Combustion Properties of Natural Gas with Hydrogen and Behaviour in Industrial Equipment

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EXECUTIVE SUMMARY

The addition of hydrogen into the natural gas network is being considered as a practical way to reduce GHG emissions from combustion equipment. This report has looked at potentially serious changes to combustion properties and surveyed the scientific literature investigating the behaviour of hydrogen enriched natural gas in industrial combustion equipment.

There are three combustion properties that increase significantly with the addition of hydrogen: flame speed, upper flammability limit, and flame temperature. This enhanced combustion has been used to improve the performance of gas turbines and internal combustion engines. Generally, addition of hydrogen increases the emissions of NOx because the higher flame temperature increases the thermal NOx formation mechanism. Flashback is a safety concern for using unmodified equipment with hydrogen enriched natural gas.

Finally, one has to consider the actual scenario for hydrogen addition to the natural gas network. The hydrogen content could vary, perhaps with the variation in solar and wind electicity output. This variation can be tracked and process controls adjusted to maintain efficient and safe operation, and control emissions of regulated pollutants (NOx). However, an industrial facility may not have the necessary instrumentation to measure the composition of the fuel gas in real time, or a sophisticated control system to make the necessary adjustments. Such users can experience degraded performance and increased emissions even with the maximum hydrogen threshold set at 10%-vol.

RECOMMENDATION: To realize the benefits of enriching natural gas with hydrogen, industrial combustion facilities need to be properly instrumented, the control systems modified, and the operators trained to take full advantage of the new fuel. This is particularly important for Power-to-Gas scenarios that introduce significant fluctuations in the fuel gas properties.

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INTRODUCTION

Natural gas is the lowest carbon intensity fossil fuel, giving 2.74 kg CO_2/kg NG or 49.5 kg CO_2/kJ fired. The replacement of coal with natural gas, especially for power generation, has produced significant reductions of GHG emissions. However, greater reductions are still required. One option is to produce renewable natural gas (RNG) by anaerobic digestion of organic wastes or gasification of biomass. RNG requires no change to downstream equipment. Another option is to introduce hydrogen into the existing natural gas infrastructure. Hydrogen would be produced from water electrolysis with nuclear or renewable electricity, steam-methane reformation (SMR) with CO_2 capture, gasification of biomass, water dissociation at high temperature, photo-electrolysis and biological processes [Ball and Wietschel (2009)][2]. Hydrogen combustion produces H_2O only, with NO_x being the major pollutant of concern. The hydrogen-enriched natural gas could serve as a bridge between the fossil fuel used today and a hydrogen fuelled economy in the medium future.

An early application of hydrogen enrichment of natural gas combustion was not motivated by GHG reduction but NOx reduction. Natural gas combustion in lean premixed mode results in lower flame temperature and reduced NOx emissions [Schefer (2003), Hawkes et al. (2004), Kim et al. (2009)]. However, the stability of the CH_4/air flame decreases and flame blow off may occur when operating close to the lean (lower) flammability limits. Even above the stability limit, there is poor combustion efficiency (increases the CO and unburned hydrocarbon (UHC) emissions) and acoustic instability (noise). Hydrogen enrichment increases combustion stability, avoids the operational issues and enables NOx reduction.

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[2] Some have been colour-coding the hydrogen as Green or Blue, and maybe someday Tourquoise. The important characteristic is that fossil carbon does not enter the atmosphere during its production.

First a word about natural gas. There is no such a thing as "a molecule of natural gas", since it is a mixture of methane, ethane, propane, nitrogen, carbon dioxide, and trace amounts of some other gases including hydrogen. The variability of natural gas is illustrated in Table 1, which gives the ranges for the major components in the gas delivered by a major utility in Ontario, Canada. There is similar variability around the world. Presumably the equipment firing natural gas can handle this variability, perhaps with some minor degradation of performance. The range of the heating value and Wobbe Number provide a target range when considering mixtures of natural gas with hydrogen.

Table 1 - Variability of composition of natural gas delivered by Union Gas.[3]

Component	Range (mole %)
Methane	87.0 - 97.0
Ethane	1.5 - 9.0
Propane	0.1 - 1.5
Nitrogen	0.2 - 5.5
Carbon Dioxide	0.05 - 1.0
Hydrogen	trace - 0.02
Gross Heating Value (MJ/m^3)	36.0 - 40.2
Wobbe Number (MJ/m^3)	47.5 - 51.5

This report looks at how combustion changes when natural gas is adulterated with hydrogen, whether combustion equipment designed for natural gas can be operated safely using natural gas blended with hydrogen, and what has

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[3] As reported on the Union Gas website, https://www.uniongas.com/about-us/about-natural-gas/chemical-composition-of-natural-gas, accessed on 2020-01-28.

been published on the actual experience with hydrogen/natural gas mixtures. The review starts with the general combustion characteristics, followed by a survey of the industrial combustion applications (primarily burners) and sections for gas turbines and reciprocating engines.

GENERAL COMBUSTION CHARACTERISTICS

As gaseous fuels, natural gas and hydrogen are at opposite ends of the spectrum on most properties: NG has a low flame temperature, high ignition energy, narrow range of flammability, and low flame speed; hydrogen has high flame temperature, low ignition energy, very wide range of flammability, and high flame speed. This raises questions of safety, flashback and explosion, when using hydrogen in combustion equipment designed for natural gas. The basic properties are listed in Table 2.

Table 2 - Basic properties relevant to combustion for methane and hydrogen.

