHG): Hydrogen incorporation reduces the carbon footprint of engines in three ways. First, less CO_2 is produced per unit of energy input; second, less methane slip is emitted; and third there is a potential increase in engine efficiency resulting in lower GHG emission. When assuming no efficiency gain the presence of 20 vol.% hydrogen in CH_4 will reduce the GHG emission by about 8 vol.% and GHG emission reduction of about 20% is obtained when assuming efficiency gain of 13%.
- *Billing:* When hydrogen is added to pipeline gas the ratio MJ/kg will change which will consequently impact the mass-based billing of CNG.

In the Table below the hydrogen limits for the different engine parts are summarized based on the literature review and interviews with manufacturers. More detailed information is given below.

Table 9 H_2 limits, knowledge gaps and recommendations for the different engine parts

Engine parts heavy-duty engine	H_2 limit	Knowledge gaps	Recommendations
Catalyst/emissions	Unknown	Lack of (research) data on total system (engine + TWC) behaviour; possible unwanted emissions (NH_3 and N_2O)	Generating practical experience with hydrogen blending using relevant heavy-duty vehicle, in joint effort with relevant OEMs
Spark plug	Unknown	Effect of hydrogen on firing end materials; heat grade selection; no consensus on potential of spark plug gap and ignition energy reduction for lifetime extension	Consult with spark plug OEMs for joined research.
Lubricating oil	Unknown	Effect of hydrogen and water in crankcase atmosphere on lube oil performances and lifetime; if needed, availability of suitable product	Consult with lube oil OEMs for joined research.
Fuel line	10 vol% for former components 30 vol% for new components	Information in this study is provided by a single manufacturer. It is unknow what the opinion is of the other manufacturers (no response).	Generating practical experience with hydrogen blending using relevant heavy-duty vehicle
CNG tank system	Steel tank: 2 vol% (UN/ECE R110) Type 4 tank: tank itself can handle full range 0-100 vol% H2 (when using identical pressure as CNG). However, the tank periphery components may be the limiting factor.	Unknown if periphery components can handle hydrogen/CNG blends and by how much hydrogen failure occurs	Development of dedicated standards for HCNG for tank periphery components. If needed replace all steel tanks (type 1) from the market Generating practical experience with hydrogen blending using relevant heavy-duty vehicle
Engine ECU/Injector	15-20 vol%, for higher percentages recalibration of the engine mapping is needed	Unknown how the ECU and stoichiometric heavy-duty engine response to >20 vol% hydrogen in CNG	Generating practical experience with hydrogen blending using relevant heavy-duty vehicle
Engine knock	Depending upon the CNG quality and engine specification (MN)	Knock sensitivity for H2/CNG blends in CNG engines Unknown how accurate the knock resistance can be predicted of H_2/CNG blends using existing MN methods for stoichiometric heavy-duty engines	Studying effect hydrogen blending on knock behaviour in a CNG engine Exploring knock mitigating measures while maintaining efficiency when H_2 is added. Studying the accuracy of existing MN methods and if needed development of a dedicated MN algorithm for relevant CNG engines

- *Catalyst performance:* exhaust gas aftertreatment systems, and particularly TWCs, are expected to be impacted by the introduction of hydrogen in the fuel mix for CNG vehicles in terms of changes in their operating conditions and in the upstream exhaust gas composition matrix. Apparently, research related to a total system approach on tailpipe emissions also involving typical aftertreatment systems is few or is just being started. This, and a concern regarding possible unwanted emissions (NH_3 and N_2O) commands continued careful consideration of the performance of CNG vehicle aftertreatment systems when admixing hydrogen into natural gas. It is advised to consult with aftertreatment system OEMs and engine/truck OEMs for a coordinated possible research program investigating this topic.
- *Spark plug:* It is generally assumed that the heat grade of and materials used in spark plugs need careful consideration regarding risk of pre-ignition. With respect to possible (initial) spark plug gap and ignition energy reduction and their effect on spark plug lifetime when introducing hydrogen in the fuel mix of CNG vehicles, there is less consensus, requiring dedicated research.
- *Lubricating oil:* It is generally assumed that lubrication oil performances and lifetime can be adversely affected by hydrogen and water in the crankcase atmosphere from blow-by. This topic is addressed in publications, but not into high detail. With possible positions ranging between simply selecting dedicated lube oils, if available, or accepting reduced engine oil change intervals and/or reduced lifetime of critical engine components, it is advised to consult with lubricating oil suppliers on this topic.
- *Fuel line:* Materials and parts used fuel line system such as the pressure regulator may have to be upgraded to avoid hydrogen embrittlement when hydrogen is present in CNG depending on the materials. It is unknow what the maximum H_2 tolerance is for installed CNG components. One manufacture indicated that former components were compatible up to **10 vol.% hydrogen** in the blends while for the current components the maximum allowable fraction of hydrogen is 30 vol.% in CNG. Additionally, the fuel injector's performance will have to be revised to adapt to different sonic speed, density and energy density. The addition of hydrogen decreases the energy flow and as a result according to interviews above **15-20 H_2 vol.%** recalibration of engine mapping is needed.
- *Engine knock:* Hydrogen addition lowers the knock resistance and thus increases the risk of the occurrence of engine knock at constant spark timing and thus limits the amount of hydrogen that can be added. In worst case, when the methane number of CNG is similar to the minimum methane number given in the engine specification then no hydrogen is allowed.
  To what extend existing methods are capable of accurately predict the knock behavior of H_2/CNG blends in stoichiometric heavy-duty engines is to our knowledge not yet investigated.
- *CNG tank system:* Interviews revealed that the maximum hydrogen allowed in steel tanks (Type 1) is restricted to 2 vol.% according the regulation 110 of the Economic Commission for Europe of the United Nations and EN 16723-2:2017 [84]. For type 4 tanks it is not expected that replacement of CNG by H_2 does not

give any issues by the tank itself when maintaining the operating pressure (250 bar). However, it is unknown if the periphery such as sealing materials can safely handle the H_2/CNG blends.

Recommendations (see also Table 9)

- The literature review and interviews showed that t it is still unknown if the ancillary components of the tank system (seals, valves etc.) and fuel-line components can handle H_2/CNG blends. Therefore, to maximize the hydrogen percentage in CNG for heavy-duty applications it is necessary to investigate and subsequently develop standards on how much hydrogen these components can handle. An alternative is to replace these components with ones that are hydrogen compliant.
- The literature review and interviews revealed that to date only limited experimental data are present on the effect of hydrogen addition to CNG for heavy-duty stoichiometric vehicles. Since these engines are dominant in today's heavy-duty vehicle market, it is highly recommended to investigate the effect of hydrogen addition on the performance of stoichiometric heavy-duty CNG vehicles on the following topics and summarized in the Table below.
- The measured knock resistance and compare the results with the different methane number methods available.
- The potential fuel efficiency gain
- Fuel line components
- Fuel tanks, including tank periphery
- Lube oil degradation
- Combined catalyst and engine performance for resulting tailpipe emissions such as CH_4, NO_x, and NH_3/N_2O
- Spark plug design, selection and possible lifetime-extending operating conditions
- Crankcase safety
- Whenever CNG is stably delivered with a fixed amount of hydrogen, it is recommended to optimize the engine performance for that specific H_2/CNG fuel blend resulting in a higher fuel efficiency, lower emissions and stable combustion. In case of a variable hydrogen content in the H_2/CNG mixture, an engine mapping should be performed to cover the full range of H_2/CNG blends supplied at the fueling station. However, as an alternative a feed-forward gas-adaptive control system can be developed and deployed for optimum engine performance. It is recommended to assess the techno-economic feasibility of such fuel adaptive control systems together with OEMs.

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3. HYDROGEN SPECIFICATIONS IN EU STANDARDS

Hydrogen is not a component that is present in natural gas. For this reason, hydrogen is traditionally not present in natural gas pipeline specifications and CNG specifications. However, the drive to increase the share of renewable gases resulted in the initiation of several projects over the last decade with the aim to develop hydrogen specifications for natural gas. To gain insights in the current hydrogen percentage allowed within the EU both from a pipeline gas specification perspective and according to CNG standards we performed a literature review and interviews with the relevant stakeholders.

3.1. INVENTORY CURRENT STANDARD IN EU COUNTRIES

As shown in Table 10, the allowed hydrogen percentages in natural gas pipeline specifications differ from country to country. In Germany hydrogen addition to natural gas is allowed. However, the maximum limit for each area requires a case-by-case examination. For example, generally 0-2% hydrogen is allowed when CNG stations are connected to the grid, 0-5% H_2 is allowed when gas turbines are present in the grid, but no CNG filling stations are connected. When no gas turbines and/or CNG stations are connected generally 0-10% is allowed [1–5]. In the Netherlands 0.5 vol.% is allowed in the distribution grid and 0.02 vol.% in the high transmission grid [6]. In several countries no or very low hydrogen blending specification are defined. However, several countries work on the development of hydrogen blending specifications. For example, the project standard EN 16726:2015 proposes a maximal H_2-content depending on the application (0-10 vol% H_2) [38]. If this standard is accepted, it will replace the Synergrid recommendation G5/42 in Belgium. France allows 6 vol% percentage of hydrogen in natural gas [3, 7]. However, it is the responsibility of the network operator to check the compliance of the gas with the technical specifications, therefore hydrogen can be blended in most networks at a rate of 6% in terms of volume, only in the absence of sensitive structures or installations on the customer's premises.

The technical origins of the specifications defined in the different countries involve the effects of hydrogen on the transmission and distribution system, end-use equipment, safety and underground storage. However, the technical origins behind the specifications are not entirely transparent and as aforementioned many countries started research programs to define or redefine hydrogen blending specifications based on the outcome of research programs and pilot projects.

To gain insights in the most critical part in the value chain (pipeline, gas engine, compressor, CNG tank, boiler etc.) that determines the H_2 specification, an assessment of the range of acceptable levels of hydrogen blend for each key system component is performed.

Table 10 Overview hydrogen gas grid specification

Country	Max. Allowed H_2 (vol.%)	Sources
Germany	Case by case (0-10%)	[1-5]
Netherlands	0.5%	[1, 6]
Spain	5%	[1, 3]
Sweden	0.5%	[1, 8]
Belgium	0.1%	[2, 3, 9]
Denmark	not specified	[2, 10]
UK	0.1%	[2, 3]
France	6%	[3, 7, 11]
Austria	4%	[3, 12]
Italy	0.5%	[4, 7, 8]
Switzerland	2%	[3, 4]
Poland	not specified	[13]

3.2. BACKGROUND (FLEXIBLE) HYDROGEN ADDITION TO NATURAL GAS

Hydrogen can be injected into the high-pressure transmission pipeline natural gas networks and/or into the low-pressure natural gas distribution network that supply gas to the domestic-, commercial and industrial end-users or gas storage.

The pipeline systems, the gas storage facilities and the end user equipment present in the natural gas system are designed, installed and maintained for a given range of local distributed natural gases. The introduction of hydrogen to natural gas may have an impact on the physical and chemical properties of the gas delivered, which may affect the integrity of the pipeline system and gas storage, performance of end-user equipment and the accuracy of the metering equipment installed in the field.

In the field a large variety of components are used in natural gas transmission and distribution system. Furthermore, regarding the end-use equipment connected to the natural gas grid, a different sensitivity towards hydrogen addition to natural gas can be observed. Consequently, the allowable fraction of hydrogen gases in natural gas depends strongly upon both the type of appliances installed in the field and the natural gas composition distributed. Based on a literature review and (experimental) knowledge within DNV, a summary of the hydrogen range for each type of equipment is presented in the Table below. Here we emphasize always a range from 0-x mole % in the Table below and not a fixed hydrogen percentage value since the amount of hydrogen addition depends upon the natural gas quality to which hydrogen is added. For example, if the Wobbe index of the natural gas at the hydrogen blending facility is at lower end of the Wobbe index no hydrogen addition is allowed (0% H_2). Below the Table we briefly summarized the reasoning behind the given hydrogen range in natural gas given for the natural grid system and for the equipment connected to the grid in Table 11.

		0-0.5%	0-2%	0-5%	0-10%	0-15%	0-20%	0-25%	0-30%	0-40%	100%	Reference
Transmission	Steel pipelines	green	green	green	green	light green	light green	light green	light green	light green	light green	14,16
	Compression Centrifugal	green	green	light green	light green	orange	red	red	red	red	orange	14, 38
	Compressor piston	green	green	green	light green	orange	orange	orange	orange	orange	orange	14
Metering equipment & components	GC's and correlative gas quality sensors	green	orange	orange	orange	orange	orange	orange	orange	orange	orange	39
	Valves, turbine meters, filters etc.	green	green	green	green	orange	orange	orange	orange	orange	orange	14
Distribution	Steel pipes	green	green	green	green	green	green	green	green	orange	orange	14
	Fittings	green	green	green	green	orange	orange	orange	orange	orange	orange	14
	Plastic pipes	green	green	green	green	green	green	green	green	green	green	14, 15
	Inhouse infrastructure	green	green	green	green	green	green	green	green	orange	orange	14, 15
End use	Gas engines	green	green	green	light green	light green	orange	orange	red	red	orange	27, 28, 34
	Gas Turbines	green	green	light green	light green	light green	orange	orange	red	red	orange	30, 31
	Industrial burners (indirect heating)	green	green	green	green	green	green	light green	light green	orange	orange	21, 22
	Industrial burners (direct heating)	green	green	green	light green	light green	light green	light green	light green	orange	orange	11, 23-25
	Feedstock	light yellow	light yellow	light yellow	light yellow	light yellow	light yellow	light yellow	light yellow	light yellow	light yellow	14
	Domestic (premix) boilers	green	green	green	green	light green	light green	light green	light green	light green	orange	14, 32, 40
	Domestic hot water heaters	green	green	green	light green	orange	orange	red	red	red	orange	14, 40, 41
	Domestic Cookers	green	green	green	light green	orange	orange	red	red	red	orange	14, 41
Transport	CNG vehicles	green	green	orange	orange	orange	orange	red	red	red	orange	11, 34
	CNG fueling stations	green	green	white	white	white	white	white	white	white	white	11, 34
Gas storage	Salt caverns	green	green	green	green	green	green	green	green	green	green	14, 34
	Other under ground storage	green	green	green	orange	orange	orange	orange	orange	orange	orange	14

Status	Color code
Currently already possible	green
Adjustments may be required (case by case)	light green
Retrofit , replacement and/or additional research required	orange
Not possible	red
Depends on industrial proces (no generic statement possible)	light yellow
Unknown	white

Table 11 Estimation of hydrogen tolerance by volume percentage in the natural gas grid system[10].

