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# A Numerical Study of Lean CH<sub>4</sub>/H<sub>2</sub>/Air Premixed Flames at High Pressure

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We perform a numerical study of the effect of including a small amount of hydrogen in lean methane–air premixed flames at high pressure and high temperature conditions. It has been shown recently (Bell and Gupta (1997)) that hydrogen addition extends the lean operating limit of natural gas engines, leading to a potential decrease in pollutant formation. We suggest here that the origin of this effect is that, at constant global equivalence ratio, the stretch resistance of these flames is considerably increased by hydrogen blending, while other flame properties, such as ignition time and burnt gas temperature, are comparatively little modified.

*Keywords:* Lean methane–hydrogen–air flames; stretched flames; extinction; pollutant formation; spark ignition engines

## 1. INTRODUCTION

The combustion of natural gas, which is essentially composed of methane, under lean premixed conditions is of current interest because it has properties, including a high equivalent octane rating, which can potentially lead to low pollutant emissions. In this paper we shall be mainly interested in natural gas combustion in conditions encountered in supercharged spark ignition engines, which occur at high pressure, and high temperature, typically 30 atmospheres and 700 K before combustion. In these conditions,

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a natural gas engine can be seen as an alternative to the Diesel engine, widely used in the bus and truck market.

In terms of pollutant formation, a natural gas engine has the advantage that it produces negligible soot compared to the Diesel engine. Furthermore the low carbon to hydrogen ratio of methane means that natural gas has the lowest ratio of CO<sub>2</sub> per kW of all fossil fuels. Concerning other pollutants, NO<sub>x</sub> production is known to increase almost exponentially with burnt gas temperature (*e.g.*, Heywood, 1988; Warnatz *et al.*, 1996) and so the use of lean mixtures (typical equivalence ratio of 0.65), which lowers the combustion temperature, makes it possible to reduce NO<sub>x</sub> production. However, in spark ignited internal combustion engines, reducing the equivalence ratio not only reduces NO<sub>x</sub> emission but also increases CO and HC (unburnt hydrocarbons) emission, so that a compromise must be chosen. The main reason for the increase of CO and HC emission in lean mixtures is that the extinction limit of the flame is approached, and the stretch associated with the turbulent field can cause local flame extinctions, eventually leading to total extinction for some engine cycles. Of course, such a behavior is not acceptable, so that the equivalence ratio has to be kept reasonably above this limit. Nevertheless, a compromise can be found which allows natural gas engines to satisfy simultaneously the present NO<sub>x</sub>, CO and HC emission requirements.

Naturally as standards evolve and become more and more stringent, it will be necessary to further improve pollutant emission of spark ignition engines. In this context, the effect of including a small proportion of hydrogen in gasoline engines (Li *et al.*, 1986) (Sher and Hacoheh, 1988) (Sher and Hacoheh, 1989), or in natural gas engines (Bell and Gupta, 1997) has been studied experimentally. The main result of this last paper is that the lean operating limit of the engine (an atmospheric engine in their case) is considerably extended by hydrogen blending, leading to a significant decrease in pollutant emission for suitable engine tunings. It will be shown here that flame extinction by turbulence, which can be modeled numerically in a classic way by extinction by strain in a counterflow configuration, is substantially more difficult in the presence of hydrogen, for a fixed global equivalence ratio. Thus for a fixed equivalence ratio, CO and HC productions are considerably reduced when a small amount of methane is replaced by hydrogen, the combustion temperature being only slightly increased. Although our study is oriented towards use of CH<sub>4</sub>/H<sub>2</sub>/air flames in automotive engines, this type of flame could be used in other applications, with the same goal of reducing pollutant emissions.

We perform a numerical study of CH<sub>4</sub>/H<sub>2</sub>/air premixed combustion. In Section 2 we treat the simple case of homogeneous combustion and we will be interested in the combustion temperature and in the induction times. We show that a small proportion of hydrogen will not substantially promote engine knock, contrary to the limiting case of pure H<sub>2</sub>/air flames. In Section 3, results concerning laminar flame velocities and flame structure of freely propagating CH<sub>4</sub>/H<sub>2</sub>/air flames will be given. In Section 4, the combustion of stretched flames in the stagnation point configuration will be studied. Finally, the prospects of CH<sub>4</sub>/H<sub>2</sub>/air combustion will be analyzed in Section 5.

All numerical calculations are performed at high pressure and high initial temperature, using Gri-Mech (Frenklach *et al.*, 1995) which is a CHEMKIN II (Kee *et al.*, 1991) kinetic scheme adapted for methane combustion at high pressure. This kinetic scheme is a C2 scheme (*i.e.*, involves the formation of species with two carbon atoms). Gri-Mech is currently accepted as being the most appropriate kinetic scheme for methane oxidation at high pressure, and has been tested against available experimental data. A comprehensive presentation of these tests is available on the web server of the authors of the scheme.

Homogeneous calculations are performed using the Gri-Mech kinetic scheme in Sandia's program CONP, distributed with the CHEMKIN II package. The calculation of the structure and burning velocities of free and stretched flames is performed using the Gri-Mech kinetic scheme in Rogg's 1-D laminar flame code, RUN-1DL (Rogg, 1991), in its CHEMKIN II version. The homogeneous calculations are performed with a version of the Gri-Mech kinetic scheme including the formation of nitrogen oxides. However the calculations of laminar flames and stretched flames are done without nitrogen chemistry, in order to limit the cost of the calculations.