		CH_4	H_2
Density	kg/m^3	0.679	0.085
LHV v	MJ/m^3	34.3	10.4
LHV m	MJ/kg	50.5	122.4
HHV v	MJ/m^3	37.6	12.1
HHV m	MJ/kg	55.4	142.0
LFL	%	5.0	4.0
UFL	%	15.0	74.2
SAF	kg/kg	17.20	34.29
Products/unit mass fuel			
CO_2	kg/kg	2.74	0.00
H_2O	kg/kg	2.25	8.92
N_2	kg/kg	13.21	26.22
Products/unit energy			
CO_2	kg/kJ	49.5	0.0
H_2O	kg/kJ	40.5	62.8
N_2	kg/kJ	238.4	184.7
Wobbe Number	MJ/m^3	50.5	45.9
Flame Speed[4]	cm/s	40.5	210
Adiabatic Flame Temperature[4]	°C	2053	2247

These properties are important in designing and predicting the progress of combustion and performance of combustion systems where hydrogen is used

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[4] At stoichiometric conditions.

as fuel. The burning velocity is closely related to the flame stability when compared to the exit velocity of the fuel-air mixture; a burning velocity that is too low results in blow-off, whereas a burning velocity that is too high gives rise to flashback. The flammability limits are the boundaries for a fuel/air mixture to maintain a flame. The Upper Flammability Limit (UFL) is the maximum fuel content, the Lower Flammability Limit (LFL) is the minimum, usually expressed as a percentage. Outside the flammability limits the fuel:air mixture does not sustain a flame. The wide flammability range for hydrogen, with a high flame speed, indicates an increased risk of fire or explosion in case of a leak.

The Wobbe Number is higher heating value on a volume basis divided by the square root of the specific gravity of the fuel gas, in symbols below.

$$WN = \frac{HHV_v}{\sqrt{\rho_f/\rho_a}} \tag{1}$$

Two fuel gases with similar Wobbe Numbers, when supplied to the same nozzle at the same delivery pressure, will give similar heat release. This makes the Wobbe Number a useful measure for relatively simple burner systems, such as found in domestic fired equipment (stove-top burners, hot water heaters) with gaseous hydrocarbon fuels. It is less useful for sophisticated industrial fired equipment, where the molecular weight, adiabatic flame temperature and lower heating value should also be matched [Baukal (2001) pp. 435-437]. Figure 1 shows the change in Wobbe Number for hydrogen enriched methane and the range of Wobbe Number for natural gas as delivered given in Table 1.

The flammability limits for a mixture of combustible gases can be calculated from the pure gas limits using LeChatelier's Principle.

$$\frac{1}{FL_{mix}} = \sum_i \frac{x_i}{FL_i} \tag{2}$$

This formula was confirmed as a good estimate for a large number of gas mixtures, including those with hydrogen, in Coward and Jones (1952). It is

clear from Figure 2 that while the LFL decreases slightly with the addition of hydrogen, the UFL increases substantially. This shows that in the case of leaks, either of the gas mixture into air or of air into the fuel lines, the chance of forming a flammable mixture increases by 50%, as measured by the spread in the flammability limits.
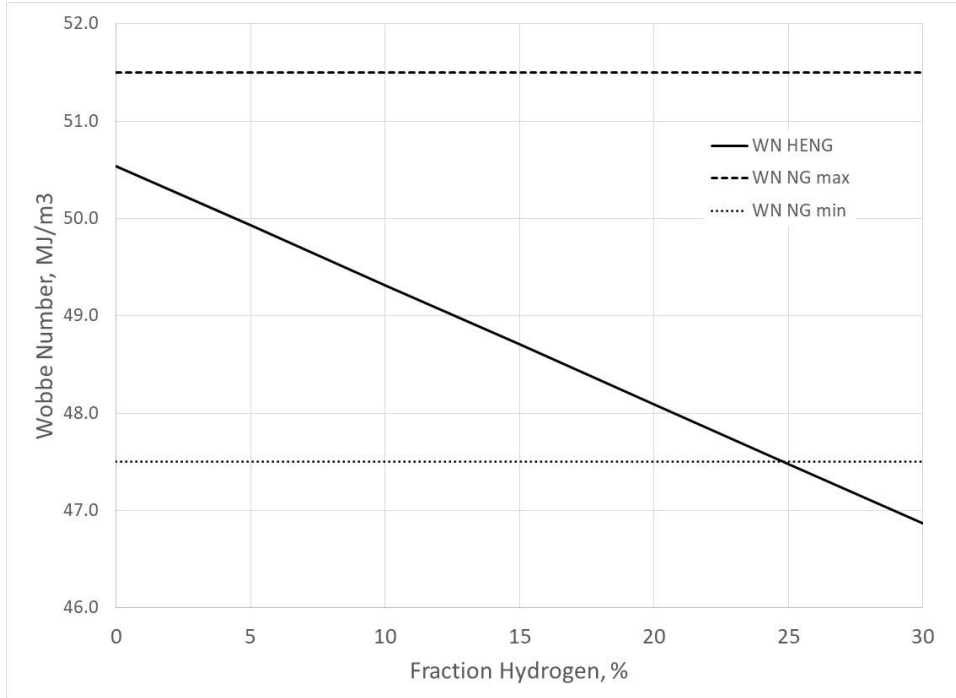

Figure 1 - Change in Wobbe Number for hydrogen-enhanced methane with hydrogen concentration. Included are the maximum and minimum Wobbe Number for delivered natural gas.

Another major difference in the combustion properties of methane and hydrogen is the laminar flame speed. This is the speed at which a flame propagates through a mixture of the combustible gas and oxidizer, most commonly air. There are two flame speeds commonly noted – that at stoichiometric conditions and the maximum flame speed. For many combustible gases, including methane, the maximum flame speed occurs near stoichiometric conditions. However, for hydrogen the maximum occurs in an excess of air, an equivalence ratio of 1.6. In the context of hydrogen enrichment of natural gas, the maximum flame speed is close to stoichiometric and for the following we will concentrate on the flame speeds at stoichiometric conditions.