3.2.1. High pressure natural gas transmission grid

The natural gas transmission grid contains several components such as steel pipelines, valves, metering equipment compressors etc. These components have different sensitivity towards the presence of hydrogen [14, 15]. Generally, these pipeline systems can handle 10 vol.% hydrogen. For higher percentages the maximum amount of hydrogen depends on its operation conditions, the pressure variation that occur in the network the type of materials present in the system, the conditions of the network [16], e.g. the presence of active crack like defects, magnitude, frequency of pressure variations, stress level and weld hardness etc.

3.2.2. Natural gas distribution grid

For distribution networks that only consists of plastic pipes 100% hydrogen can be accommodated [14, 15]. For the inhouse infrastructure about 30 vol.% hydrogen is allowed [14]. Here we remark that according to [14] fittings in the distribution grid can handle up to 10 vol.% hydrogen but it is unknown if all fittings are suited for higher fractions of hydrogen.

[10] Here we remark that we receive limited to no response to our questionnaires with regards to CNG fuelling stations operators and suppliers of components on the consequence of adding hydrogen to CNG.

3.2.3. Metering equipment & components

The gas grid consists of many components such as filters, different type of valves, odorant injection systems, different type of gas meters and gas quality analysers. Generally, these components have different sensitivities towards the presence of hydrogen in natural gas. For example, Gas Chromatographs (GC's) present in the field to measure the gas composition of the gas need to be recalibrated for hydrogen. Furthermore, the accuracy of gas flow meters (turbine meters, ultrasonic meters etc.) with increasing amounts of hydrogen in natural gas is currently studied in [17]. According to [14], except for GC's, metering equipment and components in the natural gas grid generally can handle up to 10 vol.% hydrogen and more research is needed to maximize the hydrogen content for specific components such as gas meters.

3.2.4. CNG fuelling stations

A CNG fuelling station receives the gas from the local pipeline that is initially at low pressure. The gas is compressed on site and is moved to a buffer storage (storage tank), from which the gas is transferred to the vehicle tank. According to the UNECE regulation 110, the hydrogen limit for CNG vehicles is max. 2% which puts restrictions on the amount of hydrogen in the gas delivered by the CNG fuelling station. According [18], “permission for H_2 injection is typically considered on a case by case basis and operated on a time limited demonstration basis only or by exception”.

There are generally two options available for the addition of hydrogen to CNG: addition of hydrogen to the natural gas pipeline and hydrogen addition on-site.

Literature review reveals that most projects focus on mixing the hydrogen onsite with the CNG. For example, in Malmo (Sweden) from 2003-2005 two buses (lean-burn engines, [19]) were operated on mixtures of hydrogen and CNG. The hydrogen was produced by electrolysis with electricity from a nearby wind power plant. The hydrogen was stored as a compressed gas and mixed with the CNG at the dispenser. Two mixtures were studied: 8 vol.% hydrogen in CNG and 20 vol.% hydrogen in CNG (both at 200 bars at the dispenser). Here we note that no modifications for the buses were required for the 8 vol.% hydrogen in CNG, because this mixture was considered as CNG according to the natural gas specifications. However, for the 20 vol.% hydrogen in CNG a new engine mapping was required [19]. This project revealed very few problems, except for the blending of the hydrogen and the CNG; it was found that after control measurements the blends varied a few percentages [20].

For the addition of hydrogen to the local pipeline gas, the components of the CNG fuelling station must be checked on the hydrogen compatibility [19]. These components include compressors (see below), storage tanks (see Chapter 3) and the fuel line. Here we remark that the compression power increase with higher hydrogen content in the gas (see below).

3.2.5. End use equipment: sensitivity to hydrogen

The distributed natural gas-hydrogen mixtures should be within the legal limits of the pipeline specification. For gas engines, gas turbines and gas compressors the fuel specifications given for specific engine/compressor types form the basis for manufacturers' guarantees for machine performance and maintenance intervals. In case the recommended fuel specifications are not met it can result in nullification of the guarantee. For this reason, it is highly recommended to communicate with the equipment owners when mixing hydrogen fractions that exceed the fuel specifications.

3.2.5.1. Industrial burners

In industrial processes several burner types are used in heating processes. Industrial heating processes can roughly be divided into indirect- and direct heating. Indirect heating is when the substance to be heated, referred to as the load, is separated from the combustion products, for example by heating tubes through which for example water flows for hot water and steam production. In contrast, direct heating, often associated with high process temperature, exposes the product directly to the flame and/or combustion products in an industrial kiln or furnace.

Most of the industrial burners are so-called diffusion type burners used in both direct and indirect heating processes. Experimental results show that up to 30 vol.% hydrogen in the fuel no substantial deterioration in the burners' performance occurs for the burners studied [21–25]. However, it is known that hydrogen addition can result in increased NOx emission above the legal limits. As NO_x mitigating measure flue gas recirculation can be successfully applied [22, 23]. For direct heating processes hydrogen addition can influence the product quality as a result of for example changes in flue gas composition, flame length and radiative heat transfer. For direct heating processes generally 10 vol.% H_2 is allowed. In the Joint Industry Project 'Hydrogen as a fuel for heating processes' [26] the impact of hydrogen for direct heating processes will be studied in more detail and more research is needed to maximize the allowed hydrogen limits in natural gas.

3.2.5.2. Stationary gas engines

Generally, there are two common types of lambda control used in gas engines: 1) lambda-sensor-based AFR (Air Fuel Ratio) controller, and 2) speed-density AFR control. The first type (lambda control) is dominant in older-generation gas engines. Modern gas engines are usually equipped with (a variant of) a speed-density AFR control. In response to hydrogen mixture, the AFR control based on a lambda sensor was found to maintain a virtually constant air-fuel ratio. Measurements performed in a gas engine with lambda control show that the NO_x emission increases substantially upon hydrogen addition while the engine with speed density control did not show a substantial increase in the NO_x emission [35].

Several gas engine studies summarized in [27, 28] confirm that hydrogen mixture leads to a lower knock resistance of the fuel gas. The knock resistance of a fuel gas is characterized by a methane number, which is like the octane number used to qualify gasoline. All OEM's prescribe the minimum methane number allowed in their gas specification. Consultation of gas engine manufacturers shows that there is not yet much experience in the market with natural gas-hydrogen mixtures. Based on the currently known fuel gas specifications, engine manufacturers generally allow a maximum of between 0 and 5 vol.% hydrogen in the fuel gas for the gas engines installed in the field. Additionally, several OEMs indicated that their gas engines optimized for high efficiency are suited for hydrogen contents of around 10 – 15 mole% without derating. However, they note that the amount of hydrogen mixture strongly depends on the current engine setting and on the (lowest) methane number of the natural gas to which hydrogen is added as shown in the case study.

3.2.5.3. CNG vehicles

Engine components such as fuel rails, fuel injectors and fuel tanks are homologated according to the 110R regulation [29]. Among other things, the 110R regulation relates to the approval of specific components of motor vehicles using compressed natural gas (CNG) and/or liquefied natural gas (LNG) in their propulsion system. According to this regulation a maximum hydrogen volume percentage of 2 vol.% is allowed, because of the limit of the type 1 tank. This currently sets the maximum hydrogen fraction in CNG vehicles. Several respondents indicated that not much experience is available regarding engine performance, fuel line components and tank components for hydrogen percentages above 2 vol.%.

An interview with one of the major heavy-duty engine manufacturers indicate that based on their experience no re-calibration of the ECU was needed up to a maximum hydrogen volume percentage of 20% hydrogen is necessary. Above 20 vol.% a different engine mapping is required and for percentages above 40 vol.% no experience is present. The OEMs remark that hydrogen addition to natural gas only is allowed when the methane number is not outside the engine specifications. Furthermore, it is unknown if and by how much the CNG components (fuel line, injectors, pressure regulators, fuel valve etc.) can handle more than 2 vol% hydrogen.

3.2.5.4. Gas turbines

Gas turbines can be roughly divided into two types of combustors premixed and diffusion combustion systems. Hydrogen addition to natural gas is the most challenging in premixed operation due to its high flame speed and high temperature which can cause flashback, increasing NO_x emissions and overheating of the turbine parts such as the blades. Given the above, many turbine manufacturers have stringent specifications for the allowable fraction of hydrogen in machines that were not intended to burn hydrogen-containing gases. The amount of allowable hydrogen depends upon the combustion system used for each turbine, which can lead from 0% tolerance up to 5 mol% for the majority of the current installed base [30, 31]. Typical issues are flashback, acoustic instability and shortening lifetime (high firing temperature and moisture content), increased NO_x emission. Modern turbines often can handle up to 10 mole% hydrogen in the fuel [30]. To further increase the amount hydrogen in natural gas turbines needs to be retrofitted by for example changing the fuel gas supply system, replacing seals, valves, changing the sizing of the fuel supply lines and other structural components and for concentration up to 30 mol% hydrogen often the burners needs to be replaced to prevent flash back and increased flame temperatures (NO_x and overheating turbine parts).

3.2.5.5. Domestic equipment

Domestic equipment can be roughly divided into two types of lean premixed (excess of air) and fuel rich appliances (excess of fuel). The lean premixed appliances consist of mainly domestic boilers (central heating boilers) and the fuel rich appliances consists mainly of hot water heaters (geysers) and cookers. Both categories have a different tolerance to hydrogen addition. In a study performed by DNV for Gasunie we investigated in detail the maximum allowable hydrogen fraction based on gas interchangeability analyses and measurements on these type of appliances [32]. Gas interchangeability analyses and measurements show that for fully premixed boilers up to 30-40 mole% hydrogen is allowed in natural gas. Measurements on partially premixed appliances (cookers and hot water heaters) show an increase in the burner deck temperature with increasing fraction hydrogen in natural gas. To what extent this increase in temperature will affect the long-term performance of these partially premixed appliances and will be investigated in the Hydeploy project [33] wherein the long term effect will be investigated for hydrogen blends up to 20 mole% in natural gas. As a very conservative estimation based on gas interchangeability analyses show for the Dutch natural gas grid that up to max. 6 mole% hydrogen is allowed. A recent study by Marco gas [1] show that 10 vol.% is allowed. For appliances with air-fuel ratio control (e.g. SCOT), the heat transferred increases progressively with hydrogen addition, increasing the risk of overheating/flashback; tests performed at DNV show flame flashback when 20 vol.% hydrogen is present in natural gas. More research on the effect of hydrogen addition for these types of appliances is strongly recommended. We expect that in the future boilers will be developed that can cope with 0-100% hydrogen in natural gas. Therefore, in the retrofitted case in the data base we use 0-100% hydrogen where we assume that the restriction of the Wobbe index is resolved by widening the Wobbe distribution band in the future to allow flexible hydrogen addition to natural gas.

3.2.6. Compressors

Gas compressors are an essential part of the natural gas transport network. The main categories used in natural gas networks are centrifugal compressors and piston compressors. These two types of compressors respond differently to the addition of hydrogen to natural gas. Below an overview is given for the technical suitability for compressors regarding the amount of hydrogen that can be added into natural gas. Apart from the technical suitability, an important issue is the safe and trouble-free operation. One of the most important issues with pressurized equipment is the low density of hydrogen (0.09 kg/m^3 at STP) and hydrogen containing gases as compared to natural gas (0.7-0.9 kg/m^3 at STP) and, subsequently the leakages. Feedback from the questionnaire reveal that gas leakage in pressurized equipment is regarded as a serious issue (high risk of self-ignition of the leaking hydrogen). One of the respondents to the questionnaire stated that it is important to involve Third Party assessment (components, devices and so on) from the early beginning of the design, production, commissioning, operation and maintenance to avoid any negative issues and incidents during the operation of the compressor with hydrogen containing gas mixtures. Here we remark that the compressor power should be increased for higher hydrogen content in the gas. For example, when 30% hydrogen is present in natural gas the power should be increased by approximately 40% assuming the same amount of energy transported as compared to transporting neat natural gas.

