# SHOULD WE ADD HYDROGEN TO THE NATURAL GAS GRID TO REDUCE CO<sub>2</sub>-EMISSIONS? (CONSEQUENCES FOR GAS UTILIZATION EQUIPMENT)



Main author

B.K. Slim
N.V. Nederlandse Gasunie
Gasunie Engineering & Technology
Groningen, The Netherlands

#### **Co-authors**

H. Darmeveil, G.H.J. van Dijk, D. Last, G.T. Pieters and M.H. Rotink,
N.V. Nederlandse Gasunie
Gasunie Engineering & Technology
Groningen, The Netherlands

J.J. Overdiep
Gasunie Trade & Supply
Groningen, The Netherlands

H.B. Levinsky
N.V. Nederlandse Gasunie
Gasunie Engineering & Technology, and
Laboratory for Fuel and Combustion Science, University of Groningen
Groningen, The Netherlands

## **ABSTRACT**

The current drive to conserve the finite supplies of fossil fuels, and particularly to limit the emissions of carbon dioxide within the framework of the Kyoto agreement regarding greenhouse gases, has led to renewed interest in the use of sustainable fuels. However, it will be clear that the admixture of hydrogen in the natural gas grid may have no adverse effects: for example, hydrogen embrittlement in steel pipelines and deteriorated performance of gas utilization equipment, caused by altered combustion properties, are potential "show-stoppers" for this idea.

In this paper, we consider the effects of hydrogen addition on the behaviour of industrial and domestic burners, spark-ignited gas engines and gas turbines.

We observe that hydrogen addition to natural gas tends to decrease the emissions of CO and unburned hydrocarbons from a standard industrial nozzle-mix gas burner, but increases the  $NO_x$  emissions. Further, a flameless burner can be driven out of the flameless mode by high percentages of  $H_2$ , with a concomitant increase in  $NO_x$  emissions. Whereas hydrogen addition can facilitate the flameless combustion of pure methane, it has little effect on natural gas, presumably due to the presence of easily ignitable higher hydrocarbons. In addition, using a methodology for analyzing the changes in combustion properties of natural gas in domestic appliances, it is shown that a maximum of only 5%  $H_2$  may be present in the Dutch natural gas to guarantee safe performance in domestic appliances, provided the Wobbe number is maintained at the maximum of the distribution band. Allowing the Wobbe number to decrease upon hydrogen addition, the acceptable hydrogen fraction becomes even lower. Finally, we discuss briefly the effects of  $H_2$  addition on the performance of gas engines and turbines.

# **TABLE OF CONTENTS**

#### **ABSTRACT**

- 1. INTRODUCTION
- 2. CO<sub>2</sub> REDUCTION
- 3. CHANGES IN COMBUSTION PROPERTIES
- 3.1. Gross Calorific Value
- 3.2. Wobbe number
- 3.3. Stoichiometric air Requirement (SAR)
- 3.4. Ignition properties (Knock propensity)
- 3.5. Laminar Burning Velocity
- 4. RESPONSE OF DOMESTIC APPLIANCES (to H<sub>2</sub> addition)
- 4.1. Hydrogen, Wobbe number and equivalence ratio φ
- 4.1a. Changing Wobbe number due to hydrogen addition
- 4.1b. Constant Wobbe number
- 4.2. Comparing gases with gases
- 4.2a. Lean-premixed surface burners
- 4.2b. Flashback in an existing rich-premixed appliance population
- 5. RESPONSE OF INDUSTRIAL BURNERS (to H<sub>2</sub> addition)
- 5.1. Conventional process-burner
- 5.2. Modern prototype flameless burner
- 6. RESPONSE OF SPARK-IGNITION PISTON ENGINES (to H<sub>2</sub> addition)
- 6.1. Laminar burning velocity
- 6.2. Ignition Properties (Ignition delay time / Knock propensity)
- 7. RESPONSE OF GAS TURBINES (to H<sub>2</sub> addition)
- 8. CONCLUSIONS