## 2. HOMOGENEOUS COMBUSTION

### 2.1. Equilibrium Concentrations and Temperature

In this section, we will be interested in the equilibrium temperature and concentrations of pure CH<sub>4</sub>/air flames and of CH<sub>4</sub>/H<sub>2</sub>/air flames at a pressure of 30 atmospheres. In Figure 1, we show the equilibrium concentrations of pure CH<sub>4</sub>/air flames over a wide range of equivalence ratios. As can be seen on this figure, when the equivalence ratio is increased, the molar fraction of oxygen decreases, and molar fractions of CO and HC

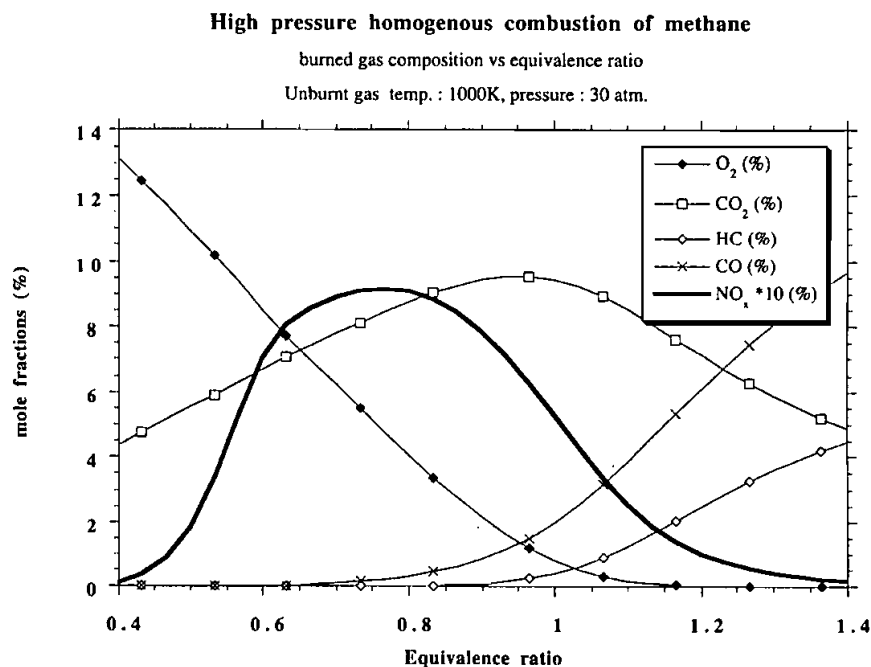


FIGURE 1 Homogeneous methane/air combustion: Equilibrium concentrations and temperature.

(the sum of all unburnt hydrocarbons, including methane) increase. The molar fraction of CO<sub>2</sub> has a maximum close to stoichiometry. When the flame is rich, there is not enough oxygen to permit the total oxidation of CO to CO<sub>2</sub>, and a very high molar fraction CO is observed. The equilibrium molar fraction of NO<sub>x</sub> also has a maximum, but for an equivalence ratio (ER) close to 0.75. In very lean mixtures, NO<sub>x</sub> production is reduced because Zeldovich's thermal mechanism (Warnatz *et al.*, 1996) is less efficient at lower temperature. In richer mixtures, NO<sub>x</sub> decreases before stoichiometry because less oxygen is available.

From the point of view of pollutant formation, it is clear from this figure that natural gas combustion is an interesting candidate, since all three main pollutants, HC, CO and NO<sub>x</sub> reach a low level for very lean mixtures. However a typical choice of operating conditions in a methane engine is an equivalence ratio of 0.65. Lower equivalence ratios are not viable because of the turbulence induced increase in CO and HC, mentioned in Section 1. Of course, all the molar fractions given here are those at equilibrium, and are different from those observed in the case of premixed turbulent combustion.

In particular, the equilibrium concentration of NO<sub>x</sub> is much larger than typically observed concentrations, through the effects of finite reaction rate and subsequent quenching, as is well known (Heywood, 1988).

In Figure 2, we show the evolution of temperature and NO<sub>x</sub> concentrations *versus* the global equivalence ratio for lean CH<sub>4</sub>/air and CH<sub>4</sub>/H<sub>2</sub>/air (with  $x = 20\%$  H<sub>2</sub>, where  $x$  is the ratio of number of moles of hydrogen to number of moles of methane). We emphasize that the global equivalence ratio (ER) is used throughout this article. If we denote by ER<sub>methane</sub> an equivalence ratio only based on methane content, and using the variable  $x$  defined above, it is easily shown that  $ER = ER_{\text{methane}} \cdot (1 + 0.25 \cdot x)$ . The comparison of curves in Figure 2 shows that hydrogen blending does not significantly increase the flame temperature nor the equilibrium NO<sub>x</sub> concentration (the NO<sub>x</sub> concentration is strongly dependent on temperature through the Zeldovich mechanism).

In Figure 3, we compare the effect on NO<sub>x</sub> concentration of:

- i) increasing equivalence ratio from 0.6 to 0.7 for pure CH<sub>4</sub>/air flames
- ii) increasing proportion of hydrogen at a constant (global) equivalence ratio of 0.6.