3.2.6.1. Centrifugal compressors

Centrifugal type compressors are very susceptible to changes in the gas density. When hydrogen is added to natural gas the density (molar weight) of the gas distributed lowers which results in a decrease in the momentum (mass x velocity) in the centrifugal compressor at further identical conditions. To maintain the required compressor head (or discharge pressure) the flow will be reduced which can result in occurrence of surge. Surging occurs when insufficient gas flow enters the compressor and/or when the increase in pressure rise across the compressor is insufficient. A rule of thumb is that centrifugal compressors can handle a maximum fluctuation in the molar weight of maximum 5 % to maintain the gas network requirements (discharge pressure and flow). By using this rule of thumb, a maximum amount of about 5 mol% hydrogen is allowed. If the grid operator accepts a reduction in the amount of energy distributed the maximum hydrogen percentage will be limited by the surge line. Based on calculations performed within DNV it was indicated that the maximum amount is approximately 10 mol% hydrogen in natural gas before surge occurs at a fixed compressor speed.

3.2.6.2. Piston compressors

Generally, the amount of energy that a piston compressor can deliver reduces substantially when hydrogen is added to natural gas. If the TSO does not want to compromise on the maximum amount of energy that the gas grid can deliver only limited amounts of hydrogen is allowed depending on the gas composition, allowable variation in piston speed and allowable temperature/pressure increase. Often the compressors are overdesigned by approximately 5% in compressor capacity. By using this margin, approximately 5 mole% hydrogen addition to natural gas is allowed. One of the manufacturers stated that they setup a maximum of 10% hydrogen in natural gas without having to take special measures to the compressor system.

The increase of the hydrogen concentration in natural gas should be discussed with the compressor manufacturer since the normal operating point of the compressor will change when adding hydrogen to natural gas.

3.2.7. Underground storage

According to [34] no problems were identified with salt cavern storage, and therefore no problems are to be expected for storage of hydrogen and natural gas mixtures. Generally, more R&D is recommended to understand the effect on underground gas storage when having 5 to 10 vol. % hydrogen in natural gas [17].

3.3. GAS BLENDING INJECTION STRATEGIES

Injecting hydrogen together into the existing gas grid is an effective means to avoid large investments in the infrastructure while ensuring the wide-spread use of these fuels by industrial, commercial and residential end users. However, the equipment connected to the natural gas grid is designed, tested, installed and maintained for a given range of local distributed natural gas. As discussed, the injection of hydrogen to the natural gas grid may have an impact on the physical and chemical combustion properties of the gas delivered, which may affect the performance of these end user equipment out of the range where they are designed for resulting in unwanted issues such as engine knock or flash-back in burners. Large-scale replacement of end-use equipment takes time, and a strategy is needed to maximize the incorporation of hydrogen into the natural gas grid.

The strategic approach proposed in this study is 1) to determine the combustion properties (gas quality) and the flow of natural gas at the point of blending and 2) to perform a renewable gas blending assessment on the types of equipment installed in the area in which the gas is being supplied to. The assessment comprises the analyses of the performance and safety of end-use equipment when hydrogen is added to natural gas as described in [36]. When strategically selecting the geographical location for hydrogen injection, the maximum percentage hydrogen allowed by the appliances installed in the field in combination with the flow of natural gas at the point of hydrogen injection must be determined. Furthermore, to increase the allowed fraction hydrogen countermeasures can be taken by for example replacing or adjusting the most critical appliance types in the field in an economically feasible way by performing cost/benefits analyses.

From Table 11 it can be seen that when ranking the categories according to sensitivity to hydrogen in the natural gas grid, heavy-duty CNG vehicles, CNG fuelling stations, gas turbines, some feedstock processes and centrifugal compressors are most sensitive to the presence of hydrogen. If this equipment is present only maximum 2 vol.% hydrogen is allowed in natural gas. To maximize the hydrogen injection, one should find a different injection location with more hydrogen-robust equipment (short term strategy) or to apply mitigating measures (long term strategy). For example, for CNG vehicles this means that the CNG steel tanks (Type 1) should be replaced, the tank periphery components should be tested or replaced by hydrogen resistant components and if needed re-calibration of the engine mapping should be done. When hydrogen is blended in the gas grid one can expect fluctuating hydrogen percentages in CNG. The disadvantage of fluctuating gas compositions is that the engine should handle the entire range of hydrogen that is allowed in the grid which can result in conservative engine parameter settings. A potential solution to allow optimal engine over the entire range of CNG/H_2 gas qualities is applying feed forward engine control as described in Chapter 3.7. Other mitigating measures for other end-use equipment is described in Chapter 4.2.

For heavy-duty CNG vehicles an alternative solution to maximize hydrogen blending is to blend hydrogen at the fuel station using a fixed percentage hydrogen in CNG. When supplying CNG/H_2 with a fixed hydrogen percentage, engine designers can make optimal use of the combustion properties of CNG/H_2 such as a higher fuel efficiency and lower pollutant formation as compared to CNG engines. This strategy would be interesting for, for example 24/7 inner-city distribution and city buses. This hydrogen blending strategy needs a separated hydrogen supply chain, an onsite storage and mixing facility at the fuelling station and the development of and dedicated CNG/H_2 engines. An additional advantage is that this option has been investigated and successfully tested (see e.g. [37]).

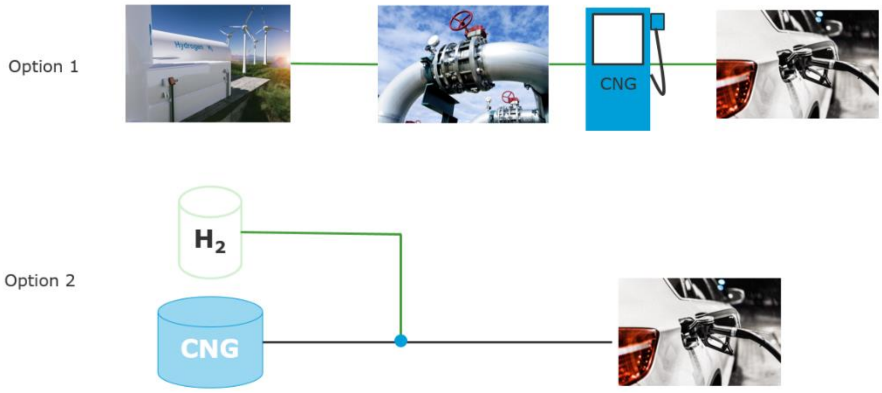

Figure 29 Different hydrogen blending strategies: 1) injection hydrogen into the gas grid and 2) blending hydrogen at the fuel station using a fixed amount of hydrogen

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4. METHODS FOR DETERMINING HYDROGEN CONCENTRATION IN NATURAL GAS

Natural gas quality analyzers are present in the natural gas grid to determine the gas quality for billing purposes and to ensure that the gas quality delivered to the end users is within the natural gas specification. To fulfill the required (high) accuracy of the measured natural gas composition often a gas chromatograph (GC) is used in natural gas grids.

Gas quality sensors can also be used in gas adaptive feed forward engine control systems. Such systems allow real-time adjustment of the engine settings based on the measured composition of the fuel that enters the engine [1, 2]. The advantage of such a feed-forward fuel-adaptive engine control system [1, 2] is that the engine only will be adjusted from its optimal setting (maximum power and efficiency) when the methane number or NOx emission is lower than specified.

The gas quality sensors available on the market can be subdivided into 'direct' and into correlative (indirect) sensors. The direct methods such as infrared sensors that convert the infrared absorption spectra of the individual components to a concentration are not able to measure hydrogen since hydrogen does not absorb in the infrared region. For these types of sensors, it is recommended to include a dedicated hydrogen sensor to measure the amount of hydrogen in natural gas blends. For correlative sensors used to measure the gas quality of natural gas it is unknown how these sensors will respond to the presence of hydrogen. Correlative sensors derive the gas quality indirectly by using the physical properties of the gas (e.g. density, calorific value, thermal conductivity, speed of sound, and so on).

Gas sensors for billing purposes

As described above often Gas Chromatographs are used to determine the gas quality of natural gas. The molecules present in natural gas will be separated when they go through the analytical column and then pass over the detector. Several types of detectors are available for gas chromatographs, including flame ionization detector (FID), flame photometric detector (FPD) and most common detector used for most gas measurements is the thermal conductivity detector (TCD). The TCD is based on measuring the differences in thermal conductivity. Since often helium is used as carrier gas which has a similar thermal conductivity as hydrogen11 this type of GC's should be replaced or upgraded when hydrogen is present in natural gas. Even when using GC's with other type of detectors, adjustments are needed to be able to detect hydrogen. Moreover, to guarantee the accuracy, new calibration gases that include hydrogen should be used. It should be noted that current standards such as ISO 6974 series [4] at present are validated for low levels of H2 (0.5 vol% in the ISO 6974). In [3] it was recommended to develop adaption of the standard.

Real time monitoring gas quality sensors

Besides GC's other novel gas quality sensors are in development and commercially available that approaches the accuracy required for billing purposes. An overview of a number of these sensors available are presented in Table 12. Several manufacturers of correlative sensors that use for example the heat conductivity and/or the viscosity, speed of sound etc. have indicated that an instrument is

[11] The conductivity of helium = 151 W/m*K; hydrogen = 180 W/m*K.

optimized and calibrated for a specific concentration range of natural gas/CNG. Deviation from this specified range of gas composition or the introduction of new components could affect the accuracy of results. To illustrate this the reliability of a commonly used correlative natural gas quality sensor has been tested within a range of 0-30% hydrogen by comparing with results from a in parallel installed for hydrogen calibrated GC. From the measurements presented in Figure 30 it can be seen that (as expected) the caloric value measured with the GC decreases as a result of the increase in hydrogen percentage in methane. However, the measurements with the correlative sensor show a substantial gradual increase in the caloric value with increasing hydrogen content in methane. The origin of the large deviation is the very large thermal conductivity of hydrogen in comparison to methane gas causing large errors in the relations used to derive the composition and caloric value. As a result, the increase in thermal conductivity measured by the correlative sensor is assigned to propane which results in an increase in the calorific value while in reality the calorific value decreases with increasing hydrogen content for the studied mixtures (0-30% H_2 in CH_4). From the results obtained above we conclude that caution is advised when using correlative quality sensors calibrated for natural gas in combination with ‘new’ gases such as biogas and hydrogen blends in natural gas.

Several manufacturers of correlative sensors (Table 12) indicate that they offer (chip based) micro-thermal conductivity sensors covering the entire range 0-100% hydrogen in CNG. Generally, the accuracy of these sensors is in the sub-percentage range with response time of only a few seconds. These sensors can be produced at potentially low costs at high numbers due to the fully integrated chip character of the sensing principle.

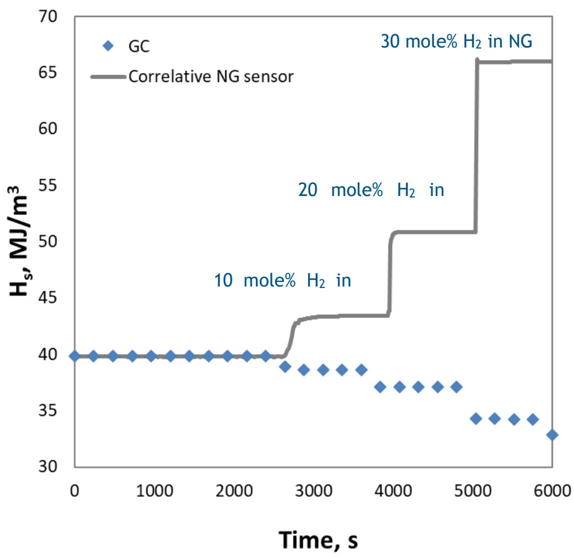

Figure 30 Measured Calorific value in methane/hydrogen using the GC (blue dots) and a correlative natural gas sensor

As indicated above, sensors that measure the gas composition based on infrared absorption are not able to detect hydrogen. A solution is to combine these infra-red based sensors with a separate sensor that measures the hydrogen fraction. The hydrogen percentage presented in Figure 31 are measured with a sensor that contains a thin film palladium-nickel alloy-based lattice that absorbs and desorbs hydrogen as it comes in contact with the sensor. The palladium catalysis converts the molecular hydrogen into atomic hydrogen, which gets absorbed into the metal lattice and changes the bulk resistivity. This change in resistance is reported in real time as the partial pressure of the hydrogen in the gas. Figure 31 shows excellent agreement between the measured hydrogen percentage in natural gas using this fast response H_2 sensor and a GC. Alternatively, Raman spectroscopy detection techniques are available that can measure the hydrogen percentage in natural gas (0-100% H_2).

Analyzers that use catalytic combustion to determine the Wobbe index and the gross calorific value with a response time of <5s can often cope the presence of hydrogen in natural gas. However, these sensors cannot measure the amount of hydrogen in natural gas. Optionally, an additional sensor can be integrated into the analyzer to determine the hydrogen percentage in natural gas

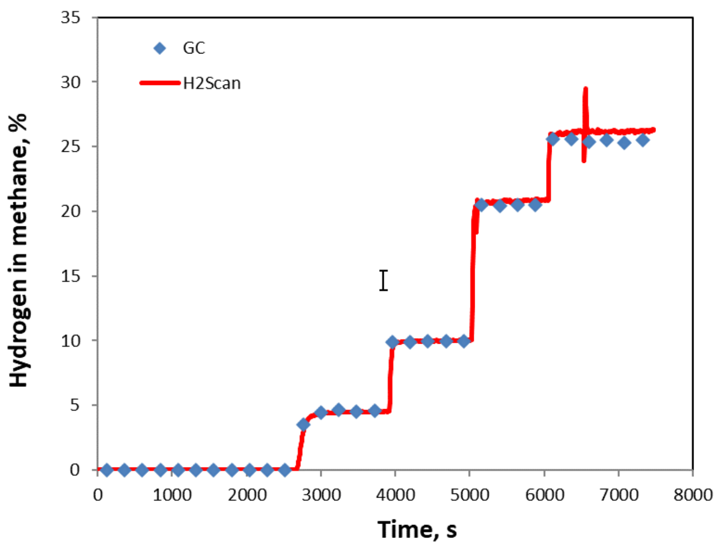

Figure 31 Measured hydrogen percentage in methane using the GC (blue dots) and a fast response hydrogen sensor (red line)

As discussed above, the real time gas quality sensors can also be used as gas quality sensor in feed-forward fuel adaptive engine control systems [1]. These gas quality sensors are currently not used in heavy-duty vehicle control systems. However, in combination with the sensors that are already present in the vehicle (lambda sensor, knock detection system, and so on) the feed forward control system can offer great benefits when variable hydrogen percentages are present in the CNG.