Acknowledgement

**REFERENCES** 

#### 1. INTRODUCTION

The current drive to conserve the finite supplies of fossil fuels, and particularly to limit the emissions of carbon dioxide within the framework of the Kyoto agreement regarding greenhouse gases, has led to renewed interest in the use of sustainable energy. Hydrogen can play an important role in a sustainable energy supply, since the utilization of H<sub>2</sub> yields no CO<sub>2</sub>, and H<sub>2</sub> can be produced from renewable sources such as solar and wind energy by the electrolysis of water or through biomass gasification. One tantalizing possibility for the gradual introduction of sustainably derived hydrogen into the energy infrastructure is to mix hydrogen with natural gas; in this fashion, the existing natural gas infrastructure can be used to distribute hydrogen to the end user. This would limit investment in costly pipelines, and natural gas could be used as a "buffer" for the varying amounts of sustainable hydrogen produced from solar/wind energy, eliminating the necessity of storage facilities. However, it will be clear that the addition of hydrogen to the natural gas may have no adverse effects: for example, hydrogen embrittlement in steel pipelines and deteriorated performance of gas utilization equipment, caused by altered combustion properties, are potential "show-stoppers" for this idea.

In this paper, we consider the effects of hydrogen addition on the behaviour of industrial and domestic burners, spark-ignited gas engines and gas turbines.

# 2. CO<sub>2</sub> REDUCTION

Since one reason for adding hydrogen to natural gas is to reduce the emissions of  $CO_2$ , it is perhaps most interesting to consider how large a reduction can be achieved. Towards this end, we perform a very simple calculation, i.e., we calculate the mass of  $CO_2$  emitted per unit of energy in the fuel. Since heating processes require a given thermal input, the reduced heating value of the fuel caused by the addition of hydrogen (the heating value of  $H_2$  is roughly a third of that of natural gas, see below), must be compensated by using more fuel. Naively, one would argue that replacing 10% of a natural gas by hydrogen would reduce the  $CO_2$  emissions by 10%. The results show however that per unit of thermal input this is simply not true: replacing 10% of natural gas by hydrogen reduces the  $CO_2$  emission by only 3%! With larger hydrogen addition, the  $CO_2$  reduction increases to 25% upon addition of 50% hydrogen. Thus, for moderate fractions of added hydrogen the  $CO_2$  reduction amounts, at best, to only half the percentage of hydrogen. This effect must be weighed against the other consequences of hydrogen addition summarized below.

# 3. CHANGES IN COMBUSTION PROPERTIES

#### 3.1. Gross Calorific Value

The calorific value of pure  $H_2$  (~ 13MJ/m³, gross, standard conditions), is only about one third of the calorific value of common natural gases, 35-40MJ/m³. One must burn three times more hydrogen (by volume) to get the same thermal input. Thus, the calorific value of natural gas decreases with  $H_2$  addition.

#### 3.2. Wobbe number

The thermal input at a given pressure drop across a fuel nozzle depends on the Wobbe number only. The Wobbe number is defined as the gross calorific value divided by the square root of the relative density. Since this primary combustion airflow is proportional to the momentum flux through the nozzle, this airflow also depends on the pressure drop of the fuel nozzle only, regardless of the gas composition, Due to the very low density of hydrogen (only  $0.0825 \text{kg/m}^3$ ) the Wobbe number of hydrogen is  $48 \text{MJ/m}^3(\text{n})$ , in the neighborhood m' of that of most natural gases. When simply adding  $H_2$  to natural gas, the Wobbe number will decrease monotonically until ~ 70%  $H_2$ , reflecting the fact that in this range the calorific value of the mixture decreases more rapidly than the square root of the density. At high hydrogen fraction, the Wobbe number again begins to increase, ultimately to the value of pure hydrogen.

#### 3.3. Stoichiometric Air Requirement (SAR)

The amount of air required for stoichiometric combustion is 25% of that of methane; one mole of hydrogen requires half a mole of oxygen for stoichiometric combustion, whereas one mole of methane, the most abundant molecule of natural gas, requires two moles of oxygen. This difference in the stoichiometric air requirement results in a higher adiabatic flame temperature of hydrogen compared to methane: 2382K versus 2226K, respectively. This is an important aspect when considering possible changes in  $NO_x$  formation due to hydrogen addition.

## 3.4. Ignition Properties (Knock propensity)

Hydrogen and natural gas have significantly different ignition properties; hydrogen spontaneously ignites much easier than methane. Spontaneous ignition can be related to the fundamental property called "ignition delay time", i.e. the time it takes for a homogeneous mixture to ignite and form a fully developed flame. This property is related to engine knock, and it is for this reason that the so-called methane number for describing the knock propensity of fuel gases in natural gas engines uses pure hydrogen as "worst" knock fuel against which other gases are compared. Although the methane number is often used to consider knock propensity, the method used to calculate it shows a *decrease* in knock tendency with addition of small quantities of hydrogen in natural gas (1).