Yu et al. (1986) investigated the laminar burning rate of mixtures of methane and hydrogen, and propane and hydrogen, in air. They produced a linear correlation of the laminar burning rate with the relative hydrogen content, R_H
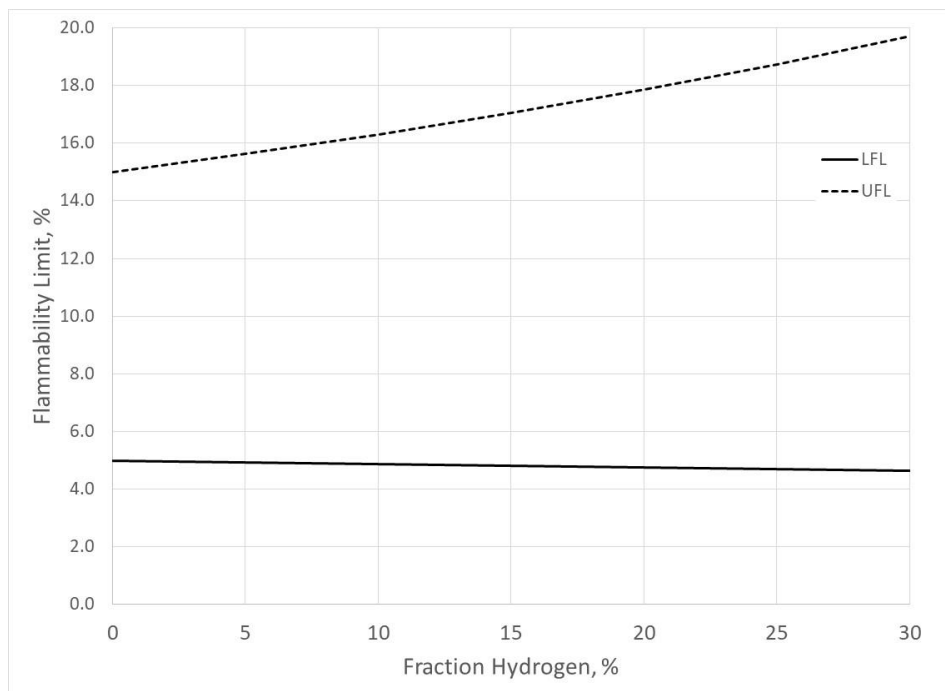

Figure 2 - Change in flammability limits with increase of concentration of hydrogen in methane.

, which works with both hydrocarbon species.

$$S_{L,mix} = S_{L,o} + 83R_H \tag{3}$$

$$R_H = \frac{x_H + \dfrac{x_H}{\left(\dfrac{x_H}{x_A}\right)_{st}}}{x_F + \left(x_A - \dfrac{x_H}{\left(\dfrac{x_H}{x_A}\right)_{st}}\right)} \tag{4}$$

The mole fractions are for air (A), hydrocarbon (F), and hydrogen (H), in the mixture. This correlations works for hydrogen enrichment levels of R_H up to one, or about 70% hydrogen.

Ilbas et al. (2006) measured experimentally the burning velocities of different compositions of hydrogen–methane–air mixtures (from 100% hydrogen to 100% methane) at ambient conditions for variable equivalence ratios using Schlieren photography technique. Their experimental study demonstrated that increasing the hydrogen increases the resultant burning velocity and widening the flammability limits. The authors as a result from their experiments suggested that a hydrogen–methane mixture of 30% hydrogen and 70% methane could be a competitive alternative fuel for existing combustion plants.

Huang et al. (2006) experimentally studied the laminar flame characteristics of CH_4/H_2/air flames in a constant volume bomb at normal temperature and pressure. The authors obtained the laminar burning velocities at various hydrogen concentrations (0 to 100 vol-%) and equivalence ratios (0.6 to 1.4) by using the Schliern photographic method and a high-speed camera. They found that the increment of the laminar burning velocities increased exponentially with the increase of hydrogen fraction in the mixture. A formula for calculating the laminar burning velocities of CH_4/H_2/air flames was proposed.

$$\frac{S_{mix} - S_{CH_4}}{S_{H_2} - S_{CH_4}} = 0.00737 \exp\left(\frac{x_{H_2}}{20.38}\right) + 0.00334 \tag{5}$$

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They provided polynomial fits for the flame flame speeds for methane and hydrogen as functions of the equivalence ratio. They claim the correlation covers the full range of hydrogen content and equivalence ratio from 0.6 to 1.4. This claim is only approximately true, for it is clear that equation (4) does not give the laminar flame speed of methane or hydrogen at the boundary values of 0% and 100% hydrogen. The author[5] has combined the work of Yu et al. (1986) and Huang et al. (2006) to obtain a more accurate fit, see Figure 3. At 30% hydrogen the laminar flame speed has increased by 12 cm/s. A premixed burner designed for a flame speed of 40 cm/s could experience flashback with a hydrogen-enriched fuel with a flame speed of 52 cm/s.
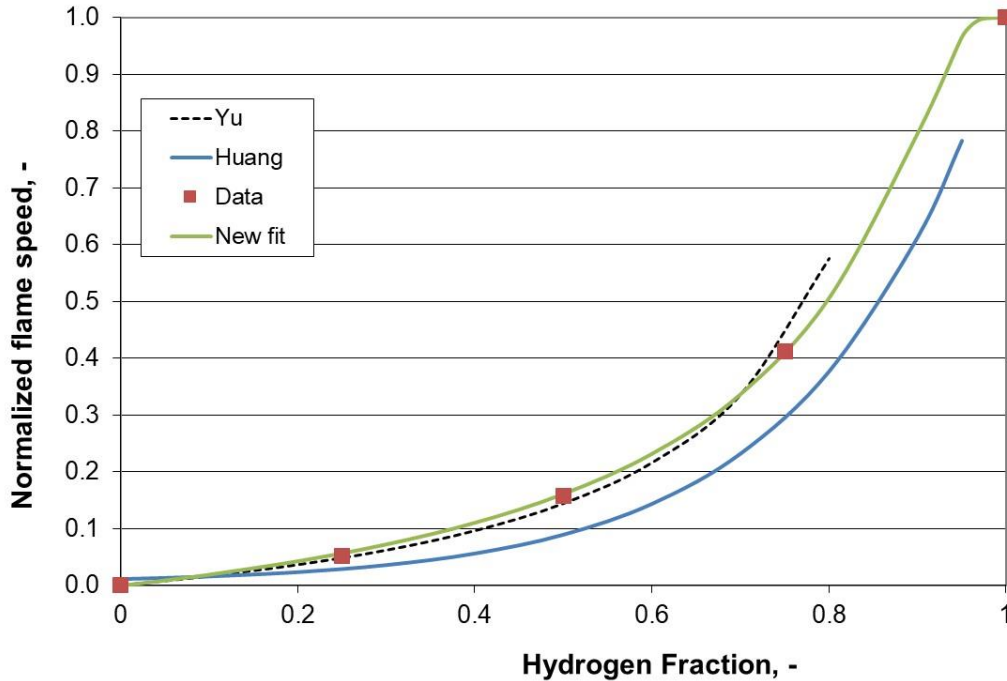

Figure 3 - Increase in laminar flame speed with increased hydrogen concentration.