The main difference with regard to the requirements of these gas quality sensors is that for heavy-duty vehicles the cost price should be low (order of several hundred euro's), the sensor should be robust and compact. However, the accuracy requirements are allowed to be lower (order of percentages) than those used for billing purposes.

In Table 12 an overview is given for several quality sensors that are commercially available on the market. This overview is not comprehensive but gives a good impression of the type- and specifications of the sensors available on the market.

Table 12 Summary table sensors overview (not comprehensive)

Analyser	Principle	Component	Physical parameters	H_2 compatible	Response time	Robust/ simple to operate	Accuracy according to manufacturers	Costs kEuro
GasPT/Orbital	correlative	C_1-C_3, N_2, CO_2	Wobbe, MN	unknown	5s	Yes	0.4%	+++
EMC 500	correlative	CO_2	Wobbe, Hs, density	unknown	30s	-	<0.5%	+++
Precisive	Direct optical (IR)	C_1-C_6	composition (Wobbe, Hs, density, MN)	Yes, potentially in combination with a H_2 sensor	<5s	Yes	<0.1%	+++
MEMS	correlative	-	Wobbe, Hs, density, MN	Yes (0-100 vol%)**	Few seconds	Yes	<±1%	++(+)
Elster Instromet Gas-lab Q1	Direct & correlative	C_1-C_8, CO_2, N_2	Wobbe, Hs, density, MN	Unknown	<10-60s	-	<0.4%	+++
Micro GCs	Direct GC	C_1-C_8, N_2, H_2	Wobbe, Hs, density, MN	Yes, but GC should be calibrated for H_2 in CNG	Several minutes	No	<0. 2*	+++(+)
WIM Compas* (Hobre)	Catalytic combustion	-	Hs, Wobbe	Yes (max. 95 vol% H_2)**	<5s	Yes	0.5-1%	++++
HiGas*	Catalytic combustion	-	Hs	Yes (max. 95 vol% H_2)	<15s	Yes		
Hobre Raman	Raman spectroscopy	C_1-C_xH_y, H_2	composition (Wobbe, Hs, density, MN)	Yes (0-100% H_2)	<10s	Yes	0.5-1%	++++
H2Scan	Correlative	H_2	H_2	Yes, suited for measuring H_2 (%) in CNG	<90s	Yes	<0.3%	+++
Bright sensor	Correlative	H_s, MN	Wobbe index	Unknown	<10s	Unknown	Unknown	+
North Dome (AviSense)	Correlative	Density or molar mass	Density or molar mass	0-100 vol% H_2**	<5s	Yes	±0.02kg/m3	++(+)

* Optional a GC or conductivity sensor can be integrated into the analyzer to determine the hydrogen percentage in natural gas

** According to manufacturer (in response to the questionnaire)

+ 0-200 Euro, ++ 200-2.000 Euro, +++2.000-20.000 Euro, ++++ >20.000 Euro

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5. COMPARISON BETWEEN ICE AND FUEL CELL POWERED HEAVY-DUTY VEHICLES

5.1. FUEL CELL POWERED HEAVY-DUTY VEHICLES

5.1.1. What is a Fuel Cell?

The basic fuel cell is a device, where the oxygen and hydrogen gas react in a controlled way. Hydrogen and oxygen flow inside the cell and react to produce water and electricity.

$$2\,H_2 + O_2 \leftrightarrow 2H_2O$$

The fuel cell transforms chemical energy into electrical energy without the intermediate indirect route via thermal and mechanical energy as is the case in combustion engines. The different fuel cell technologies differ by the working temperature, the working pressure, the sensitivity of the fuel and the behavior in partial load ranges [1]. This section will give a closer look to the fuel cell technologies polymer electrolyte membrane (PEMFC) and solid oxide (SOFC) and the behavior with the different fueling gasses.

5.1.2. Functionality of a polymer electrolyte membrane fuel cell

A basic structure of a PEMFC is shown in Figure 32. The hydrogen flows inside the fuel cell on the anode side, and the oxygen on the cathode side (see also reactions above). Both electrodes are made of porous carbon and separated by a polymer electrolyte membrane. This membrane lets only pass positive hydrogen ions through. It is impossible that an uncontrolled reaction starts. The electrons will go through the external circuit and the water is produced at the cathode. State of the art in such systems is the use of platina catalysts, which is used at both the cathode and anode to accelerate the reaction.

The membrane in a low-temperature PEMFC (LT-PEMFC) must be hydrated with liquid water to ensure the conductivity. That is why the operating temperature of the LT-PEMFC is below 90 °C. A high temperature PEMFC (HT-PEMFC) consists of a silicon carbide matrix saturated with liquid phosphoric acid. The high operating temperature of 140-200°C reduces the amount of platina and increases the tolerance to CO poisoning. In general, PEMFC have the highest power density of all the fuel cell types (see also Table 13), which makes it interesting for mobile applications. The lifetime can be up to 30.000-40.000 hours under optimal conditions (temperature, constant load and optimal humidity) [1].

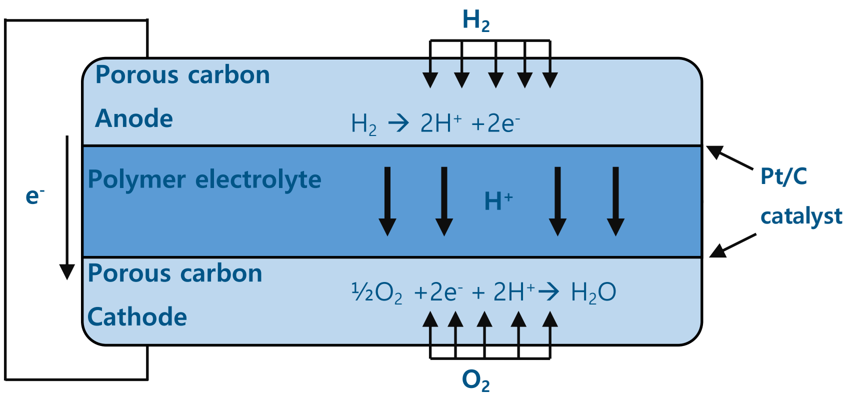

Figure 32 Schematic of H2-O2 PEMFC [1]

5.1.3. Functionality of a Solid Oxide Fuel Cell

A basic structure of a SOFC is shown in Figure 33. The SOFC is mainly developed for stationary power applications. Similar to PEMFC, the hydrogen flows inside the fuel cell on the anode side and the oxygen on the cathode side. The anode is made of porous nickel and the cathode of porous conducting ceramic. Both electrodes are separated by a solid ceramic electrolyte. Only oxygen ions can diffuse through this membrane. Here too, it is impossible that an uncontrolled reaction starts. The electrons will go through the external circuit. The working temperature of the SOFC is between 600 °C and 1000 °C (At these temperatures the oxygen ion can diffuse through the ceramics). The challenges of such a high temperature are the high requirements for material, mechanical issues and thermal expansion. The high temperature on the other side has also advantages like a fuel flexibility (internal reforming of hydrocarbons), a high efficiency and the possibility to use the waste heat in a combined heat and power application. The lifetime of this systems can be up to 100.000 hours, but only a limited number of ramps up processes is allowed within the expected lifetime (to minimize thermal stress) [1].

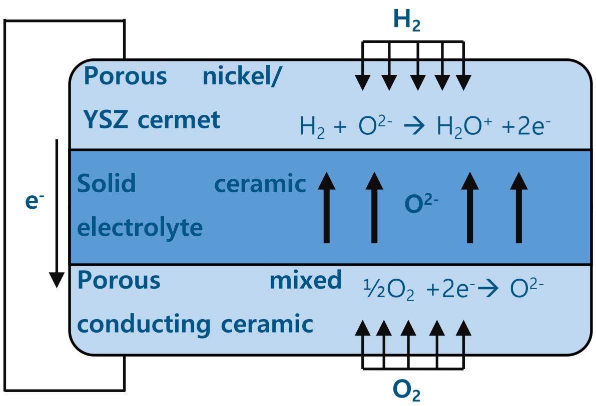

Figure 33 Schematic of H_2-O_2 SOFC [1]

5.1.4. System overview

To use a fuel cell with fuels other than hydrogen, a gas reforming process is necessary. There are basically two types of reformers, which can be combined;

- Steam reforming process (water and natural gas transformed to a hydrogen rich gas: $CH_4 + H_2O \rightarrow CO + 3H_2$).
- Dry reforming or so-called CO_2 reforming ($CH_4 + CO_2 \rightarrow 2CO + 2H_2$).

A HT-PEMFC additionally needs a water gas shift reaction to create CO_2 and H_2 out of CO + H_2O. After this step, there is still carbon monoxide in the fuel which can be poisonous for both HT- and LT-PEMFC (see also Table 13). To a certain CO level, HT PEMFC can work with it, but for the LT PEMFC an extra preparation of the fuel necessary [2].

Figure 34 gives an overview over the different auxiliary units are needed for PEMFC and SOFC to use natural gas as fuel. PEMFC is sensitive to impurities in the hydrogen fuel and requires a reformer that can meet these specifications. For the SOFC a reformer is not always necessary, light hydrocarbons (e.g. methane, ethane, propane and butane) can be internally reformed due to the high operating temperature, but a feed containing heavy hydrocarbons (e.g. gasoline and diesel) require an external reformer [3].

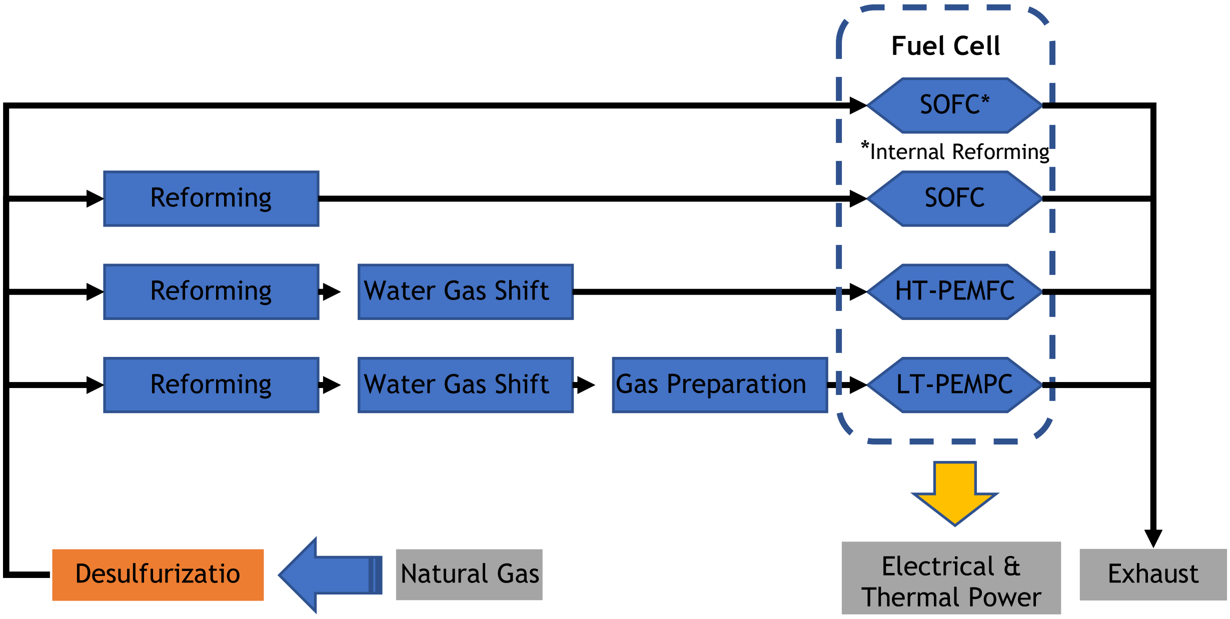

Figure 34 Overview over different fuel cell systems fueling with natural gas [4].

5.1.5. Fuel cells in heavy-duty applications

Today there are two popular options to decarbonize the heavy-duty sector. On the one hand there is the possibility to use battery-powered electric vehicles and on the other hand the hybrid electric vehicles, i.e. fuel cell with electric motor. The hybrid vehicles were developed to get over the disadvantages of battery powered electric (long fueling time and limited range) and the vehicles with a conventional internal combustion engine (GHG emission and efficiency). One possible hybrid is the combination of a fuel cell and a battery storage. In these hybrids PEMFC are state of the art because of the fast start/response time and the low operation temperature. Nevertheless, there are disadvantages concerning the fluid management, the fuel requirements, and the thermal management. That's why SOFC comes in the focus of interest [5].