#### 3.5. Laminar Burning Velocity

The laminar burning velocity is the velocity with which a one-dimensional flame front propagates with respect to the unburned gas. The laminar burning velocity of a stoichiometric pure hydrogen-air flame is roughly 6 times higher than the equivalent methane-air flame; so, the addition of  $H_2$  increases the burning velocity of natural gas. The burning velocity increases roughly linearly up to 30% of  $H_2$  in the mixture, at 1% per 1% $H_2$  addition. The burning velocity is closely related to the flame stability when compared tot 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. Occurrence of these phenomena in practice can have dire consequences.

# 4. RESPONSE OF DOMESTIC APPLIANCES (to H<sub>2</sub> addition)

## 4.1. Hydrogen, Wobbe number, equivalence ratio φ and burning velocity

In order to understand the behaviour of domestic gas appliances when hydrogen is added to natural gas, the changes in Wobbe number and equivalence ratio  $\phi$  have to be considered.

## 4.1a. Changing Wobbe number due to hydrogen addition

First, the Wobbe number of the mixture changes in the way mentioned above. This results in a decrease in the thermal input to the appliance. Second, the primary equivalence ratio  $\phi$  (the reciprocal of the air-factor  $\lambda$ ) decreases with increasing hydrogen content, provided the volumetric flow rate of primary combustion air remains constant. The airflow is constant in both naturally aspirated appliances, as mentioned above, and in fan-assisted appliances, since one generally does not change the fan speed when changing gases. Thus, due to the decrease in stoichiometric air requirement, the fuel-air mixture becomes leaner with increasing  $H_2$  addition.

The change in burning velocity arising from  $H_2$  addition under these conditions exhibits two different behaviors, depending on whether the mixture is fuel-rich or fuel-lean. We note here that the burning velocity increases as  $\phi$  moves closer to the stoichiometric value  $\phi = 1(\lambda = 1)$ . When hydrogen is added to a fuel-lean mixture , $\phi < 1$ , this mixture becomes leaner still. In that case the leaner mixture offsets the effect of the faster-burning  $H_2$  on the burning velocity. Depending on the amount of hydrogen added, this offset can be so large that no change of the burning velocity is observed. In contrast, when  $\phi > 1$ , the fuel-rich mixture moves towards  $\phi = 1$ , and thus the effect of increasing burning velocity due to the fast-burning hydrogen is amplified by the effect of decreasing  $\phi$ .

## 4.1b. Constant Wobbe number

When the Wobbe number of the mixture is held constant, for example, by addition of another gas (e.g. ethane) to compensate for the effect of hydrogen, both the thermal input and the equivalence ratio remain essentially unchanged. In this scenario, the burning velocity of the mixture increases upon addition of hydrogen, due to the faster burning rate of hydrogen compared to natural gas.

In the vast majority of domestic appliances the fuel-air mixture is fuel-rich,  $\phi > 1$ , across the whole distribution band. As will be shown below, it is advantageous to keep the Wobbe number of a hydrogen-containing mixture constant and equal to the upper Wobbe number of the distribution band. In that case, the increase of the burning velocity is solely due to addition of a faster-burning fuel and the acceptable percentage of hydrogen is maximized.

#### 4.2. Comparing gases with gases

The question now arises as to how serious the changes in fuel-gas composition are for appliance behavior. To assess those changes, there are two possibilities:

- 1 one can test every appliance installed in the area to receive hydrogen-containing gases (which is extremely impractical due to the large number of appliances concerned) or test a subset of appliances (which raises questions about how representative the sample is);
- 2 one can compare the burning properties of the distributed fuel gases with those of the test gases. If the hydrogen-containing fuel gas has a lower burning velocity than the fastest burning test gas for which the performance of the appliance is guaranteed, one assumes that flashback does not occur.

In Europe, there is a series of standard test gases, bracketing the actual distribution limits, based on Wobbe number. The relevant test gases in the Netherlands (for the "L-group" of gases, whose "adjustment" gas, referred to as G-25, has  $W = 43.7 \text{ MJ/m}^3$ ) and a test gas for flashback or overheating of the burner (also G-25). Thus, for appliances whose performance with these gases is guaranteed, we can compare the changes in relevant combustion characteristics caused by hydrogen addition with those arising from the use of the test gases. Let us consider two examples.