The 'new fit' is

$$\frac{S_{mix} - S_{CH_4}}{S_{H_2} - S_{CH_4}} = 1 - \exp\left(-0.548R_H\right) \qquad (6)$$

[5] In an unpublished report on the combustion properties of industrial gases.

Jackson et al. (2003), provided an experimental and numerical study for an understanding of the effects of hydrogen addition to CH_4 on lean premixed flame extinction and response to stretch. They used a counter-flow flame configuration created by opposing jets of fully premixed fuel and air. The extinction strain rates were measured with a nozzle exit temperature of 300 °C over range of H_2 values of 0, 5, 10, and 20 vol-%. The equivalence ratios were ranging from 0.75 to less than 0.4. Their results indicate that increasing H_2 in the fuel significantly increases flame speeds and thus extinction strain rates increased. Also, increasing H_2 decreases the dependency of extinction equivalence ratio on the strain rate of the flow. For all of the mixtures tested in this paper, extinction temperatures depend primarily on equivalence ratio and not fuel composition for the range of H_2 content studied, which suggests that extinction can be correlated to flame temperature and O_2 concentration. H_2 addition greatly increases the maximum allowable strain rate before extinction temperatures are reached. Inspection of the model-predicted species profiles suggest that the enhancement of CH_4 burning rates with H_2 addition is driven by early H_2 breakdown increasing radical production rates early in the flame zone to enhance CH_4 ignition under conditions where otherwise CH_4 combustion might be prone to undergo extinction.

Zhao et al. (2008) used a flow tube reactor to study the effect on ignition and burn-off temperatures for hydrogen addition to methane. They defined ignition as the temperature at which 10% conversion of methane occurred and burn-off as the temperature when 90% conversion occurred. The authors concluded from their tests that hydrogen could decrease the ignition temperature and burn off temperature of methane evidently for a wide concentration range. The ignition temperature and burn off temperature decreased continuously with the increase of hydrogen. For 1% vol. methane combustion, the ignition and the burn off temperatures decreased 45 °C and 42 °C respectively when the H_2/CH_4 was equal to 0.05. The ignition and the burn off temperatures decreased about 170 °C and 180 °C when the H_2/CH_4

was 2.5. They reported that CO was generated in a wider temperature range when the hydrogen was added.

Hu et al. (2009) also performed an experimental and numerical study on laminar burning characteristics of the premixed methane–hydrogen–air flames. The tests were conducted at room temperature and atmospheric pressure. The unstretched laminar burning velocity and the Markstein length were obtained over a wide range of equivalence ratios and hydrogen fractions. The authors reported results indicated that the enhancement of chemical reaction with hydrogen addition is due to the increase of H, O and OH mole fractions in the flame as hydrogen is added. This causes the unstretched laminar burning velocity to increase. Also they found that the peak value of the unstretched laminar burning velocity shifts to the richer mixture side with the increase of hydrogen fraction. They identified three regimes determined by the hydrogen fraction: the methane-dominated combustion regime (H_2 < 60%); transition regime (60% ≤ H_2 ≤ 80%); and methane-inhibited hydrogen combustion regime (H_2 > 80%). They found that the laminar burning velocity increases linearly with the increase in H_2 fraction in the first and last mentioned regimes and increased exponentially in the transition regime.

Hawkes et al. (2004) conducted a direct numerical simulation to study the effects of H_2 enrichment (0% and 29% vol.) on lean premixed methane–air flame stability (from the perspective of the turbulent flame speed) and pollutant formation (emissions of CO and NO). The authors reported that the influence of adding H_2 into the flame results in an increase in the turbulent flame speed, flame surface area, and the burning rate per unit area. This leads to shorter flame time-scale. The reported results indicated that the above mentioned factors were found to lead to an enriched flame that is less thermal-diffusively stable and more resistant to extinction than the pure methane flame. Regarding the CO emission levels, lower CO emissions per unit methane consumption were found in the enriched flame. A 50% increase in NO production per unit heat release was reported for the enriched flame relative

to the pure methane flame, attributed to locally high temperatures and radical concentrations.

The influences of pressure and thermo-diffusive effects on spherically propagating premixed hydrogen–air turbulent flames were studied by Hayakawa et al. (2008) using a constant volume fan-stirred combustion vessel. They reported that the ratio of the turbulent to unstretched laminar burning velocity increased with decreasing equivalence ratio and increasing mixture pressure, in addition the turbulent burning velocity was dominated by turbulence Reynolds number (the ratio of turbulence intensity to unstretched laminar burning velocity), and the sensitivity of the flame to the stretch due to the thermo-diffusive effects.

Cocchi et al. (2005) set up a semi-empirical model for the prediction of emissions of a diffusion combustion system. The model has been tuned and validated with experimental data collected from a single can of a silo-type 10 MW_e gas turbine, fed with natural gas-H_2 blends. The authors assumed that NO_x is generated through the thermal route only, and therefore it was strongly dependent on flame temperature and on the residence time of the mixture at high temperature. They found that as the hydrogen concentration increased (from 0% to 100% hydrogen) the NO_x emission increased, the emissions at hydrogen operation are about three times higher than at methane operation.

Zhao et al. (2019) used these considerations of the combustion behaviour (flashback limit, ignition time, flame characteristics, combustion noise, emissions) to establish test procedures for domestic fired equipment. They applied the procedures to a cooktop burner and found that hydrogen enrichment up to 15 vol-% did not significantly affect the combustion performance. The flashback limit was 20 vol-% hydrogen addition. So for hydrogen addition above 15 vol-% the equipment has to be modified for safety.

The theoretical and experimental work done on fundamental combustion of the methane/hydrogen mixtures shows that flame temperature, flame speed

and flame stability all increase with increased hydrogen concentration. The mechanisms of methane combustion are dominant until the hydrogen concentration approaches 60%. The higher flame temperature and increased concentration of transient H, O and OH radicals lead to increased formation of NO even within the methane-dominated regime.