Table 13 Comparison between PEMFC and SOFC

	PEMFC	*SOFC*
Fuel	H_2 , methanol [1]	H_2, CH_4, CO [1]
Operating Temperature	80 °C [1]	600 - 1000 °C [1]
Catalyst	Platinum [1]	Perovskites [1]
Electrical Efficiency	40 - 60 % [5]	55 - 65 % [6]
Start-up time	Fast [7]	Slow [7]
Application focus	Vehicles	Stationary engines
Power density (W/cm2)[8]	0.7+	0.15-0.7
Typical stack size [8]	1-100 KW	2 KW up to 100 MW range
Substances poisonous to fuel cell[9]	Sulphur CO (> 10ppm, LT-PEMFC, >3% HT-PEMFC)	Sulphur
Challenges [10]	Expensive catalyst (Pt) Sensitive to fuel impurities	Long start-up time High temperature corrosion and breakdown of cell components

Table 14 displays an overview over the most promising fuel cell trucks so far. There is still no large-scale production, but the number of manufactures rises. Compared to battery electric vehicles the fueling/charging time is lower (around 15 to 20 minutes compared to several hours).

As mentioned, fuel cell vehicles are always hybrid vehicles. Since a fuel cell can just provide energy (unidirectional) a short time storage (e.g. battery or super capacitor) is needed to store the recuperation energy (break energy). Also, the battery allows that the fuel cell runs at a constant operating point with a high efficiency and avoids high power gradients, which would lead to a faster degradation. The manufacturers interviewed see PEMFC as the most promising option for road vehicles, as these have a lower operating temperature and a shorter start-up time. If the fueling stations provide pure hydrogen (99.999 %), the main advantage of the SOFC, the high fuel tolerance plays a minor role.

Table 14 Overview Fuel Cell Trucks and Buses (Announced Data)

Manufacture	*Hyundai*	*Nikola*	*HYZON*	*Toyota*	Solaris
Type	Fuel cell electric truck	Hydrogen Electric		Beta	Urbino 12 hydrogen
Power	190 kW	735 kW	500 kW	226 kW (FC)	70 kW (FC)
Range	<400 km	<1200 km	<600 km	< 500 km	350 km
Weight	40000 kg	40000 kg		40000 kg	
Status	2020	Production later than 2021	2021	Proof of concept 2018-2019	Commercial
FC Type	PEM	PEM	PEM	PEM	PEM
Source	https://www.hyundai.com/worldwide/en/company/newsroom/hyundai-motor%E2%80%99s-delivery-of-xcient-fuel-cell-trucks-in-europe-heralds-its-commercial-truck-expansion-to-global-markets-0000016544	https://www.ttnews.com/articles/nikola-sets-truck-production-schedule-envisions-autonomous-model	https://fuelcellsworks.com/news/hyzon-motors-inc-opens-europes-first-dedicated-hydrogen-truck-production-facility/	https://www.expressandstar.com/news/motors/2020/12/14/toyota-begins-testing-second-generation-hydrogen-power-for-trucks/	https://www.solarisbus.com/en/vehicles/zero-emissions/hydrogen

5.2. HYDROGEN ICE POWERED HEAVY-DUTY VEHICLES

Hydrogen internal combustion engine (ICE) can be divided into two main fuel injection strategies: port fuel injection and direct injection [11, 12].

In port fuel injection, hydrogen is injected before and/or during the intake stroke (see Figure 5) upstream of the intake valve where it mixes with air. Benefits of this fuel injection strategy is its relative simplicity and the availability of these injections systems. This fuel injection strategies have already successfully being applied in some H_2-ICE vehicles [13–15]. However, an engine operating on hydrogen using the port fuel injection suffers from several drawbacks. An important drawback is the occurrence of backfire (flashback) [11, 12, 16, 17] in the intake manifold. This is caused by the short quenching distance together with the high burning velocity of hydrogen as compared to hydrocarbons (see also Table 3).

The development of direct injection systems for hydrogen engines was found to solve the increased risk of possible explosions in the intake manifold, crankcase of exhaust manifold [11, 17, 18]. According to [11], several studies have demonstrated that “high load hydrogen high pressure direct injection (HPDI) operation under optimal conditions, can achieve similar efficiency as traditional diesel engines (~ 40-45%).

Like the H_2/CNG internal combustion engines described above, the H_2-ICE also suffers from high NO_x emissions due to the high adiabatic flame temperature of hydrogen. Fuel lean operation, high EGR rates and multi-injection strategies [17] were effectively used to reduce the NO_x emission.

Between 2000 and 2010 several concept cars were developed by OEMS, such as the BWM Hydrogen 7 and Ford P2000 [11, 12]. However, according to Ref [11] the development of H_2-ICE powered vehicles is stopped in favour of developing FCEV powered vehicles and the research focus is on addition of hydrogen to hydrocarbon fuels (e.g. gasoline, diesel and CNG).

As shown in the Table below, using liquid hydrogen substantially increases the range as compared to using compressed hydrogen (248 bar). Here we remark that modern systems can increase the pressure up to 750 bar which increased the vehicle range.

Table 15 BMW Hydrogen 7 and Ford P2000 vehicle specifications

	BWM Hydrogen 7 [12, 14]	Ford P2000 [12, 15]
Introduction	2003	2001
Vehicle type	Demonstration vehicle	Sedan
Engine	6.0 Litre V12	2.0 litre
Power	260 HPS	150 HPS
Engine operation strategy	Lean-burn/stoichiometric*	unknown
Fuel injection system	Gaseous port injection	Port injection
Catalytic converter	3-way catalyst	None
On-board storage	170 L liquid hydrogen	1.5 kg compressed H2 (248 bar)
Range	200 km (cruising range)	96 km
Fuel economy (Miles per gallon equivalent)	17 (cold start) -30 (highway) (FTP-75 cycle)	unknown
NOx (g/km)	0.0005 (FTP-75 cycle)	0.4598 (EPA-75 test cycle)

*Lean-burn in the low load region and stoichiometric operation in the high load region.

In a recent study performed by Westport and AVL [20] a modelling-based analysis is done for three hydrogen engine concepts; spark ignition (port fuel and direct fuel injection) and high-pressure direct injection with pilot injection (HPDI). The results of the analysis show that the H_2-HPDI with its diesel-like combustion cycle, efficiency and torque outperforms the spark ignition concepts in terms of efficiency, retained power density and combustion robustness. The study identifies H_2-HPDI as a very attractive solution for zero-CO_2 emission solution for the heavy-duty transportation sector because of the technical characteristics to meet the increasingly stringent emission regulations and low total costs of ownership (TCO) as compared to the other engine concepts and FCEV (near term). It should be noted that to our knowledge H_2-HPDI powertrains are not yet commercially available.

5.3. A COMPARISON BETWEEN HYDROGEN ICE AND HYDROGEN FUEL CELL VEHICLE TECHNOLOGIES

A major challenge for hydrogen fueled engine (either ICE of FCEV) is the low density of hydrogen. This means that a relatively large volume and/or better efficiency is required for an adequate driving range and power output of the engine as compared to methane. For most heavy-duty on-road vehicles there is limited space available to store the hydrogen, which means that the storage pressure has to be increased to be able to store more hydrogen or to use liquid hydrogen. This solution is currently being applied in hydrogen FCEV vehicles, where hydrogen is stored at 700 bars (as compared to 350 bars for CNG vehicles).

Gillingham [18] provides a rough sketch of the engine efficiencies at different loads for spark ignition (SI), compression ignition (CI) and a single fuel cell (equivalent output to the other engine types), see also Figure 35. The efficiency of the fuel cell reaches much higher values than the ones for the SI and CI engine as can be seen in Fig 35. However, as the load increases the efficiency drops because of the increasing demand for the subsystem components like compressor, vent, magnetic valves and management system. To compensate for the drop in efficiency, while delivering adequate power to the vehicle, additional cells need to be added, which is expensive [18]. Alternatively, the peak power demand is usually provided by a high-power high-capacity auxiliary energy storage source, e.g. a battery or an ultracapacitor. As a result, most fuel cell vehicle power train usually operated at an optimal steady state (see the red points in the figure 36), where the efficiency curve is relatively flat in a wide range around the efficiency peak.

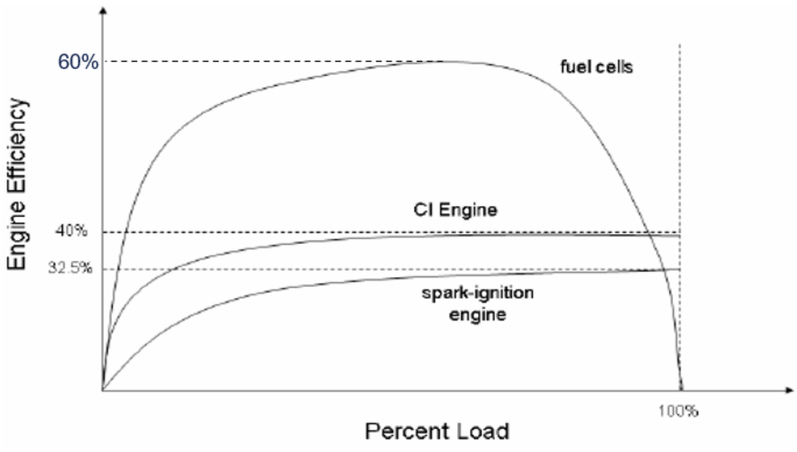

Figure 35 Engine efficiency versus load curves for spark ignition engine, compression ignition engine and a single fuel cell. The image is taken from [18].

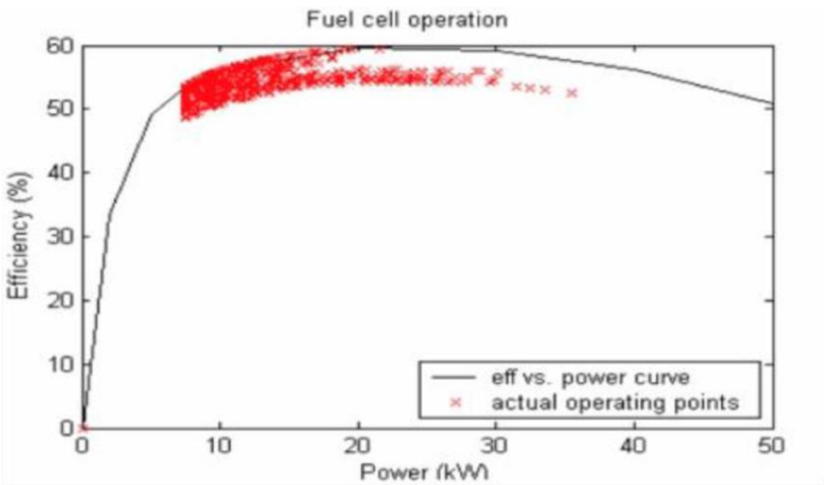

Figure 36 Fuel cell efficiency curve with actual operating points [21].

Based on the interviews with manufactures and the literature review a comparison is made between the hydrogen ICE and fuel cell technology (see Table 16).

Table 16 Comparison between H_2 ICE and H_2 fuel cell

	H2 ICE	H2 fuel cell (PEMFC)
Technology	Combustion	Electrochemical conversion
Fuel requirements	Less fuel quality requirements as compared to FCEV (hydrogen quality)	High purity H_2 (99.9999%, grade 5 H_2)
Max. Engine efficiency	~40%[18]	~60%[5]
Fuel Economy [miles per gallon equivalent]	30-40 [14, 18]	50 [18]
Maintenance [Euro/km] [19]	0.19-0.20*	0.48-0.53**
Total costs of ownership after 5 years of operation [€/km] [20]	1.4-1.5	1.6-1.8
Main advantages	Relatively low cost (known technology) Low re-engineering effort Less impact of external conditions (dust, cold conditions)	Higher efficiency No tailpipe emissions Fuel cell vehicles commercially available
Main disadvantages	Potential significant NOx emissions (Mitigating measures available such as EGR, injection strategy, fuel-lean limit) Development in research phase	High cost (e.g. precious metals) Higher maintenance cost.

* Assuming similar maintenance costs as for LNG/CNG truck
** Stack needs to be replaced after approximately 500.000 km [22-23]

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APPENDIX A: OVERVIEW OF HEAVY-DUTY VEHICLES CURRENTLY AVAILABLE IN THE EU MARKET

The data was retrieved from public database (internet mainly) and the NGVA vehicle Catalogue 2019. The inventory revealed that all heavy-duty vehicles currently being sold in the EU market are equipped with a 4-stroke spark-ignited stoichiometric engine and a three-way catalyst.