#### 4.2a. Lean-premixed surface burners

Many modern appliances (and industrial equipment as well) are equipped with lean-premixed burners, particularly in regions having stringent  $NO_x$  regulations. These appliances are either fit with radiant surface burners or low-pressure-drop metal burners. An increase in burning velocity for both these types of burner can lead to overheating of the burner, and ultimately to failure. As discussed above, hydrogen addition allowing a decrease in Wobbe number for fuel-lean flames yields only a modest change in burning velocity, which we shall consider unimportant here (of course, how important it is depends on the circumstances and the method described here can be used to calculate the effects). Regarding the situation of hydrogen addition at constant Wobbe number, the burning velocity will increase with hydrogen content, without the benefit of compensation by a leaner mixture. The potential consequences for the burner temperature are shown in Figure 1.

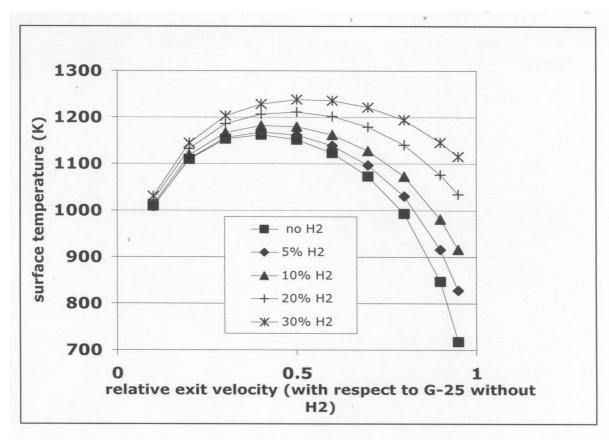


Figure 1: Surface temperature of the burner as a function of exit velocity, with varying hydrogen content in the fuel.

Qualitatively, the increase in burning velocity can cause increased heat transfer from the flame to the burner. For a radiant surface burner, in the radiant mode, this increased heat transfer can exceed the acceptable limit. In Figure 1 we plot the surface temperature of a burner as a function of exit velocity of the fuel-air mixture, calculated using the method for radiant surface combustion presented in (2). The velocity used to scale the horizontal axis was the exit velocity at which the burner surface is at room temperature (no heat transfer to the burner) using G-25. In the calculations, we have taken the 1% burning velocity per 1% hydrogen addition to be adequate (this can be performed more accurately if need be). The points corresponding to "no H<sub>2</sub>" are from G-25, and the other points consider hydrogen addition at constant equivalence ratio, which would be the situation for constant Wobbe number. Here we see that the burner temperature at the maximum of the curve for "no H<sub>2</sub>" (relative velocity = 0.4) increases by only 60 K upon increasing the hydrogen content to 30%. The largest difference is at higher exit velocity, since at high velocity the natural gas flame gives little heat to the burner surface (the burning velocity is only slightly higher than the exit velocity), while 30% hydrogen addition has increased the burning velocity by 30%, significantly increasing the heat transfer. Measurements of burner temperature show excellent agreement with the results given here. Be that as it may, the real question is whether this increase in burner temperature is acceptable.

A burner in the L-band is only tested for flashback and overheating using G-25. Thus, formally, no increase in burner temperature above that for G-25 is guaranteed by the approval regime. Interestingly, the burner is also tested with G-26. Officially, this test is performed at high thermal input to assess incomplete combustion. However, we note that the equivalence ratio will change by ~8% upon switching from G-25 to G-26. If the burner had been adjusted at  $\varphi = 0.8$  for G-25,  $\varphi$  will increase to more than 0.85 when switching gases. This will increase the burning velocity by more than 20%, and will have the same effect on burner temperature as an equivalent amount of hydrogen at W = 43.7 MJ/m³. Accordingly, provided the burner was adjusted at the G-25 exit velocity in Figure 2, the temperature increase caused by hydrogen up to ~ 20% should have the same consequences for burner performance as G-26. However, the test with G-26 is only performed briefly, and at high thermal input; to guarantee burner performance, this test must be performed at part load (such as at relative velocity 0.4 in Figure 1). Since the burners at part load are only tested with G-25, the burner

temperature may not exceed the value obtained with this gas. Therefore, the approval regime will not accept any H<sub>2</sub> addition.

We see that we can do an adequate job of determining the acceptable amount of hydrogen without having to do any testing, provided testing regime is appropriate for the phenomena being tested. In this case the test gas for flashback should be reconsidered.