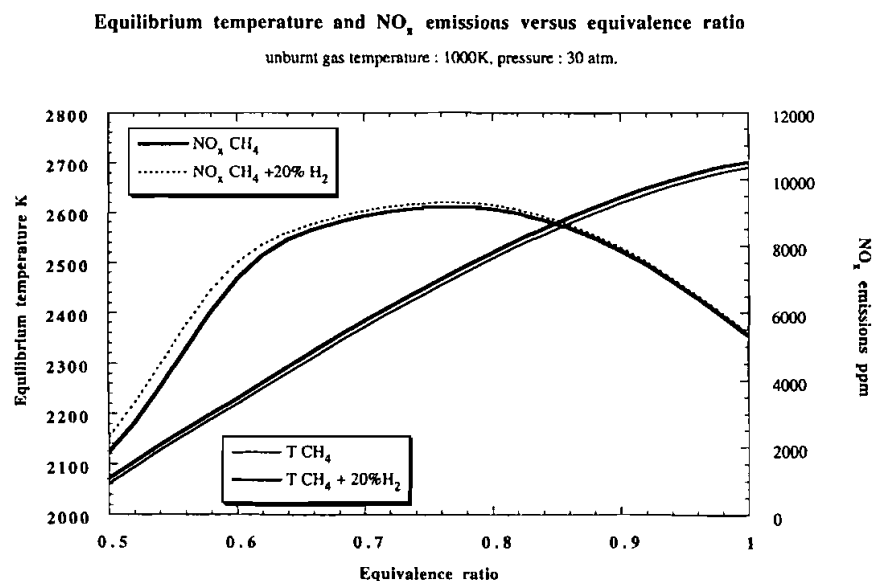


FIGURE 2 NO<sub>x</sub> concentration and temperature *versus* equivalence ratio for methane/air and methane/hydrogen/air homogeneous combustion.

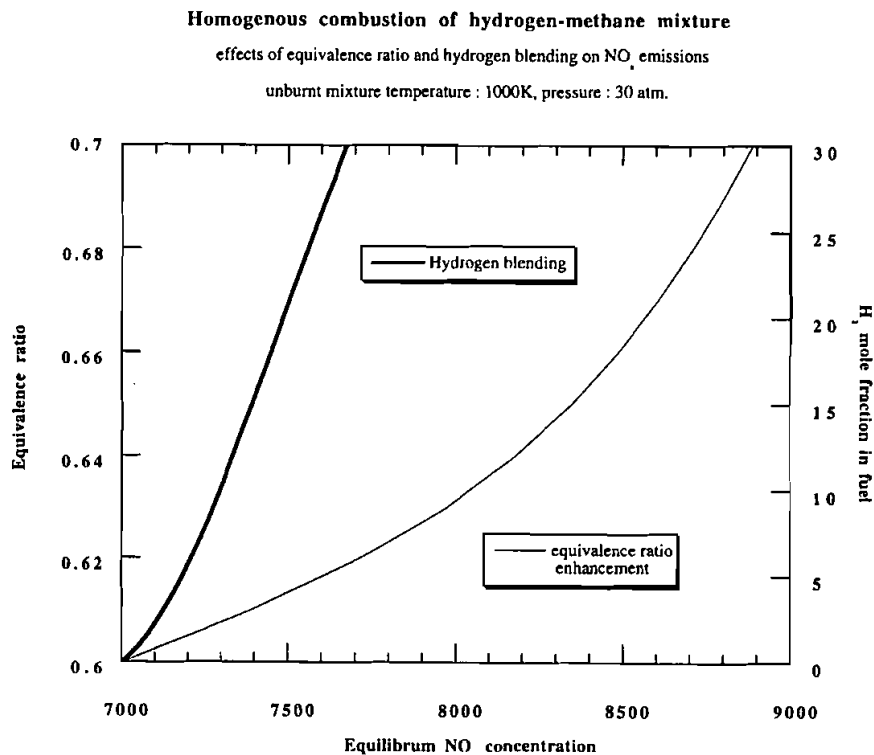


FIGURE 3 Compared influence of hydrogen blending and equivalence ratio increase on equilibrium  $\text{NO}_x$  concentrations.

To motivate this comparison, we anticipate the results on stretched flames that will be reported in Section 4. These results will show that both types of variation induce a higher resistance to stretch, the effect of hydrogen being however stronger. If we compare the equilibrium concentrations of  $\text{NO}_x$  for the same stretch resistance (see later), it will be seen that hydrogen blending results in equilibrium concentrations of  $\text{NO}_x$  noticeably lower than those obtained by increasing the equivalence ratio (the reader is warned that hydrogen proportion and equivalence ratio are plotted here as ordinates).

## 2.2. Induction Times

A property of homogeneous combustion relevant to practical application is the induction time. If we have in mind the behavior of spark ignition engines, induction times control the resistance to knock of a given engine.