A-1. Heavy-duty Trucks

Brand	Name	Max. Power kW (hp)	Engine	EGR?	Engine specifications
Iveco	S-WAY NP CNG	294 (400)	FPT Cursor 9	No	6 cyl., 9L, 294kW, 1700Nm
Iveco	Eurocargo Natural Power 12-16 Ton	150 (240)	Tector engine		4 cyl, 4.5 L, 152 kW, 750 Nm
Mercedes	Econic NGT	222 (302)	M 936 G	Yes	6 cyl, 7.7 L, 222 kW, 1200 Nm
Renault	D Wide CNG	235 (320)	NGT9		6 cyl.,9 L, 236 kW, 1356 Nm
Scania	P/G CNG Version 280	205 (280)	G280		5 cyl.,9L; 206 kW, 1350 Nm
Volvo	FE CNG	239 (250)	G9K320		6cyl., 9 L, 239 kW, 1356 Nm

A-2. Heavy-duty Buses

Brand	Name	Max. Power kW (hp)	Engine	EGR?	Engine specs
Iveco	Urbanway Natural Power	213-243 (290-330)	FPT Cursor 8	No	6 Cyl., 7.8 L, 213 (10m) -243 (18m), 1100 (10m) -1300 (18m) Nm
Iveco	Crealis Natural Power	213-243 (290-330)	FPT Cursor 8	No	6 Cyl., 7.8 L, 213 (12m) -243 (18m), 1100 (12m) -1300 (18m) Nm
Iveco	Crealis Natural Power	263 (360)	FPT Cursor 9	No	6 Cyl., 9L, 265kW, 1650 Nm
Iveco	Crossway LE Natural Power	264 (360)	FPT Cursor 9	No	6 Cyl., 9L, 265kW, 1650 Nm
MAN	Lion´s City CNG	200-228 (272-310)	E2876 LUH	Yes	6 Cyl., 13L, 200-228kW, 1050-1250 Nm
Mercedes	Citaro (G) NGT	240 (302)	M 936 G	Yes	6 Cyl., 7.7 L, 222 kW, 1200 Nm
Mercedes	Citaro NGT hybrid	240 (302)	M 936 G	Yes	6 Cyl., 7.7 L, 222 kW, 1200 Nm
Mercedes	Conecto (G) NGT	222 (302)	M 936 G	Yes	6 Cyl., 7.7 L, 222 kW, 1200 Nm
Scania	Citywide LF CNG	206 (280)	OC09 (G280)	Yes	5 Cyl., 9L, 206 kW, 1350 Nm
Scania	Citywide LE CNG	235 (320)	OC09 (G280)	Yes	6 Cyl., 9L,250 kW, 1350 Nm
Scania	Interlink LD CNG	206-235 (280-320)	OC09 (G280)	Yes	7 Cyl., 9L, 208-250 kW, 1350 Nm
ISUZU	Cityport CNG	224 (300)	CUMMINS L9N	Yes	6 Cyl., 9 L, 239 kW, 1356 Nm
SOLARIS	Urbino 18 CNG	239 (320)	CUMMINS L9N	Yes	6 Cyl., 9 L,239 kW, 1356 Nm
VAN HOOL	Exqui.city 18	206 (280)	MAN E0836 LOH	No	6 Cyl., 7 L, 206 kW,1000 Nm
VAN HOOL	Exqui.city 24	250 (340)	FPT Cursor 9	No	6 Cyl., 9L, 294kW, 1700Nm

APPENDIX B: BACKGROUND INFORMATION METHANE NUMBER METHODS DESCRIBED IN THE LITERATURE

AVL and MWM method

Most methods used to calculate the methane number are derived from the AVL methodology [14], based on experimental work performed on a stochiometric engine. AVL uses a methane-hydrogen scale; pure methane is a knock resistant fuel and is assigned a value of 100, while hydrogen is knock sensitive and is given the value of 0. The AVL methodology includes hydrocarbons up to butane (higher hydrocarbons are treated as butane), CO_2, CO, H_2, O_2, H_2S and N_2. Engine manufacturers developed their own method based on the data of the AVL work and some implement modifications to fit the methodology to their engines.

The MWM method (published in the standard EN 16726 [7]) is based on the same data as the AVL methodology. In contrast to AVL, the MWM method ignores the effect of nitrogen, stating that nitrogen has no impact on the knock resistance of lean-burn engines [15]. Also, MWM extended the tool to include a maximum of 3% of higher hydrocarbons (n-pentane, hexane and heptane). Both MWM and AVL use complex relations to iteratively find the methane number for a given gas composition.

Traditionally used methods, such as MWM and AVL suffer from a number of shortcomings. For example, the suitability of hydrogen as a reference gas has been disputed [2, 9], as has been the method of accounting for butane and higher hydrocarbons [4]. Also, both MWM and AVL do not discriminate between isomers of higher hydrocarbons (butanes, pentanes, etc.) which are known to show different knocking behavior [4]. Since October 2020, the MWM MN method is incorporated into ISO standard 23306 [12].

CARB and GRI method [3, 5]

Both CARB and GRI/ISO methods use a correlation between the motor octane number (MON) and methane number [9-11]. The MON scale is used as octane number for liquid fuels and ranges from 100 corresponding to iso-octane to 0, which is assigned to n-heptane. Natural gases have a much higher knock resistance than iso-octane, typically in the range of 115-130+ [3, 11], often exceeding the maximum range of 120 on the ASTM octane number scale [11]. This is the reason that a methane number (separate knock scaling) was developed.

According to ref [11], there are two formulas for calculating the MON;

$$MON = 137.78 \times X_{CH4} + 29.948 \times X_{C2H6} - 18.193 \times X_{C3H8} - 167.062 \times X_{C4H10} + 181.233 \times X_{CO2} + 26.994 \times X_{N2} \quad (1)$$

Where, Xi is the mole fraction of component i. The limitation for the components for calculating the MON of natural gas are summarized in the Table below [11]:

Table B-1 Maximum mole fractions for the GRI octane test [13]

Species	Limitations [%]
CH_4	≥75
C_2H_6	≤14
C_3H_8	≤25
C_4H_{10}+	≤1.0
CO_2	≤1.8
N_2	≤3.5

The other equation for calculating the MON is based on the reactive H/C ratio, which is the ratio of the hydrogen to carbon atoms excluding the carbon atoms in the inerts (specifically CO_2) [1, 3, 5, 13];

$$MON = -406.14 + 508.04 \cdot \frac{H}{C} - 173.55 \cdot \left(\frac{H}{C}\right)^2 + 20.170 \cdot \left(\frac{H}{C}\right)^3 \quad (2)$$

According to Ref [3], the above relation is only for H/C ratios above 2.5 or inert concentrations below 5%.

The GRI method also uses the following equation to calculate the methane number [1, 3, 5, 13]:

$$GRI\ MN = 1.445 \times MON - 103.42 \quad (3)$$

Since there are two equations for calculating the MON (1 and 2), there also two methane numbers that can be calculated following the GRI method. If for the same gas, large differences are observed, then the ISO standard 22302 recommendations are shown in the Table below [13].

Table B-2 **ISO standard 22302 recommendation for the GRI MN method [13]**

Difference between GRI MN values	Recommendation according to ISO
>6	two MNs are in doubt, and another method such as AVL should be used instead
>10	Gas composition is unusual (e.g. contains more N_2 or CO_2)

The CARB method only uses the relation between the MON and the H/C ratio to calculate the MON for the natural gas under investigation (equation 2). To calculate the methane number the following relation is used [3];

$$CARB\ MN = 1.624 \times MON - 119. \quad (4)$$

Waukesha Knock Index (WKI) [16]

The Waukesha Knock Index (WKI) to characterize the knock resistance of a gaseous fuel is described by Sorge et al. [16]. The WKI method is developed from experimental data where blends of natural gases were tested using a stationary engine [16]. The method for calculating the methane number uses either a polynomial equation (similar as PKI MN, see below) or a C/H ratio method (similar to the method used by GRI and CARB). The polynomial equation is used in case the species concentrations meet the following criteria:

- Methane: 60 – 100 vol%
- Ethane: 0 – 20 vol%
- Propane: 0 – 40 vol%
- N-Butane: 0 – 10 vol%
- N- Pentane: 0 – 3 vol%
- Hexane+: 0 - 2vol%
- Nitrogen: 0 – 15 vol%
- Carbon dioxide: 0 - 10 vol%

Most natural gases typically fall within this concentration range, which means that in most cases the polynomial equation is used to calculate the methane number.

For gas compositions that fall outside this range of concentrations, a C/H ratio method is used, where the C/H ratio can be converted into a methane number using a given calibration curve [16]. Adjustments are made for inert gases when using the C/H ratio method.

The method also includes the effect of iso-butane by assigning 58% of the iso-butane concentration to propane and 42% of the iso-butane to normal butane. A similar approach is used to include the effect of iso-pentane (68% to n-butane and 30% to n-pentane) [16]. The method also enables calculation of the methane number calculation for gaseous fuels that contain hydrogen, carbon monoxide and H_2S.

Cummins Methane Number (CMN) [6]

In November 2015, Cummins Westport launched their fuel quality calculator, which calculates the methane number and lower heating value for a given gas composition. The online tool uses a traffic light to indicate if the fuel meets the required specification for a number of Cummins Westport Engines. The online tool includes the effect of iso-butane and iso-pentane, higher hydrocarbons including n-hexane, n-heptane, n-octane, n-nonaan and n-decaan, hydrogen (up to 0.03 mole%), oxygen, nitrogen, carbon monoxide, carbon dioxide, H_2S (up to 6 ppmv), sulfur (up to 0.001 weight %) and siloxanes (up to 0.0003%).

Wärtsilä Methane Number (WMN) [17]

On the Wärtsilä website the methane number for gaseous fuels can be calculated. The tool is based on the PKI methane number method (see below). The tool provides information whether the fuel can be used in Wärtsilä engines based upon the calculated methane number. The tool calculates the methane number for hydrocarbons (up to octane), including iso-butane and iso-pentane, carbon monoxide, carbon dioxide, hydrogen, nitrogen and H_2S. The validity of the tool is restricted to the following concentration ranges

- Methane: 70 - 100 mol%
- Ethane: 0 – 30 mol%
- Ethene: 0 - 5 mol%
- Propane: 0 – 30 mol%
- Propylene: 0 – 5 mol%
- N-Butane: 0 - 10 mol%
- I-Butane: 0 – 10 mol%
- N- Pentane: 0 – 5 mol%
- I- Pentane: 0 – 5 mol%
- Neo- Pentane: 0 – 5 mol%
- Mix C6H14: 0 – 4.02 mol%

- Mix C7H16: 0 – 3.44 mol%
- N-C8H18: 0 – 1.82 mol%
- Nitrogen: 0 – 20 mol%
- Carbon dioxide: 0 – 20 mol%
- Carbon moxide: 0 – 10 mol%
- Hydrogen: 0 – 30 mol%
- Hydrogensulfide: 0 – 1 mol%

PKI MN method

DNV GL developed a methane number method ("PKI MN") that characterizes gases for their knock resistance based on the combustion properties of the fuel mixtures themselves [8, 10, 12]. In contrast to the methods described above, which use a methane-hydrogen scale, the PKI MN method is based on a methane-propane scale (PKI, Propane Knock Index).

Additionally, while AVL and MWM use complex relations to iteratively calculate the methane number, the PKI MN method uses a polynomial equation:

$$PKI = \sum \alpha_{i,n} x_i^n + \sum \beta_{i,n} x_i^n x_j^m \quad (5)$$

Herein i= CH_4, C_2H_6, C_3H_8, i-C_4H_{10}, n-C_4H_{10}, n-C_5H_{12}, i-C_5H_{12}, neo-C_5H_{12}, CO_2, CO, H_2 and N_2, n = 1-4 and m =1,2. The α and β coefficients can be found in Ref [10].

The Table below summarizes the range of fuel composition for which the PKI MN method is valid. It should be noted that this covers a range of natural gases distributed including fractions of species such as H_2 and CO.

Table B-3 PKI MN range of gas compositions

Species	Range (mol%)
CH_4	70-100
C_2H_6	0-20
C_3H_8	0-20
i-C_4H_{10}	0-5
n-C_4H_{10}	0-5
n-C_5H_{12}	0-2
i-C_5H_{12}	0-2
neo-C_5H_{12}	0-2
C_6+	0-1.5
CO	0-10
CO_2	0-20
N_2	0-20
H_2	0-20
H_2S	0-0.5

To put the method on a scale analogous to the currently used Methane Number methods, the propane-based scale (PKI) has been converted to a 0-100 scale, referred to as PKI MN using the following equation:

$$PKI\ MN = a \times PKI + b \times (PKI)^2 + c \times (PKI)^3 + d \times (PKI)^4 + e \times (PKI)^5 + f \times (PKI)^6 + g \quad (6)$$

Table B-4 Coefficients for converting PKI to PKI MN [10, 12]

Coefficient	Value
a	-9.757977
b	1.484961
c	-0.139533
d	7.031306×10-3
e	-1.770029×10-4
f	1.75121×10-6
g	100

The PKI MN method is developed and verified for a high-speed, lean-burn, spark-ignited CHP engine. In 2017, the PKI MN methodology was also applied to develop dedicated methane number algorithms for a mono-gas variable-speed, stoichiometric, spark-ignited gas engine typically used in heavy-duty road transportation and a dual-fuel, ultra-lean-burn medium-speed engine used on ships [8]. The results show that the ranking of the knock resistance of fuel compositions differs among the different engine platforms [8]. For the engines tested, the method has shown superior performance as compared to AVL and MWM methods [8, 9]. Since October 2020, the PKI MN method is incorporated into ISO standard 23306 [12].