#### 4.2b. Flashback in an existing rich-premixed appliance population

Let us now consider a population of appliances for which we are not sure whether the entire population can perform properly with the test gases we just described. This is de facto the situation in many European countries; the test gases defined above have only been legally in place since the beginning of the 1990's (3), and many appliances dating from before this time are still in use. Furthermore, without discussing the adequacy of the test gases for specifying the acceptable range of distribution gases, just because an appliance could briefly accommodate a wide range of gases when it was new, does not mean it can still do so when it is 10 years old. This will depend among other things upon the degree of maintenance. So, taking the Netherlands as an example, the only reasonable assumption is that the appliance population can accept the range of gases currently being distributed. In the Netherlands Wobbe numbers 43.4 MJ/m³ – 44.4 MJ/m³. When considering flashback in rich-premixed flames (being representative for most domestic burners), we take the lowest Wobbe number as the gas for comparison; this gas will result in the equivalence ratio closest to stoichiometric, and thus having the highest burning velocity.

We have varied the hydrogen content at Wobbe numbers between 44.0 and 44.4 MJ/m³. To avoid the complications of practical burners (difficulty in varying equivalence ratio, finding a burner that flashes back easily, etc.), we have considered flashback in simple Bunsen tubes, and recall that many of the American standards for gas interchangeability (4) have been determined using them. Experimentally, we determined the (average) exit velocity of the fuel-air mixture at which the flame just began to enter the tube (incipient flashback) for the "limit" gas at W = 43.4 MJ/m³, at two equivalence ratios,  $\varphi$  = 1.2 and 1.25 that are typical for practical appliances that are susceptible to flashback. We then performed the same measurements for the hydrogen-containing gases, accounting for the change in equivalence ratio experienced if these gases had been supplied to practical appliances. The results are shown in Figure 2.

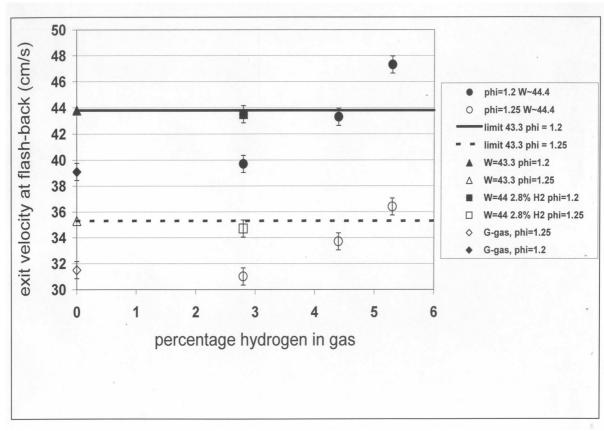


Figure 2: Experimentally determined exit velocity at flashback as a function of the hydrogen percentage in natural gas.

In this figure, the higher the exit velocity at flashback, the easier the gas will flashback. We see that at W =  $44.4~\text{MJ/m}^3$  the velocity at flashback for the hydrogen-containing gas equals that of the "limit" gas at about 5% H<sub>2</sub> in the gas. Thus, we must conclude that we cannot guarantee the performance of an appliance at higher percentages, at W =  $44.4~\text{MJ/m}^3$ . What we also see in the figure is that at W =  $44.0~\text{MJ/m}^3$  the H<sub>2</sub>-containing gas has equal flashback propensity as the limit gas at  $2.8\%~\text{H}_2$ ! At lower Wobbe number, even less hydrogen may be added to the gas (caused by the change in equivalence ratio at lower Wobbe). The maximum H<sub>2</sub> percentage as a factor of Wobbe index, derived from experimental data, is shown in Figure 3.

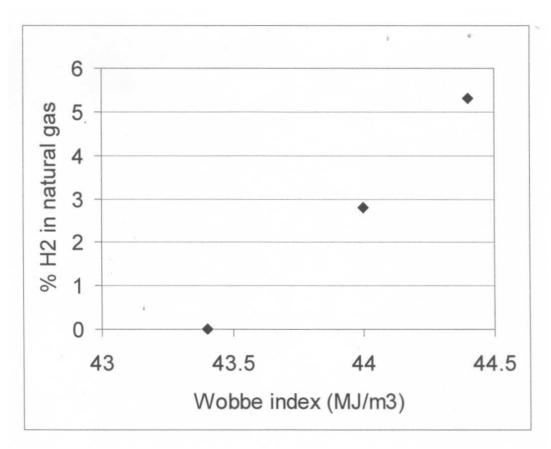


Figure 3: Experimentally determined maximum H<sub>2</sub> percentage as a factor of Wobbe index.