INDUSTRIAL APPLICATIONS

There has also been research into the implications of hydrogen addition for industrial combustion equipment, primarily related to effects on the efficiency of the combustion system and the emissions. These are the practical implications, the impact on the economic performance and the regulated pollutants.

Hoelzner et al. (1994) investigated the effect of hydrogen enrichment on emissions and efficiency at various boiler outputs (20, 15, 10, and 6 kW_{th}). Two boiler units were used and operated over the full range of hydrogen concentration with natural gas (0, 25, 50, 75, and 100 vol-% H_2). The emitted quantity of nitrogen oxides were represented by mg NO_2 per kWh of supplied firing energy as a function of fuel gas composition and thermal boiler output. At 6 and 10 kW_{th} the NOx emission level increases gradually with the H_2 addition. For the 15 and 20 kW_{th} the trend was different, NO_2 emissions increased until the H_2% reaches 50% then the NO_2 emission starts to decrease with the H_2 addition. The overall results showed that at the nominal capacity of 20 kW_{th} the NO_2 emissions are lower than 100 mg NO_2 kWh with all fuel gas mixtures and higher NO_2 emission levels were obtained at part load. They attributed the increase in NO_x formation to the high combustion temperature of hydrogen and that could be lowered by increasing in the excess air which could reduce the formation of nitrogen oxides.

Slim et al. (2006) discussed the effects of hydrogen addition on the behaviour of industrial burners. They tested two 150 kW burners, a conventional process-burner and a prototype flameless burner. Both burners were fired in a lightweight unloaded furnace; following a temperature ramp to furnace temperatures in excess of 1200 K. In the experiments they conducted the firing rates were kept constant to compensate for the decrease in heating value of the fuel by increasing the flow rate. CO, O_2, NO_x, CO_2 and methane measurements of the exhaust gases were conducted. Their results for the

conventional process burner indicated that the NO_x emission increased with H_2 addition of 20-50% due to the steady increase in flame temperature with H_2 addition. The CO emissions were reduced. The authors observed the flames being stabilised closer to the burner than when being fired with natural gas alone and also a very modest increase in burner head temperature (5 °C) was observed. For the modern prototype flameless burner, the hydrogen was added at a constant furnace temperature. The emissions reported were extremely low (3mg NO_2/m^3 at 3% O_2) up to 55% H_2. Above 55% H_2 concentration the NO_x emissions steadily increased and at this region a visible flame was present which was not visible at lower H_2%. Hydrogen addition above 55% completely changes the combustion mode of this burner, from flameless to close to conventional diffusion flame. These flames provide the local high temperature zones for formation of thermal NOx. Increasing the H_2% in the mixture reduces the CO emissions. Regarding the change of the combustion mode the authors suggested that the addition of hydrogen to natural gas should lower the temperature necessary to observe flameless behaviour and that would allow the use of flameless burners in a much wider range of processes.

In a similar study related to the industrial application, Joua et al. (2008) evaluated the benefits of partially replacing natural gas with the hydrogen-rich waste refinery gas recovered from the catalytic reforming and catalytic cracking operations. They used a high-pressure cogeneration boiler (280 tons/h boiler capacity), originally designed for fuel oil and natural gas in a full-scale petrochemical plant. The authors reported that the use of waste refinery gas to partially replace natural gas has practical benefits for reducing energy consumption and greenhouse gas emission. They show that adding hydrogen rich waste refinery gas to the fuel oil/ natural gas reduces 14,500,000 m^3 of natural gas, 12,900 tons/year of CO_2 emissions, and 9.1 ppmv or 4.3% per year of NO_x emission for a full scale plant operation.

Gogolek et al. (2010) studied the effects of hydrogen enrichment with a flameless burner in a pilot-scale furnace. They found no increase in NOx for hydrogen enrichment up to 10%.

Ayoub et al. (2012) used a laboratory-scale pilot furnace with a flameless burner to study the combustion of methane/hydrogen mixtures over the full range. Unlike Slim et al. (2006) above, they did not find flame formation at any hydrogen content. However, the burner in this study was 20 kW, much smaller than the 150 kW burners used by Slim et al. (2006). This underlines the importance of doing testing at the appropriate scale and that caution is needed when using small-scale experimental results in decisions with implications for full-scale industrial processes.

Khalil and Gupta (2013) also used a flameless burner in a much smaller test furnace with hydrogen enrichment from 25 vol-% to 58.5% vol-% and fuel-lean conditions. They found that hydrogen improved stability and lowered the CO emissions without a significant increase in NOx emissions.

Athens et al. (1995) tested two different ultra low NO_x burners; one is a small heat release forced draft burner and the other a large heat release round flame burner for the effect of hydrogen addition on the combustion process of CH_4/air mixtures. The authors reported that the NO_x emissions for these ultra low NO_x burners were reduced for mixtures with very high hydrogen concentrations. Since ultra low NOx burners have minimized the effect of the thermal NOx formation mechanism, this reduction was attributed to a reduction in the amount of NO_x generated by the prompt NO_x mechanism due to the reduction of the concentration of CH radicals for these mixtures.

The combustion of hydrogen in a hot, bubbling bed of quartz sand fluidized by air has been studied in a laboratory-size fluidized bed by Baron et al. (2009). The tests were performed by injecting hydrogen via six horizontal fine tubes of Cr/Ni. Different mixtures of H_2/O_2 were tested with the composition varying from nearly stoichiometric to very lean mixtures. With hydrogen as fuel the authors found that the ignition of hydrogen bubbles occurs at approximately

500 °C, which is 100 K below the ignition temperature predicted by well-established kinetic models. The amounts of NO found in the off-gases were at a maximum (24 ppm), when the bed was at 500 °C for $(O_2/O_2 stoich)$ equals 1.05. As the bed warmed up from 800 to 940 °C, NO concentration fell from 8 to 6 ppm. The authors explained that NO was produced partly via the intermediate N_2H. They also observed emission of green light indicating the presence of both NO and free atoms of oxygen for $1.05<(O_2/O_2 stoich)<1.1$.