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APPENDIX C: GAS COMPOSITION USED IN KNOCK EXPERIMENTS

natural gas	G-gas			3% H2			6% H2		
	mole fraction	mole %	WKI (exlc.C6)	mole fraction	mole % (exlc. He en norm.)	WKI (exlc.C6)	mole fraction	mole % (exlc. He en norm.)	WKI (exlc.C6)
CH4	0.819205357	81.97	82.03	0.793632532	79.40	79.45	0.768707616	76.90	76.95
C2H6	0.029286238	2.93	2.93	0.02842307	2.84	2.85	0.027631553	2.76	2.77
C3H8	0.004433323	0.44	0.44	0.004289496	0.43	0.43	0.004196696	0.42	0.42
n-C4H10	0.000804632	0.08	0.08	0.000778013	0.08	0.08	0.00075775	0.08	0.08
i-C4H10	0.000687649	0.07	0.07	0.000665082	0.07	0.07	0.000647587	0.06	0.06
n-C5H12	0.000196101	0.02	0.02	0.000190517	0.02	0.02	0.000185808	0.02	0.02
i-C5H12	0.000199299	0.02	0.02	0.000193846	0.02	0.02	0.000189493	0.02	0.02
neoC5H12	0	0.00	0.00	0	0.00	0.00	0	0.00	0.00
C6H14	0.000738865	0.07	0.07	0.000715063	0.07	0.07	0.000698584	0.07	0.07
CO2	0.009623531	0.96	0.96	0.00936	0.94	0.94	0.009134535	0.91	0.91
O2	0	0.00	0.00	0	0.00	0.00	0	0.00	0.00
N2	0.134183679	13.43	13.44	0.129930072	13.00	13.01	0.125643219	12.57	12.58
H2	0	0.00	0.00	0.031409149	3.14	3.14	0.061824087	6.18	6.19
H2O	0	0.00	0.00	0	0.00	0.00	0	0.00	0.00
He	0.000641325		0.00	0.000416296		0.00	0.000383071		0.00
Ar	0	0.00	0.00	0	0.00	0.00	0	0.00	0.00
CO	0	0.00	0.00	0	0.00	0.00	0	0.00	0.00
Total		100			100			100	

natural gas	11% H2			15% H2			20% H2		
	mole fraction	mole %	WKI (exlc.C6)	mole fraction	mole % (exlc. He en norm.)	WKI (exlc.C6)	mole fraction	mole % (exlc. He en norm.)	WKI (exlc.C6)
CH4	0.730756808	73.10	73.15	0.689369086	68.94	68.98	0.654556889	65.46	65.49
C2H6	0.026297859	2.63	2.63	0.02482791	2.48	2.48	0.023546639	2.35	2.36
C3H8	0.004008513	0.40	0.40	0.003810397	0.38	0.38	0.003634694	0.36	0.36
n-C4H10	0.000722384	0.07	0.07	0.000684335	0.07	0.07	0.000651699	0.07	0.07
i-C4H10	0.000616347	0.06	0.06	0.000584411	0.06	0.06	0.000557839	0.06	0.06
n-C5H12	0.000177423	0.02	0.02	0.000167681	0.02	0.02	0.000159548	0.02	0.02
i-C5H12	0.000252092	0.03	0.03	0.000239922	0.02	0.02	0.000229782	0.02	0.02
neoC5H12	0	0.00	0.00	0	0.00	0.00	0	0.00	0.00
C6H14	0.000670114	0.07	0.07	0.000631829	0.06	0.06	0.000599545	0.06	0.06
CO2	0.008635747	0.86	0.86	0.008160136	0.82	0.82	0.00776833	0.78	0.78
O2	0	0.00	0.00	0	0.00	0.00	0	0.00	0.00
N2	0.118924938	11.90	11.90	0.112081204	11.21	11.22	0.106363855	10.64	10.64
H2	0.108592924	10.86	10.87	0.159380303	15.94	15.95	0.201931178	20.19	20.21
H2O	0	0.00	0.00	0	0.00	0.00	0	0.00	0.00
He	0.00034485		0.00	6.27854E-05		0.00	0	0.00	0.00
Ar	0	0.00	0.00	0	0.00	0.00	0	0.00	0.00
CO	0	0.00	0.00	0	0.00	0.00	0	0.00	0.00
Total		100			100			100	

APPENDIX D: EXPERIMENTAL ENGINE KNOCK DATA FOR CNG MIXTURES

Literature review revealed that the amount of experimental gas engine knock data are very scarce. An exception is the data published in 2017 [1] where the knock resistance of different LNG compositions was tested on a stoichiometric CNG truck engine (FPT cursor 9). The measured knock resistance (Knock Limited Spark Timing, KLST [1]) was compared with PKI, AVL and MWM calculated methane numbers. The results are shown in Figure D-1 below.

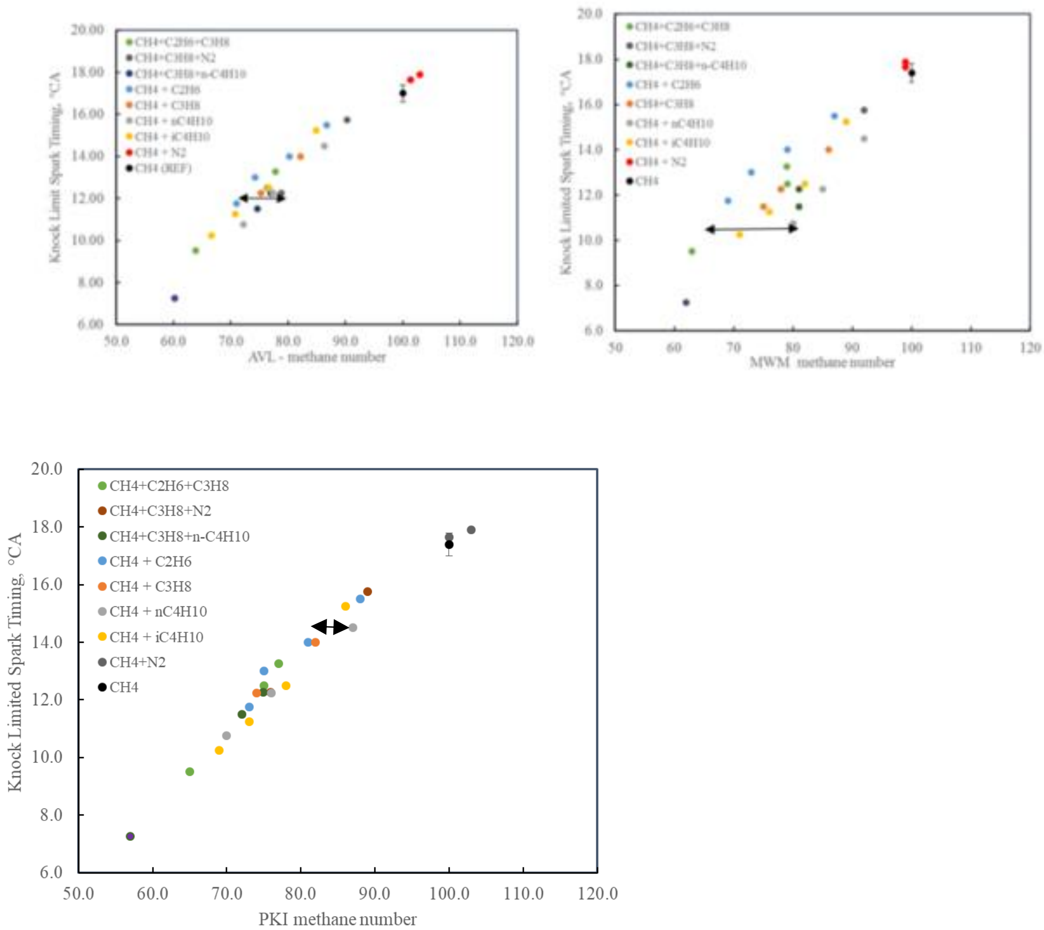

Figure D-1 Experimentally determined knock resistance (KLST) versus AVL (top), MWM (middle) and PKI (bottom) methane numbers for a CNG truck engine. The horizontal arrows indicate the spread in the calculated Methane Numbers at a fixed KLST.

The results in Figure D-1 illustrate that the importance of which engine platform was used to validate the method. For example, the largest difference between experimentally (KLST) and calculated (methane number) knock resistance is observed for the MWM method. This is not surprising, since this method is validated for lean-burn gas engines which can have different in-cylinder conditions (pressure and temperature) than the stoichiometric truck engine used in this experiment. A better agreement between calculated and experimental data is found for AVL, which is validated for a stoichiometric engine. Although the PKI MN method was validated on a fuel-lean CHP engine, it was expected to provide a similar result as acquired using MWM. However, as can be seen from Figure 3, an excellent agreement is found between measured and calculated knock resistance. This result shows that the importance of testing the different Methane number methods by comparing the results with experimental data.

It should be noted that the study in ref [1] was limited to LNG compositions and did not include the effect of hydrogen addition. In fact, experimental data where knock behavior of hydrogen – CNG mixtures were studied for CNG engine could not be found.

For this reason, **it is highly recommended to experimentally investigate the knock behavior of CNG-hydrogen mixtures in CNG engines and compare the results with the outcome of the above discussed methane number methods**. This offers an opportunity to select the methane number method that unambiguously calculate the impact of hydrogen addition to natural gas on the occurrence of engine knock.

References

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APPENDIX E: CALCULATED RESULTS FOR CASE STUDY

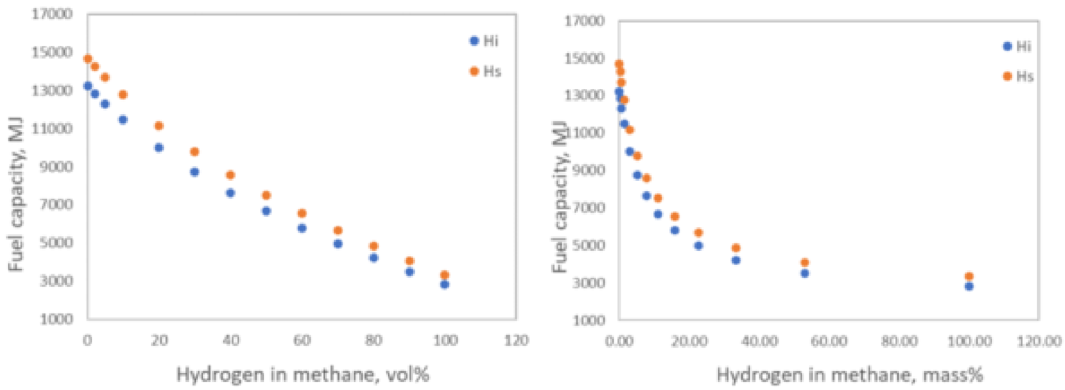

Figure E-1 Energy content CH_4/H_2 using a 1575 m3 tank at a pressure of 200bar, see assumptions in Table 8.

It can be observed a non-linear evolution of energy content as a function of hydrogen content in methane expressed in vol% (Figure E-1, left). The explanation is that CH_4 and H_2 do not behave as perfect gases at high pressure. Therefore, their compressibility factor (Z) needs to be considered. Interestingly, the behaviors of the two molecules are different: for methane the attractive forces are more dominant, resulting in compressibility factors lower than 1 (favorable to energy content by volume at high pressure), while for hydrogen the repulsive forces are dominant resulting in a compressibility factor greater than 1 (unfavorable to energy content by volume at high pressure).

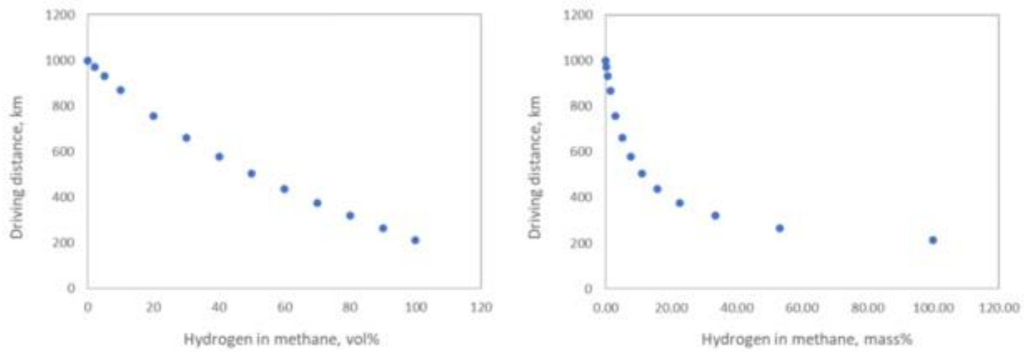

Figure E-2 Driving distance (mileage) using assumptions in Table 8.