These results do not imply that practical appliances will start flashing back at 6% H<sub>2</sub>, since we must test each appliance separately to determine the point at which they flashback, but rather that, based upon the comparison of the gases themselves, we cannot guarantee performance at more than 5%. Stated differently, if there was an appliance that was at its flashback limit at W = 43.4 MJ/m³, then it would flashback at a hydrogen percentage of more than 5%. Given this modest amount, and the severe restrictions on Wobbe number, one can wonder whether hydrogen addition under these conditions is worthwhile.

Also shown are the results for pure Groningen gas ("G-gas", the model for G-25 and having the same Wobbe number). Here we see that the lower limit gas flashes back more easily than Groningen gas, which is not surprising since the limit gas has a lower equivalence ratio, and thus a higher burning velocity. However, we recall that G-25 is the specified test gas for flashback, according to the European standard. These results suggest (again) that this choice should be reconsidered. If we use the formal test gas for flashback as our "limit" gas (according to the approval regime) we see from Figure 2 that only  $\sim 3\%$  H<sub>2</sub> is acceptable at W=44.4.

Given the fact that the richer the equivalence ratio, compared to the "limit" gas, the more  $H_2$  can be added, the results suggest that a wider Wobbe range can accommodate more added  $H_2$ . With an eye towards extending these data to other countries, we are currently performing these comparisons using the European H-gas ranges as limit gases.

## 5. RESPONSE OF INDUSTRIAL BURNERS (to H<sub>2</sub> addition)

To determine the response of industrial burners to changes in gas composition two 150kW burners were tested: a commercially available generic process burner and a prototype "flameless" burner. Both burners were fired in a lightweight unloaded kiln, following a temperature ramp to furnace temperatures in excess of 1200K. Emissions (CO, O<sub>2</sub>, NO<sub>x</sub>, CO<sub>2</sub>, methane) measurements of the exhaust gases were performed using standard gas analysis equipment.

#### 5.1. Conventional process-burner

The  $NO_x$ -emissions of the standard burner with varying hydrogen addition are shown in Figure 4 for several furnace temperatures. In the experiments the firing rates were kept constant, compensating for the decrease in heating value of the fuel by increasing the flow rate. Up to 50% of hydrogen in the fuel mixture shows an increasing  $NO_x$  emission with  $H_2$  addition of 20-50%. We ascribe this increase to the steady increase in flame temperature with  $H_2$  addition, as described above.

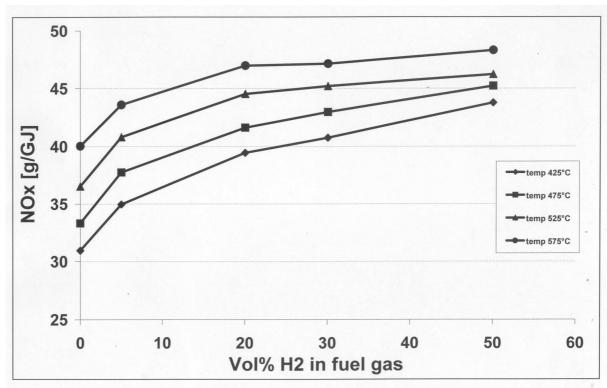


Figure 4: Experimentally determined  $NO_x$  emission as a function of the hydrogen percentage in natural gas and the furnace temperature.

Although hydrogen addition is often touted as a method for "greening" natural gas, one can wonder whether a significant increase in  $NO_x$  emissions is desirable. Certainly for industries at their regulatory limits of allowable  $NO_x$  emissions this increase is unacceptable.

Experiments have shown that hydrogen addition can have a positive effect on CO and methane emissions. We attribute this effect to the increased burning velocity caused by the presence of hydrogen, leading to the flames being stabilised closer to the burner than when being fired with natural gas alone. Thus, less methane escapes the flame to be converted to CO in cooler regions of the furnace. A very modest increase in burner head temperature (5K) was observed.

#### 5.2. Modern prototype flameless burner

The  $NO_x$ -emissions of the 150kW prototype flameless burner with increasing  $H_2$ -addition is shown in Figure 5. Here, the hydrogen was added at a constant furnace temperature. Although the emissions are exceptionally low (3mg/m $^3$  at 3%  $O_2$ ) up to 55%  $H_2$ , further increasing the hydrogen percentage in the natural gas results in a steady increase in  $NO_x$ -emissions. This increase was accompanied by the recovery of a visible flame, which is not visible at lower  $H_2$  content.