These times, typical of volume reactions, should be compared to typical times for turbulent flame propagation. If the induction time at the unburnt gas temperature is less than the turbulent flame propagation time, knock is likely to occur. We have seen in the previous subsection that hydrogen blending does not significantly increase burnt gas temperature for a given equivalence ratio. We are now interested in the variation of induction times caused by the blending. Because of the high knock resistance of natural gas engines, characterized by a high octane number (approximately 130), these engines can be supercharged and use values of compression ratio much higher than in classical gasoline spark ignition engines. However, the octane number of pure hydrogen is low, so that the variation of induction times through blending is an important issue.

In Figure 4 the variation of induction time at a pressure of 30 atm. and global equivalence ratio of 0.6 is plotted *versus* the temperature of the unburnt mixture. The curves correspond to pure CH<sub>4</sub>, CH<sub>4</sub> with 10% H<sub>2</sub>, with 20% H<sub>2</sub>, and pure hydrogen. The inclusion of a proportion of

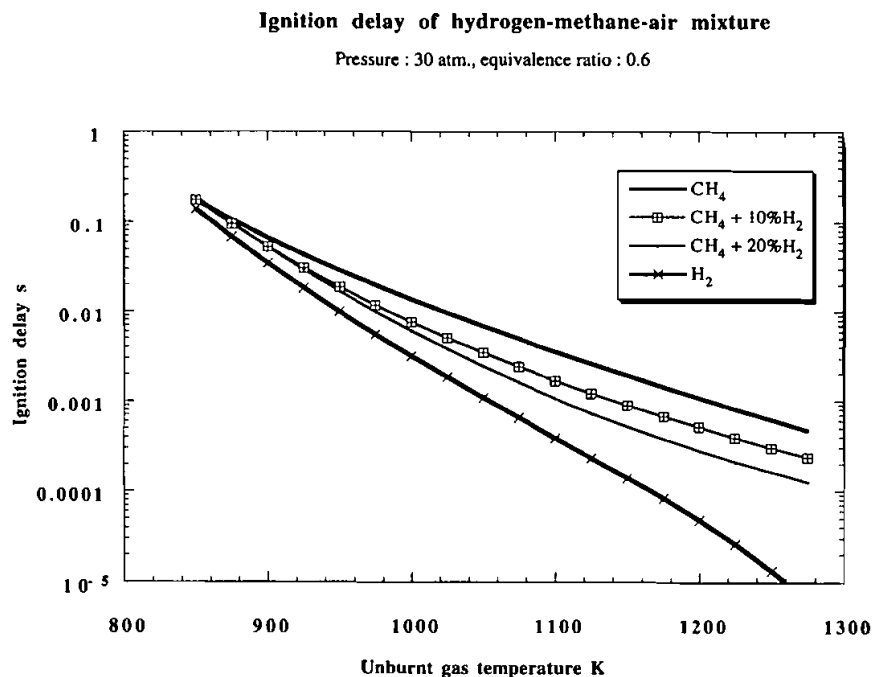


FIGURE 4 Induction times *versus* temperature of unburnt mixture for methane/air and methane/hydrogen/air combustion.



hydrogen decreases the induction time (*i.e.*, promotes the tendency to knock) and the effect increases rapidly as the unburnt gas temperature is increased. For temperatures below 1000 K, the induction time is less than two to three times smaller when 10% or 20% H<sub>2</sub> is added, but is nearly an order of magnitude smaller for pure hydrogen/air flames at this temperature. Compared to hydrogen blending, increasing the equivalence ratio has a weaker effect on the induction time, typically-10%.

We can see here a drawback of including a proportion of hydrogen in natural gas. However, this decrease in induction time, although important, is not dramatic for a small proportion of hydrogen. Furthermore, one of us has remarked (Delorme *et al.*, 1997), that the spark advance has to be reduced when hydrogen is included in natural gas at a given equivalence ratio. This property is related to the reaction rates of stretched flames, which will be discussed in Section 4. This reduction of spark advance limits the tendency of the engine to knock.

### 3. FREELY PROPAGATING FLAMES

In this section we will be interested in the laminar velocity and flame structure of premixed flames at high pressure and temperature. The comparison of pure methane flames and methane/hydrogen flames, begun in the previous section, will be continued. In this section and in Section 4, we use a version of the kinetic scheme (Frenklach *et al.*, 1995) without nitrogen oxide chemistry, in order to limit the computational time.

Figure 5 shows the laminar burning velocity of lean flames *versus* equivalence ratio for various proportions of hydrogen. Inclusion of hydrogen leads to an increase in velocity. This increase is relatively small for 5% or 10% of hydrogen, but reaches 30% for an addition of 20% hydrogen. However for typical values of the parameters (equivalence ratio 0.6) the increase in laminar flame velocity is relatively small.

Figure 6a shows the profiles of the major species of a high pressure pure CH<sub>4</sub>/air flame at an equivalence ratio of 0.6; the minor species are shown in Figure 6b. We will compare our results with hydrogen blending to these reference results. In these figures, the burnt gases are on the right. Let us note for the moment that the concentration of C<sub>2</sub> radicals (*i.e.*, the radicals involving two carbon atoms) are not negligible compared to that of C<sub>1</sub> radicals, showing the usefulness of a C<sub>2</sub> kinetic scheme at high pressure. Moreover, these C<sub>2</sub> species would be even more important at a higher

**Laminar flame speeds of hydrogen-methane-air mixtures**effects of H<sub>2</sub> fraction and equivalence ratio on flame speed

Pressure : 30 atm.; unburned gaz temperature : 700K.