Vetkin et al. (2015) use a theoretical analysis of radiative heat transfer in the context of a tube furnace in a petroleum refinery to determine the effect of hydrogen enrichment on the furnace performance and NO emissions. They find that the radiative transfer in the furnace would not be significantly changed up to 30% hydrogen content. Interestingly, they predict a maximum radiative transfer around 90% hydrogen content due to the combination of higher flame temperatures with the high hydrogen content and the presence of carbon species due to the remaining methane. NO emissions are found to increase with increasing hydrogen content.

Lo Basso et al. (2017) provide guidelines and tools to operators of boilers firing hydrogen enriched natural gas. This is a very practical article aimed at the people in the plant having to adjust operating procedures, an indication of this likelihood of short-term implementation of hydrogen enrichment in Europe.

A cautionary note is sounded in Leicher et al. (2017). They looked at the effect of hydrogen addition to natural gas in the context of power-to-gas and its potential impact on industrial combustion equipment. With power-to-gas, the hydrogen content of natural gas delivered to an industrial facility will fluctuate, within set limits. If it is equipped with the necessary instrumentation to monitor and control the operation with the hydrogen addition, then the negative effects on efficiency and NOx emissions can be minimized. However, systems without the necessary instrumentation will not be able to adjust to the changes and the impact will be negative even at the relatively low level of

10% hydrogen addition, which some jurisdictions in Europe are considering as the threshold level.

Swirl Stabilized Burners

Swirl burners are now standard industrial equipment. The swirl induced in the combustion air produces internal flue gas recirculation to preheat and stabilize difficult fuels or to control flame dilution thereby lower peak flame temperatures and reduce of thermal NO_x formation. The scaling of pollutant emissions in industrial flames is very difficult and the relation between the combustion characteristics of swirling flames and pollutant formation still needs to be established [Hsieh et al. (1998)]. Different researchers investigated the effect of enriching methane with hydrogen in a lean premixed swirl stabilized burners such as Schefer et al. (2002), Schefer (2003) and Kim et al. (2009) and on non-premixed swirl stabilized burners such as Cozzi et al. (2006).

Schefer et al. (2002) examined the effect of different amounts of hydrogen addition (0%, 20%, 40% and 45%) at several fuel-air flow rates (fuel-lean conditions) on the combustion characteristics of a premixed, swirl-stabilized flame at atmospheric pressure. The burner used consisted of a center-body with an annular, premixed fuel-air jet. Swirl was introduced to the flow using seven 45 º swirl vanes. Flame stability, blowout maps and emission levels (CO and NO_x) were obtained. In the study the flame structure was identified qualitatively by luminous photographs and quantitatively by OH planar laser-induced fluorescence (PLIF) measurements. The authors reported that the addition of a moderate amount of hydrogen to the methane/air mixture increased the peak OH concentration and a significant change in the flame structure. Their results also indicated that hydrogen addition increased the strain resistance of the flame as well as the OH levels in the flame. As they reported that the improved stability with hydrogen enrichment was a result of the higher OH, H, and O radical concentrations, which increase several key reaction rates. A significant reduction in CO emissions was reported by hydrogen addition as the lean stability limit of natural gas was approached.

The NO_x emissions for an equilibrium adiabatic flame temperature were not affected by the hydrogen addition.

Schefer (2003) investigated the effects of hydrogen addition (0, 10, and 20 vol-%) on the flame stability limits, blowout velocity and the behaviour of OH mole fraction of a premixed, swirl-stabilized flame over a range of operating (fuel-lean) conditions. The burner used in this investigation has a different configuration than that used in Schefer et. al. (2002), it consisted of a center-body with an annular, unconfined premixed methane/air jet introduced through five, 45 º swirl vanes. The author results indicated that under fuel-rich conditions the flame was lifted from the burner surface due to the mixing with entrained ambient air that was needed to form a flammable mixture and as the fuel/air mixture ratio was decreased toward stoichiometric, the resulting increase in flame speed allowed the flame to propagate upstream through the low-velocity wake region and attach to the center-body face. In addition, the maximum blowout velocity occurred at stoichiometric conditions, and decreased as the mixture became leaner. Near the lean stability limit the overall OH mole fraction decreased and the flame decreased in size. The addition of up to 20% hydrogen to the methane/air mixture resulted in a significant increase in the OH concentration and extended the lean stability limits of the burner. This reduction in the lean stability limit allows stable burner operation at the lower flame temperatures which allows a reduction in NO_x emissions.

Kim et al. (2009) examined the effect of hydrogen addition on methane–air lean-premixed flames for a laboratory-scale unconfined swirl-stabilized premixed combustor operated at 5.8 kW. The authors investigated the combustion characteristics of hydrogen-enriched methane–air flames at fixed thermal load but different swirl strengths (30 º, 40 º and 60 º) using particle image velocimetry (PIV), gas analyzers (OH radicals and flue gas concentration) and micro-thermocouple diagnostics to provide information on flow field. The experimental matrix was: H_2/CH_4 concentration of 0, 4, and 9 (wt %); with an equivalence ratio of 0.717, 0.706 and 0.694. Their results

show that the higher combustibility of hydrogen makes the reaction faster, raises the temperature in the reaction zone and reduces the recirculation flow in the reaction zone. They reported that the upstream of flame region is more dependent on the swirl strength than on hydrogen addition to methane fuel. At lower swirl strength condition the NO concentration in the reaction zone reduces with increase in hydrogen content in the fuel mixture, which is due to the faster reaction of hydrogen which accelerates the flow and reduces the residence time. At higher swirl strength their results showed an increase in NO concentration with an increase in percentage of hydrogen in the fuel mixture.

Cozzi et al. (2006) also investigated the effect of H_2 addition (0% up to 100%) on the structure and stability of a lean swirled non-premixed natural gas flame at atmospheric pressure in a confined environment. The authors concluded that by increasing the hydrogen content flame stability increases, the visible flame size decreases, fuel jet penetration increases, CO and NO_x emissions (for 0% up to 80% H_2 fractions) increase and there is a large increase in soot formation (from the observation of central highly luminous yellow plume extending above the visible blue zone).