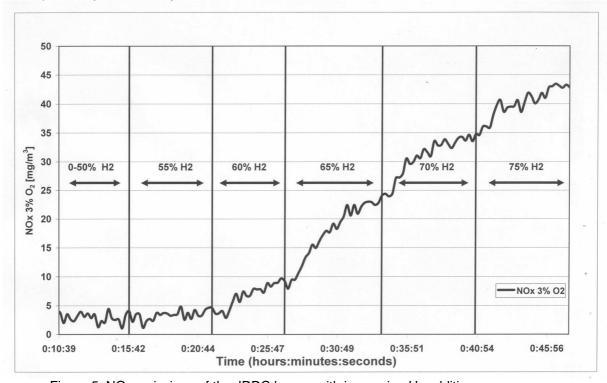


Figure 5: NO<sub>x</sub>-emissions of the JDDC burner with increasing H<sub>2</sub>-addition

Apparently, hydrogen addition completely changes the combustion mode of this burner, from flameless to flame, with the concomitant increase in  $NO_x$  emissions. We suggest that this effect also reflects the increased burning velocity of the hydrogen-natural gas mixture: since the fuel and air in this particular burner are concentrically supplied, an increase in burning velocity causes the flame to stabilise at the end of the gas lance. The lower burning velocity of natural gas results in a "blown-off" flame; however, the fuel and air oxidize completely in the hot furnace gases characteristic of the "flameless" process.

Since one generally considers the flameless mode as a kind of homogeneous oxidation, it could be suggested that the addition of hydrogen to natural gas (given its ease of ignition) should lower the temperature necessary to observe flameless behaviour. If possible, this would allow the use of flameless burners in a much wider range of processes than those at the high temperatures (> 1200K) to which these burners are currently limited. Experiments to determine the minimum temperature at which flameless combustion is observed, using the CO emissions as an indicator of poor combustion, show a negligible difference by adding  $H_2$  up to 25%. Interestingly, the burner fails to generate flameless combustion using pure methane as a fuel, even at a furnace temperature of 1200K. However, the addition of 15%  $H_2$  to the methane decreased the CO emissions to <100ppm, thus improving flameless behaviour for this fuel. Presumably, in natural gas the higher hydrocarbons facilitate homogeneous oxidation, and the results indicate that they are more effective at this than hydrogen.

# 6. RESPONSE OF SPARK-IGNITION PISTON ENGINES (to H<sub>2</sub> addition)

The behavior of spark-ignited piston engines when hydrogen is added to natural gas is mainly influenced by two essential combustion properties of the fuel gas that are changed by the administration of hydrogen.

## 6.1. Laminar burning velocity and flame temperature

The addition of H<sub>2</sub> increases the burning velocity of natural gas.

Most modern natural gas engines operate on lean fuel mixtures. As opposed to stoichiometric engines, this has a number of positive features such as, high thermal efficiency, low likelihood of knock, reduced emissions (especially  $NO_x$ ) and permits using higher compression ratios while reducing heat transfer. However, there are a number of difficulties associated with the lean burn operation. These arise primarily from the associated slower flame propagation velocity, less complete combustion, increased cycle-to-cycle variations (stability) and even occasional flame failure (misfiring). These contribute to poor engine power output and excessive hydrocarbon emissions that may be encountered with lean operation.

The addition of some hydrogen to either methane or natural gas showed an enhancement in performance particularly when operating on relatively lean mixtures (5).

We have performed experiments showing an increase of about 3% (absolute) in overall efficiency by adding 20% of hydrogen to the fuel gas, mainly due to a faster combustion in the first half of the combustion duration and a shorter ignition lag period. If the equivalence ratio is not actively controlled and the Wobbe number is allowed to decrease, the mixture becomes leaner by adding hydrogen. In this case the  $NO_x$  and  $CH_4$  emissions are virtually unchanged. The leaner mixture counterbalances the increase in flame temperature with  $H_2$  addition, due to both a slower burning rate of the leaner mixture and the cooling effect of the excess air. If the equivalence ratio remains unchanged, due to either an active mixture control system or adding higher hydrocarbons to the fuel gas to keep the Wobbe number constant, the  $NO_x$  emission more than doubled at 20% hydrogen, reflecting the increase in flame temperature, similar to the behavior observed for the industrial burner.