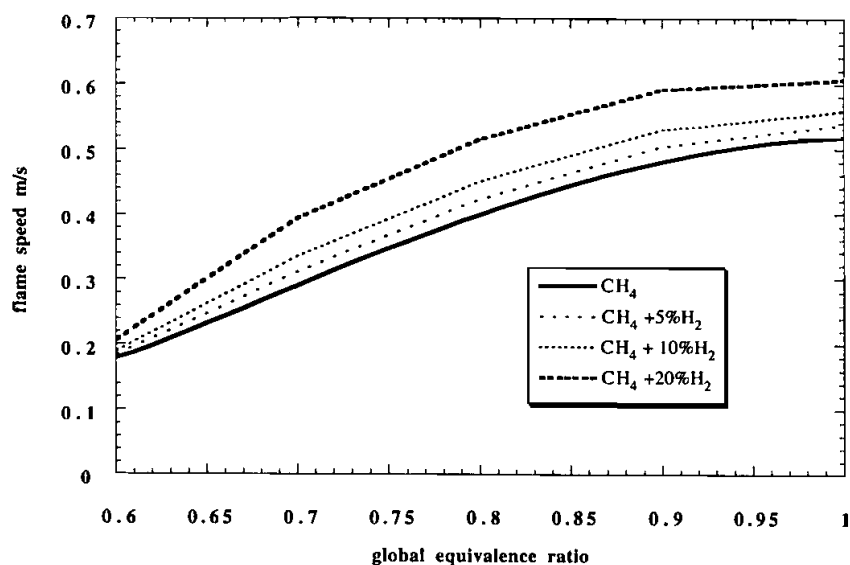


FIGURE 5 Influence of hydrogen proportion on laminar flame velocity.

equivalence ratio (Warnatz, 1981), for instance, the radical recombination reaction,  $2\text{CH}_3 + \text{M} \rightarrow \text{C}_2\text{H}_6 + \text{M}$ , is enhanced with increased equivalence ratio.

Figures 7a and 7b show, respectively, the major and minor species of a CH<sub>4</sub>/H<sub>2</sub>/air flame with the same global equivalence ratio 0.6, but with 20% of hydrogen. Contrary of Figure 6, H<sub>2</sub> is now plotted with the major species. Apart from H<sub>2</sub>, other species and the temperature are not greatly modified compared to Figures 6a and 6b; this result is compatible with the small change in laminar velocity seen in Figure 5 at this particular value of the equivalence ratio. Radical concentrations relevant to hydrogen/oxygen chemistry, such as H, OH and O are only slightly increased by hydrogen blending.

We now turn to the problem of stretched flames, where the difference between the two cases, with or without hydrogen, will be much more significant.

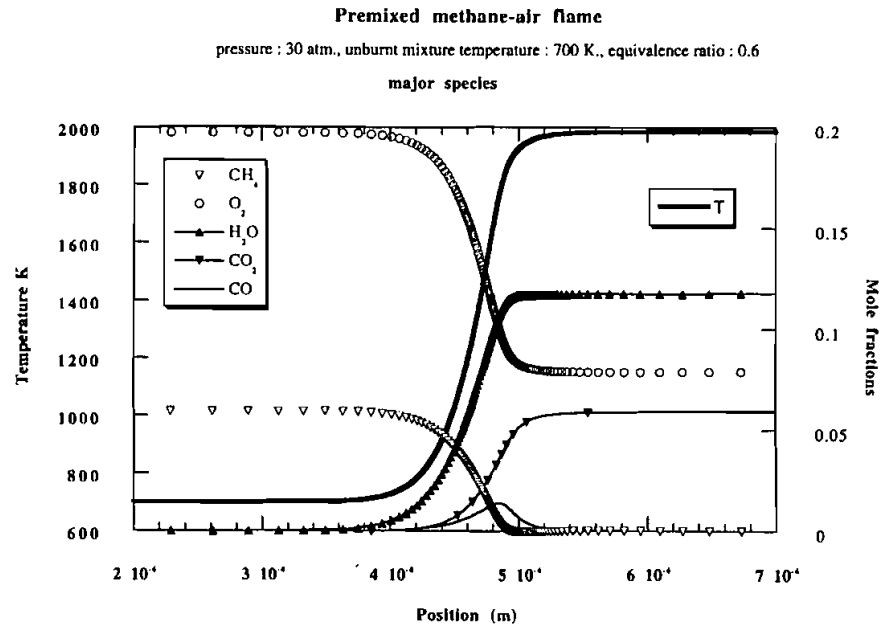


FIGURE 6a Structure of a laminar methane/air flame: major species.