Porous Radiant Burners

Porous radiant burners has the combustion occurring in the pores of the solid burner face and the heat is transferred primarily by the thermal radation from the surface. The distributed combustion and heating of the porous medium lowers the flame temperature so they are classified as ultra low NO_x burners.

Gauthier et al. (2007) experimentally tested premixed combustion of natural gas-hydrogen mixtures in a porous burner made of open cell metallic foam. Their results showed that H_2/CH_4/air mixture fuel combustion in a porous burner reduces the pollutant emissions of CO and NO_x and CO_2. Importantly, they measured the flashback limit burning velocity (velocity of the gases at the inlet of the burner when flashback occurred) for the different hydrogen concentrations (0, 20, 50, 100 vol-%). They reported that the addition of H_2 from 0% to 20% has small effect on the behaviour of the burner. Above the 20% H_2, the equivalence ratio has to be lowered to reduce the flame speed to operate within the stability limits.

Tseng (2002) investigated numerically the effect of hydrogen addition on premixed combustion of methane in porous medium burners. GRI-Mech 2.11 was used for the detailed chemical kinetic model. As a result the author reported that adding hydrogen in the fuel, the lean limit can be reduced to a lean equivalence ratio of $\Phi = 0.26$, from 0.33 for pure methane. Regarding the flame speed, the author reported that increasing the hydrogen fraction in the fuel increases the flame speed and reduces the flame thickness. CO and NO_x emissions were increased with the addition of hydrogen.

GAS TURBINE APPLICATIONS

Various studies have examined the feasibility of using hydrogen-enriched natural gas as a method for achieving NO_x reduction in gas turbines. As noted in the introduction, this application was perhaps the earliest advocacy for enriching natural gas with hydrogen. The stability characteristics of a prototype premixed, burner was studied by Schefer (2002). In his study the flame stability was characterized by varying amounts of H_2 addition (70, 80, 90 and 100 vol-%) and only the fuel lean conditions were studied. The author reported that the increase in hydrogen content from 70% to 100% results in a significant shift in flame blowout conditions to leaner fuel/air ratios. As an example, from the author report that at 60 m/s velocity increasing H_2 concentration from 70% to 100% reduces the equivalence ratio at flame blowout from 0.40 to 0.12.

When operating the gas turbine combustor under very fuel lean conditions to reduce emissions two main important points should be considered: flame stability (flashback) and the thermo-acoustic (e.g. pressure oscillations) instability. Tuncer et al. (2009) conducted laboratory-scale premixed tests to study these modes of instability. The hydrogen-enriched confined methane combustion was used and the lean blowout, pressure amplitudes and emissions were examined. The authors reported that taking advantage of the lower lean blow-off associated with hydrogen and the combustor can be operated under very lean conditions with low flame temperature and thus favourably impact thermal NOx emissions. They reported that hydrogen enrichment can cause an abrupt change in the dominant acoustic mode and at higher hydrogen concentrations, flashback is observed and appears to trigger a shift in the pressure oscillation mode to lower frequencies. Also they concluded that hydrogen enrichment shifts the flame center of mass more towards the dump plane as the burning velocity is increased with hydrogen addition.

A novel NOx control technology for premixed combustion of hydrogen enriched natural gas is studied in Goke et al. (2013). The Rich-Quench-Lean combustor starts combustion in a fuel rich zone, then it is quenched with the addition of air and steam, and finally combustion is completed with the addition of air to reach fuel lean conditions. Hydrogen, natural gas and hydrogen at 10 vol-% and 50 vol-% with natural gas was used as fuel. Steam addition was able to reduce NOx emissions and above 20% steam the NOx emissions were below 10 ppm. However, above 20% steam there is an increase in the CO emissions.

Flameless oxidation combustion technology has been used in many industrial furnaces under atmospheric conditions. This technique was known by its very low NO_x emissions. Lückerath et al. (2008) investigated the combustion behaviour of the FLOX burner at high pressure (20 bars with 475 kW thermal powers) in order to assess its applicability for gas turbine combustors. In their study, the flame zone (size & structure), lean extinction limits, mixing process and emissions (NO_x and CO) were investigated as a function of equivalence ratio and fuel composition. Among the fuels tested were mixtures of natural gas and H_2. With admixtures of H_2 to the natural gas, the authors reported that the range of stable operation could be extended but the NOx emissions increased due to inhomogeneities in the temperature distribution inside the burner. The lower CO emissions were recorded in their tests.

Imteyaz et al. (2018) constructed a stability map for gas turbine combustors firing hydrogen enriched natural gas with and without oxygen enrichment of the air. The addition of hydrogen expands the operability between flashback and blowoff. However, the model combustor is small, only 5 cm diameter, so these results need to be verified at a larger scale.

INTERNAL COMBUSTION ENGINE APPLICATIONS

Several studies have been reported on the performance and emission characteristics of the internal combustion engines using hydrogen-natural gas fuel blends. Karim et al. (1996) reviewed the effects of adding hydrogen with methane on the main combustion characteristics of the fuel for engine application. The authors claimed that hydrogen as an additive in natural gas can strongly improve the performance of such engines, especially in terms of power, efficiency and emissions allowing the engine to work with leaner mixtures. In addition, hydrogen does not affect the anti-knocking performance of NG fuel. The fuel mixture (H_2/CH_4+H_2) of 0, 20, 30, 50, 70, and 80 volume % were used in the study.

Wall (2007) studied the effect of H_2 (0, 10, 20 vol-%) addition on natural gas in an internal combustion engine. The results showed a decrease in the emissions of NO_x, CO and unburned hydrocarbons with an increase of the H_2 concentration. The hydrocarbon fuel consumption was reduced by adding as low as 5 to 10% due to extended lean operation limit.

Pechlivanoglou (2007) presented a summary of the key points on the hydrogen-enhanced combustion investigation and experimentation and discusses the different issues regarding this technique in Otto and Diesel engines. In both types of engines the author reported that the addition of hydrogen to the injected fuel leads to more rapid combustion, improves the lean flammability limits and reduces emissions. Rapid and efficient combustion leads to lower fuel consumption and high overall engine efficiency.