## 6.2. Ignition Properties (Ignition delay time / Knock propensity)

Hydrogen and natural gas have significantly different ignition properties; hydrogen spontaneously ignites much easier than methane, and thus results in knocking more easily.

Engine knock arises from auto ignition of the unburned part of the fuel and air charge (the so-called end-gas) while the spark-ignited flame propagating through the combustion chamber. The compression of the fuel and air mixture in the firing cycle raises the temperature of the charge to the point of rapid auto ignition. The smooth and efficient operation of the engine is upset and severe knocking can lead to mechanical failure.

However, our ability to characterize and predict the knock limits of a spark-ignition engine upon  $H_2$  addition is limited by our lack of fundamental knowledge of the processes that cause knock in the engine. The effects of  $H_2$  addition on the ignition delay time of gas-air mixtures (being the relevant property related to engine knock) are currently being investigated.

We are currently deriving a method by which the knock propensity of natural gases can be compared, unambiguously and without reference to specific engines or arbitrary standards, similar to that described for flashback above.

# 7. RESPONSE OF GAS TURBINES (to H<sub>2</sub> addition)

A combustion system particularly sensitive to variations in gas composition is the lean-premixed gas turbine: unwanted spontaneous ignition before reaching the burner and flashback of the flame into the burner can have both disastrous effects on the integrity of the machine. At the other extreme, flame blowout is equally unwanted, and even partial flame lift can result in undesirable acoustic instability. A particular concern regarding ignition is the presence of hydrogen; since this gas

ignites easily, there is apprehension that even small quantities of hydrogen in natural gas will be catastrophic for turbine behaviour. To illustrate this apprehension; one major turbine manufacturer allows only traces of hydrogen in the fuel gas, while another manufacturer allows only 8.5% of H<sub>2</sub>.

On the other hand, addition of large amounts of hydrogen (up to 70%) significantly improves flame stability and allows stable operation in a gas turbine type test burner at the extreme lean fuel/air ratios required for reduced  $NO_x$  emissions (7). However, given the aforementioned sensitivity of premixed turbines to gas composition, the machines can tolerate little fluctuations of the  $H_2$  content of the gas which must be considered.

## 8. CONCLUSIONS

The consequences of hydrogen addition to natural gas for the performance of end-use equipment, such as increased safety risk caused by decreased flame stability and the negative environmental impact of increased  $NO_x$  emissions, must be weighed against any potential benefits in terms of reductions in greenhouse emissions when considering transition scenarios involving using natural gas/hydrogen mixtures to integrate hydrogen, sustainable or otherwise, into the energy infrastructure.

If, as discussed above, we consider the effects of hydrogen addition on the behavior of industrial and domestic burners, spark-ignited gas engines and gas turbines it becomes clear that each generic application has its own characteristic response for which a specific gas-quality specification regarding hydrogen is necessary. The possible future modification of equipment design for increased hydrogen tolerance will thus also require a tailor-made approach.

# Acknowledgement

We gratefully acknowledge the EET Program of the Dutch Ministry of Economic Affairs and Gasunie Trade & Supply for their support of this work.

## **REFERENCES**

- 1. Leiker, M., Cartellieri, W., Cristoph, K., Pfeifer, U., and Rankl, M., ASME Paper No. 72-DGP-4, April 1972.
- 2. Sepman, A., Tiekstra, G., Mokhov, A.V., and Levinsky, H.B., Proceedings of the Second Joint Meeting of the U.S. Sections of the Combustion Institute, Pittsburgh, Paper 279, 2001.
- 3. European Norm EN-437, European Committee for Standardization, 1993.
- 4. A.G.A. Laboratories, "Interchangeability of Other Fuel Gases with Natural Gases", Research Bulletin 36, 1946.
- 5. Karim, G.A., Wierzba, I., and Al-Alousi, Y., Int. J. Hydrogen Energy Vol. 21, pp. 625-631, 1996.
- 6. Bade Shresta, S.O., and Karim, G.A., Int. J. Hydrogen Energy Vol. 24, pp. 577-586, 1999.
- 7. Schefer, R.W., and Oefelein, J., "Reduced Turbine Emissions using Hydrogen-enriched Fuels", FY 2003 Progress Report Hydrogen, Fuel Cells, and Infrastructure Technologies.