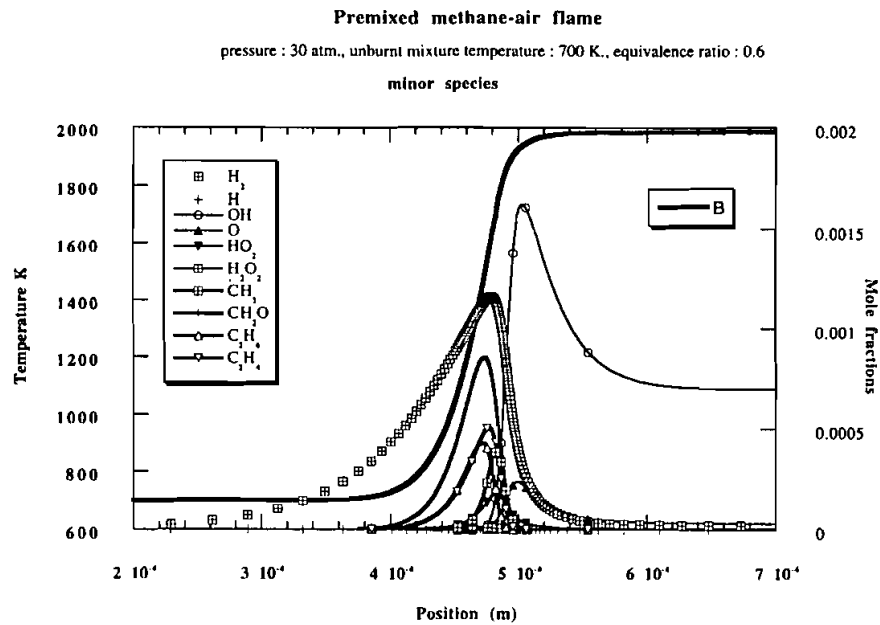


FIGURE 6b Structure of a laminar methane/air flame: minor species.

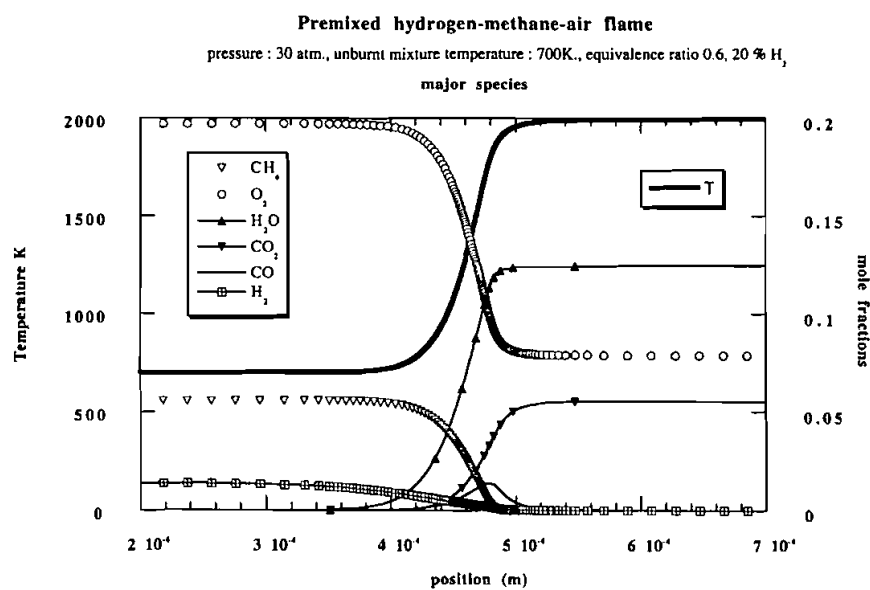


FIGURE 7a Structure of a laminar methane/hydrogen/air flame: major species.

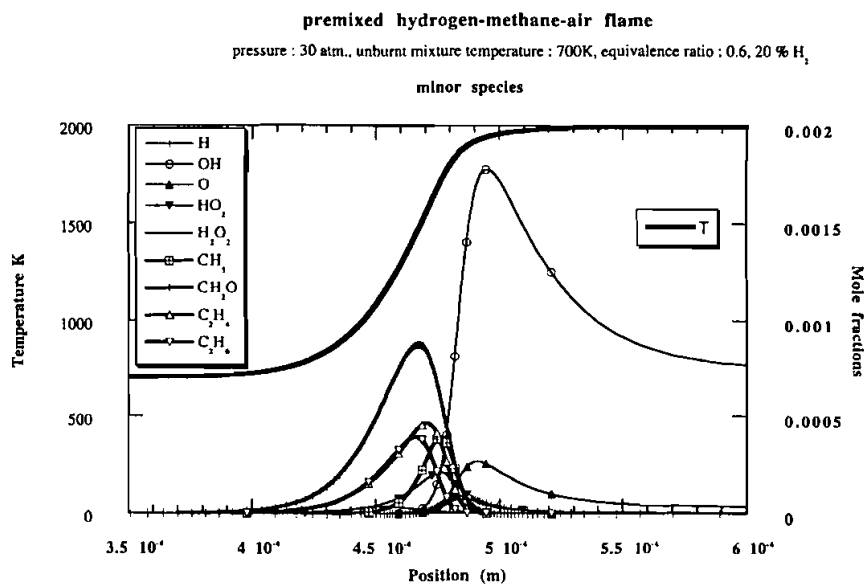


FIGURE 7b Structure of a laminar methane/hydrogen/air flame: minor species.