Ma et al. (2007) analyzed the effect of hydrogen addition (10, 30, 50 vol-%) on a natural gas engine's thermal efficiency and emissions. An experimental investigation was conducted on a spark ignition natural gas engine using variable composition hydrogen. The results showed that hydrogen enrichment could significantly extend the lean operation limit, improve the

engine's lean burn ability, and decrease burn duration. NO_x was found to increase with hydrogen addition if spark timing was not optimized for hydrogen's high burn rate.

A numerical investigation on the characteristics of natural gas–hydrogen blends and their effect on engine performance were carried out by Morrone et al. (2009). The mixture fuel tested was natural gas–hydrogen blend from 0% to 30%. Their results showed that there is an increase in engine efficiency only if maximum brake torque spark advance is used for each fuel. A follow-up article [Mariani et al. (2012)] used the same levels of hydrogen enrichment but also studied the effect of exhaust gas recirculation (EGR) to reduce NOx emissions. The presence of hydrogen improves the stability of the combustion with EGR, which would not work with natural gas alone. EGR at 10% of the gas input produced an 80% reduction of NOx emissions for NG alone. Since these are only numerical studies, it should be weighted lighter than the experimental work described here.

Reyes et al. (2013) studied the cycle-to-cycle variation of a single cylinder test unit firing the full range of hydrogen enrichment. They found no difference for 0% to 80% hydrogen content, only for pure hydrogen.

Hora and Agarwal (2015) looked at hydrogen enrichment up to 30 vol-% hydrogen as a means of improving the performance of engines firing compressed natural gas. There was an improvement with hydrogen content in various measures of performance (in-cylinder pressure, rate of pressure rise, heat release rate, combustion duration). However, there was an increase in NO emissions.

Similar work was done with a dual-fuel diesel engine by Ouchikh et al. (2019). The hydrogen was added up to 30 vol-%. Improvements were found as in Hora and Agarwal (2015), but the greatest thermal efficiency was found at 10 vol-% hydrogen. Sandalci et al. (2019) used 'hythane' (methane with 10 vol-% hydrogen) co-fired in a diesel engine at 30% and 50% of fuel input. The results

were generally negative, with emissions of gaseous pollutants increased and only a 5% reduction in particulate emissions.

Klell et al. (2012) outlined various benefits for production, storage, distribution as well combustion from the mixing of hydrogen with natural gas for internal combustion engines and transportation. They address legislative changes needed as well as standards, regulations and infrastructure.

Mehra et al. (2017) provide a comprehensive review of the use of HCNG (hydrogen enriched compressed natural gas) in internal combustion engines.

CONCLUSION

The addition of hydrogen into the natural gas network is being considered as a practical way to reduce GHG emissions from combustion equipment. While the introduction of renewable natural gas will require no changes from the end user, there are potentially serious changes to combustion properties from hydrogen addition that would require investments by the end-user. This report has looked at those combustion properties and surveyed the scientific literature investigating the behaviour of hydrogen enriched natural gas in industrial combustion equipment.

There are three combustion properties that increase significantly with the addition of hydrogen: flame speed, upper flammability limit, and flame temperature. This enhanced combustion has been used to improve the performance of gas turbines and internal combustion engines. Generally, addition of hydrogen increases the emissions of NOx because the higher flame temperature increases the thermal NOx formation mechanism. However, gas turbines have used hydrogen to stabilize lean combustion that minimizes NOx and ultra-low NOx industrial burners have lower NOx emissions because the prompt NOx mechanism is reduced.

Flashback is a safety concern for using unmodified equipment with hydrogen enriched natural gas. Some porous burners had flashback for hydrogen content above 20 vol-%, which could be controlled by adjusting flow rate and air supply.

Finally, one has to consider the actual scenario for hydrogen addition to the natural gas network. For example, Power-to-Gas would have excess renewable electricity used for electrolysis of water to produce hydrogen that would be injected into the natural gas network. The hydrogen concentration delivered to an industrial client would vary with the sun and wind, of course with a threshold. These variations can be tracked and process controls adjusted to maintain efficient and safe operation and control emissions of regulated

pollutants (primarily NOx). However, an industrial facility may not have the necessary instrumentation to measure the composition of the fuel gas in real time, or a sophisticated control system to make the necessary adjustments. Such users can experience degraded performance and increased emissions even with the maximum hydrogen threshold set at 10 vol-%.

Hydrogen addition to natural gas does produce many benefits, especially that of reducing the emission of GHGs from industrial combustion processes. However, it can also produce negative effects for existing industrial facilities if implemented without proper consideration of the actual end-use.

RECOMMENDATION: To realize the benefits of enriching natural gas with hydrogen, industrial combustion facilities need to be properly instrumented, the control systems modified, and the operators trained to take full advantage of the new fuel. This is particularly important for Power-to-Gas scenarios that introduce significant fluctuations in the fuel gas properties.

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NOMENCLATURE

FL_i	Flammability limit of a compound, either upper or lower (vol-%).
FL_{mix}	Flammability limit of a mixture, either upper or lower (vol-%).
$\dot{m}_f$	Mass flow of fuel gas (kg/s).
η	Combustion efficiency (%).
n_i	Number of moles of product from complete combustion of one mole of fuel at stoichiometric conditions (mol/mol).
$\dot{Q}$	Heat release rate (MW).
Re	Reynold's number (-).
S	Laminar flame speed (m/s).
S_{mix}	Mixture laminar flame speed or burning rate (m/s).
T_{ad}	Adiabatic Flame Temperature (K).
v_f	Kinematic viscosity of fuel gas (m^2/s).
ρ_a	Density of air (kg/m^3).
ρ_f	Density of fuel gas (kg/m^3).
ρ_p	Density of burnt fuel products (kg/m^3).
w_i	Weight fraction of species i in gas mixture (-).
x_i	Mole or volume fraction of species i in gas mixture (-).
LHV$_m$	Lower Heating Value, mass basis (MJ/kg).
LHV$_v$	Lower Heating Value, volume basis (MJ/Nm^3).
LFL	Lower Flammability Limit (vol-%).
SA$_m$	Stoichiometric air-to-fuel ratio, mass basis (kg/kg).
SA$_v$	Stoichiometric air-to-fuel ratio, volume basis (m^3/m^3).
UFL	Upper Flammability Limit (vol-%).

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