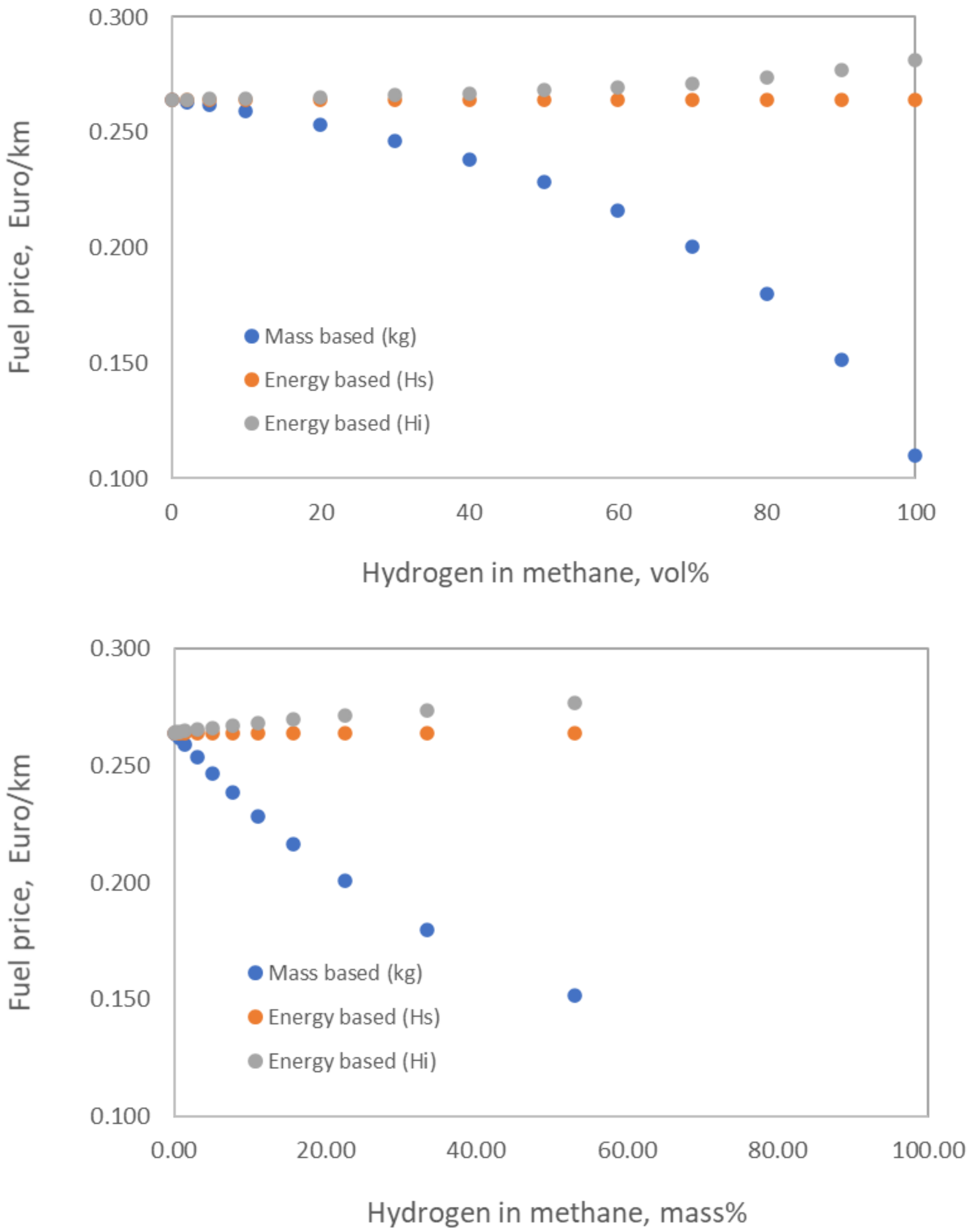

Figure E-3 Fuel price/km using assumptions presented in Table 8: billing based on kg, Hs, Hi.

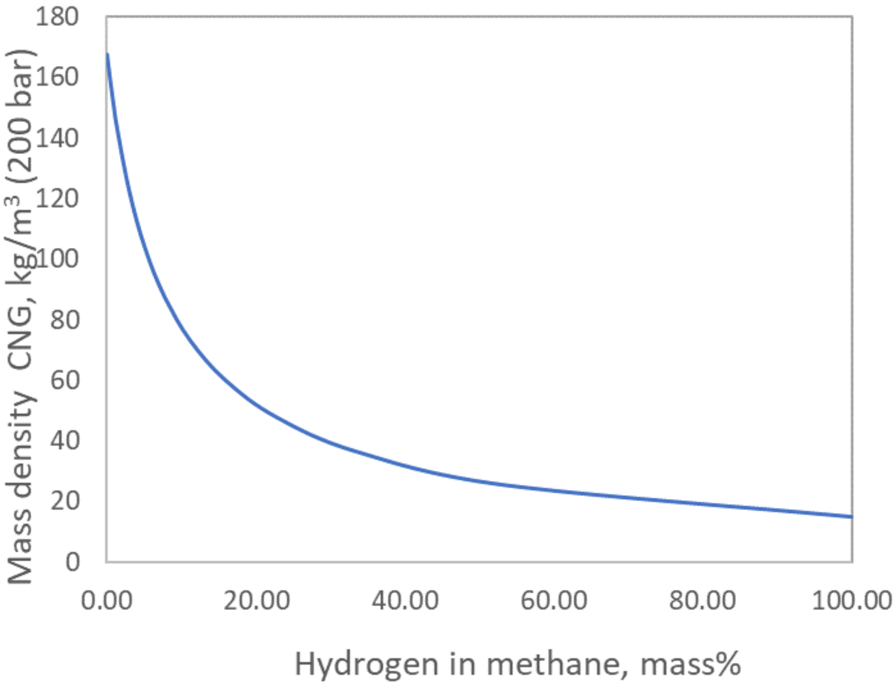

Figure E-4 Mass density of CNG as function of hydrogen content in CNG

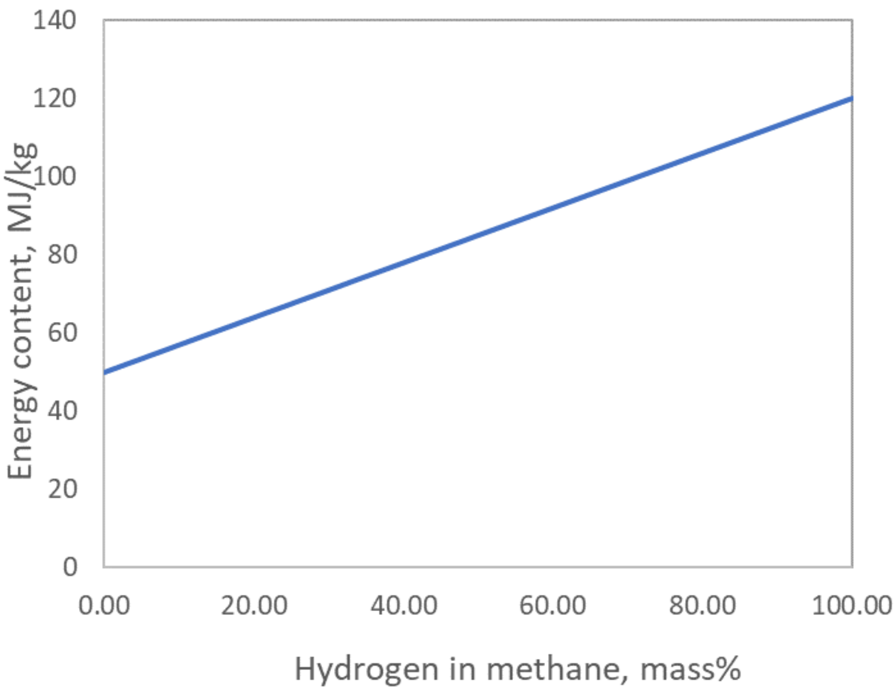

Figure E-5 Energy content as function of hydrogen content in CNG

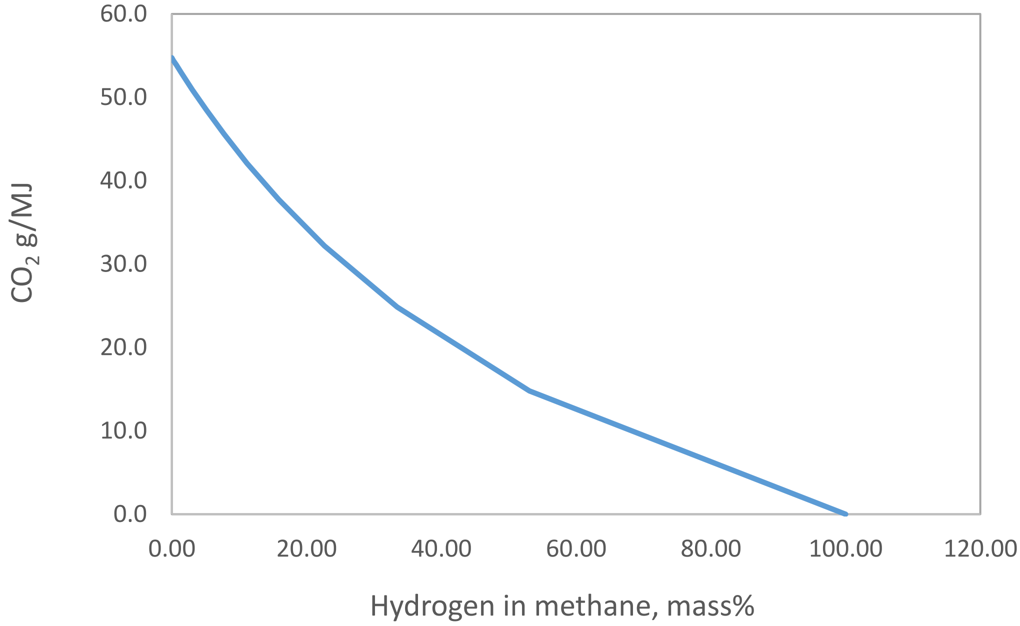

Figure E-6 CO2 emission as function of hydrogen content in CNG

6. GLOSSARY

AFR	Air/Fuel Ratio
AVL	Anstalt für Verbrennungskraftmaschinen List.
BMEP	Brake Mean Effective Pressure, parameter to express the effective work per combustion cycle.
BTDC	Before Top Dead Center.
BTE	Brake Thermal efficiency.
CI	Compression Ignition (engine), often referred as “Diesel engine”.
CMN	Cummins Methane Number.
CNG	Compressed Natural Gas, natural gas compressed to high pressures (up to 350 bar).
COV-IMEP	Coefficient of Variance (COV) of the Indicated Mean Effective Pressure (IMEP), parameter widely used to quantify the combustion stability.
CR	Compression Ratio, the ratio of maximum cylinder volume (swept volume + clearance volume) to minimum cylinder volume (clearance volume).
Direct Injection	Fuel dosing method in which fuel is injected directly into the cylinder.
DPF	Diesel Particulate Filter, device to remove particles/soot from the exhaust gas.
DNG	Dutch Natural Gas, natural gas consisting of roughly 82.0 mole% CH4, 2.7 mole% C2H6, 0.4 mole% C3H8, 0.9 mole% CO2 and 14.0 mole% N2
ECU	Engine Control Unit, electronic control unit for a group of or all key engine parameters.
EGR	Exhaust Gas Recirculation.
FCEV	Fuel Cell Electric Vehicle.
FID	Flame Ionization Detector.
FPD	Flame Photometric Detector.
GC	Gas Chromatography
GHG	GreenHouse Gas emissions, refer to the weighted sum of carbon dioxide (CO2), methane (CH4), nitrous oxide (N2O), perflourocarbons (PFCs),

	hydroflourocarbons (HFCs) and sulphur hexafluoride (SF6), using their 100-year global warming potentials[1].
HCNG (or H_2/CNG)	Hydrogen Compressed Natural Gas, mixture of compressed natural gas and hydrogen.
HPDI	High Pressure Direct Injection, fuel dosing method in which fuel is injected directly into the cylinder at high pressure, typically near the end of the compression phase of the combustion cycle.
ICE	Internal Combustion Engine.
IMEP	Indicated Mean Effective Pressure, parameter to express the indicated work per combustion cycle.
ITE	Indicated Thermal Efficiency.
KLST	Knock-Limited Spark Timing, spark timing delivering borderline knock at the given operating conditions.
LFL	Lower Flammability Limit, equivalence ratio below which combustion is impossible
LNG	Liquefied Natural Gas, natural gas that has been converted to a liquid state by cooling to below -163°C (at ambient pressure).
MN	Methane Number, parameter to quantify the resistance to knock of gaseous fuels.
MWM	Motoren Werke Mannheim, German engine manufacturer (now part of Caterpillar).
OEM	Original Equipment Manufacturer.
PEMFC	Polymer Electrolyte Membrane Fuel Cell, fuel cell based on a membrane that only lets positive hydrogen ions pass through.
PKI MN	Propane Knock Index Methane Number, methane number calculation method developed by DNV.
Port Injection	Fuel dosing method in which fuel is injected into the cylinder-individual intake manifold ports.
SCR	Selective Catalytic Reduction, catalyst system using a reducing agent (e.g. ammonia) to remove nitric oxides from exhaust gas of lean-burn operated engines.
SI	Spark Ignition (engine), often referred as "Otto engine".
S_L	Laminar flame Speed, speed at which a flame propagates in laminar conditions

[1] https://www.eea.europa.eu/data-and-maps/indicators/ghg-emissions-outlook-from-iea

STP	Standard Temperature and Pressure, here defined as 273.15 K and 1.01325 bar respectively.
SOFC	Solid Oxide Fuel Cell, fuel cell in which the electrodes are separated by a solid ceramic electrolyte that only allows diffusion of oxygen ions.
TCD	Thermal Conductivity Detector.
TCO	Total Costs of Ownership, a total cost which includes all costs related to owning a vehicle: vehicle purchase and resale, fuel costs, maintenance and repair costs, etc.
TDC	Top Dead Centre, position of the piston at minimum cylinder volume.
TSO	Transmission System Operator, entity entrusted with transporting energy in the form of natural gas or electrical power on a national or regional level, using fixed infrastructure.
TWC	Three-Way Catalyst, catalyst system to remove nitric oxides, hydrocarbons and carbon monoxide from exhaust gas of stoichiometric operated engines.
UFL	Upper Flammability Limit, equivalence ratio above which combustion is impossible
WHTC	World Harmonized Transient Cycle, standardized transient engine emission test cycle.
WKI	Waukesha Knock Index, engine knock measurement method.

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