#### 4. STRETCHED FLAMES

In Section 3, we were interested in the characteristics of lean laminar flames with or without hydrogen blending. However, in practical applications of natural gas premixed combustion, flames are submitted to a turbulent flow field. As a result, they experience strong local stretch which can cause local extinctions, or at least very slow combustion. Although the complete coupling of turbulence and chemistry is a very difficult problem, it can be simplified if one makes use of the idea of flamelets to study the local behavior of a flame submitted to strain, in the configuration of the stagnation point flow. This simplification is not perfect, as for instance effects of curvature and instationarity are neglected (see however Stahl and Warnatz (1991) for a study of the effect of a strain rate varying with time and Mishra *et al.* (1994) for a numerical investigation of the effect of curvature in a spherical geometry). The main idea here is to obtain an order of magnitude of the strain rates necessary to extinguish the flame.

This order of magnitude can then be compared to typical values of strain observed in practical configurations. The highest strain rates in spark ignition engines are generated by the smallest scales of turbulence. Using Kolmogorov scaling laws and previous experiments in spark ignition engines (Daneshyar and Hill, 1987), a typical strain rate has been estimated by Blint (1991) to be of the order of  $5 \cdot 10^4$  to  $1 \cdot 10^5 \text{ sec}^{-1}$ . Actually this strain depends of course on operating conditions of the engine, particularly on rpm.

Although combustion independent phenomena, such as quench layers will lead to some HC emission even for stoichiometric flames, it is more than probable that the dramatic increase in unburnt hydrocarbon emission at the lean running limit is related to strain induced local extinction of the flame and incomplete combustion. The very high strain rates mentioned above are thus believed to be at the origin of increased CO and HC pollutant formation in very lean flames.

This effect limits the use of lean equivalence ratios, a typical value being 0.65, chosen as a compromise between  $\text{NO}_x$  emission, which decreases for lean flames, and HC and CO emissions, which increase. Bell and Gupta (1997) have shown that hydrogen blending makes it possible to extend the operating conditions to leaner flames, leading to an effective reduction of pollutant formation by a suitable new choice of the compromise. Some results concerning the effect of blending, not necessarily for lean flames, can also be found in the experimental study of Yamaoka and Tsuji (1992) in the "back to back" flame configuration. We will show below that the observed

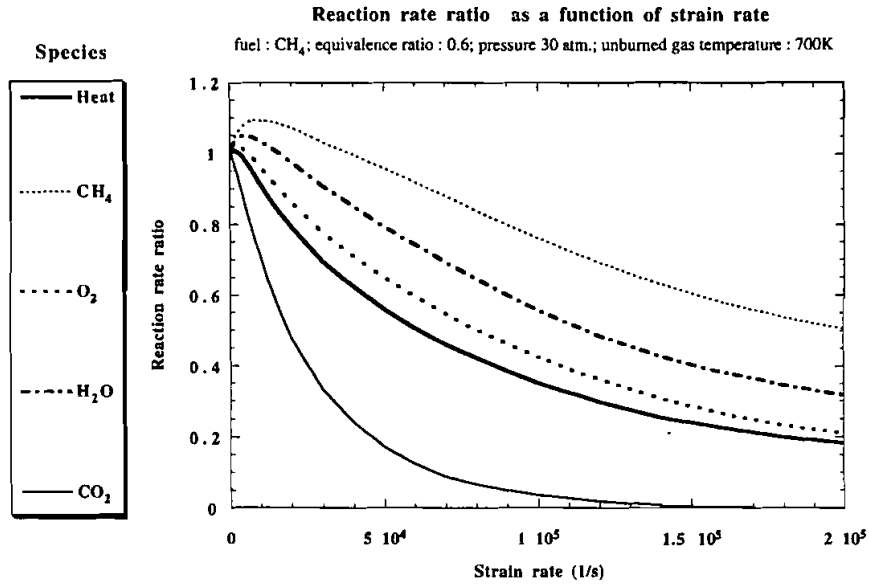
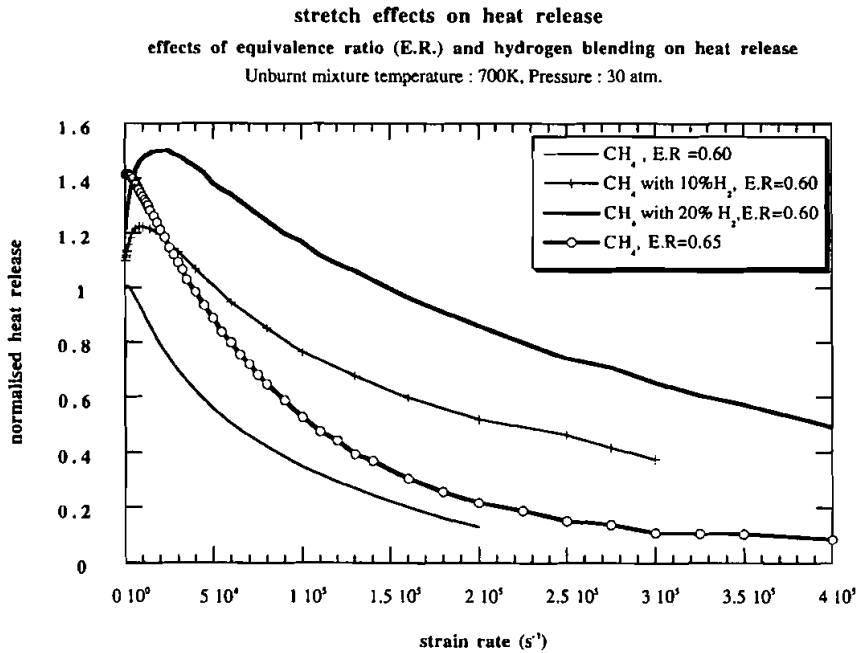
reduction in pollutant formation by hydrogen blending can be explained by a strong increase in resistance to strain produced by addition of a small amount of hydrogen.

In this section, we perform a numerical study of CH<sub>4</sub>/H<sub>2</sub>/air mixtures in the “fresh to burnt” stagnation point flow configuration (Rogg, 1988). As in the previous section, the calculations will be performed at high pressure and temperature. We recall that numerical studies of pure methane/air flames at atmospheric pressure have become classic, see for instance (Rogg, 1988; Giovangigli and Smooke, 1987; Lee and Chung, 1994), as well as experimental studies of the same type of flames (Law *et al.*, 1986; Ishizuka and Law, 1982). However data at high pressure and temperature is extremely scarce.

To analyze the effect of strain rate on the flame properties, we make use of the concept of reaction rate ratios (ratio of the integrated reaction rate to the integrated reaction rate at zero strain) introduced by Cant *et al.* (1994). In this definition, the integration is performed over the length of the computational domain. To make the comparison between pure CH<sub>4</sub> and CH<sub>4</sub>/H<sub>2</sub> flames, we normalize our results by the integrated reaction rate for pure methane/air flames at zero strain.

In Figure 8 we show the integrated reaction rate of the major species and the heat release rate for pure CH<sub>4</sub>/air lean flames at high pressure and temperature. The reaction rates are plotted against strain rate. It can be seen that the high pressure flame can withstand a value of strain rate of the order of  $10^5 \text{ sec}^{-1}$ . The resulting decrease in the heat release ratio is a factor three. However, the reaction rate ratios of the different species respond in a different way to strain. For example, the CO<sub>2</sub> production rate drops very quickly. This fact provides an explanation for the strong increase in CO emission, observed in experiments for very lean turbulent flames. The CH<sub>4</sub> consumption rate also decreases with strain rate, leading to higher HC emissions (CH<sub>4</sub> is a major component of this type of emission). The effect on CH<sub>4</sub> is however less strong than for CO<sub>2</sub>.

In Figure 9, we show the effect of 10% and 20% hydrogen blending on the integrated heat release ratio. The resistance of the flame to strain is greatly improved by hydrogen, and this effect is probably the main cause of the improved behavior of natural gas engines with hydrogen blending, observed experimentally by Bell and Gupta (1997). In this figure we also plot the effect, on heat release ratio, of increasing the equivalence ratio from 0.60 to 0.65 (the integrated heat release is always normalized by the integrated heat release at zero stretch of a flame with an equivalence ratio of 0.6). At very small strain, this effect is greater than for hydrogen blending,

FIGURE 8 Integrated reaction rates *versus* strain rate for strained methane/air flames.FIGURE 9 Integrated heat production rate *versus* strain rate for strained methane/air and methane/hydrogen/air flames.

but as the strain is increased, the heat release ratio with only 10% hydrogen blending at equivalence ratio 0.6 quickly overtakes that of richer pure CH<sub>4</sub> flame. It may be noted that at a strain rate of  $2 \times 10^5 \text{ sec}^{-1}$ , the heat release rate of the lean CH<sub>4</sub>/air flame is only 10% of the value of the unstrained flame, whereas with 10% hydrogen blending, at the same global equivalence ratio, it is 50%, and with 20% hydrogen blending it is 90%. In other words, at this strain rate, the flame doped with 20% hydrogen is predicted to react nine times faster than the pure methane flame.

We now turn to the structure of these stretched flames. In the following figures, contrary to the case of freely propagating flames, the burnt gases will be shown on the left. In Figures 10a and 10b we show, respectively, the major and minor species of stretched pure lean CH<sub>4</sub>/air flame at high pressure. As before, we use an equivalence ratio of 0.6. This flame is shown for a value of the strain rate equal to  $6 \times 10^4 \text{ sec}^{-1}$ . Compared to the free flame of Figures 6a and 6b, it can be seen that profiles are much steeper in Figures 10a and 10b.

In Figures 11a and 11b we show the major and minor species of the same type of flame, for the same values of the parameters, but with 20% of hydrogen. The higher value of heat release ratio, seen in Figure 9, is reflected in Figure 11b by a large increase in the concentration of OH, H and O radicals, this increase is much higher than the increase in the concentration of radicals containing carbon atoms. In some sense, the hydrogen–oxygen chemistry is sustaining the flame, while the products typical of hydrocarbon chemistry stay at the low levels observed in Figure 10b.

## 5. DISCUSSION

With the results of the previous section, we are now in a position to synthesize our results and explain the improvement of pollutant emissions observed experimentally. The major effect of hydrogen blending is the improved behavior of the flame in response to strain, which indicates that the flame is able to withstand higher turbulence levels when hydrogen is included. At a fixed value of the global equivalence ratio, the result is reduced emissions of CO and HC compared to the methane/air flame.

A similar effect could be obtained by simply increasing the equivalence ratio, but as seen in Section 2, a higher equivalence ratio means a higher flame temperature and higher values of NO<sub>x</sub> concentrations. On the contrary, hydrogen blending does not significantly alter the flame temperature.



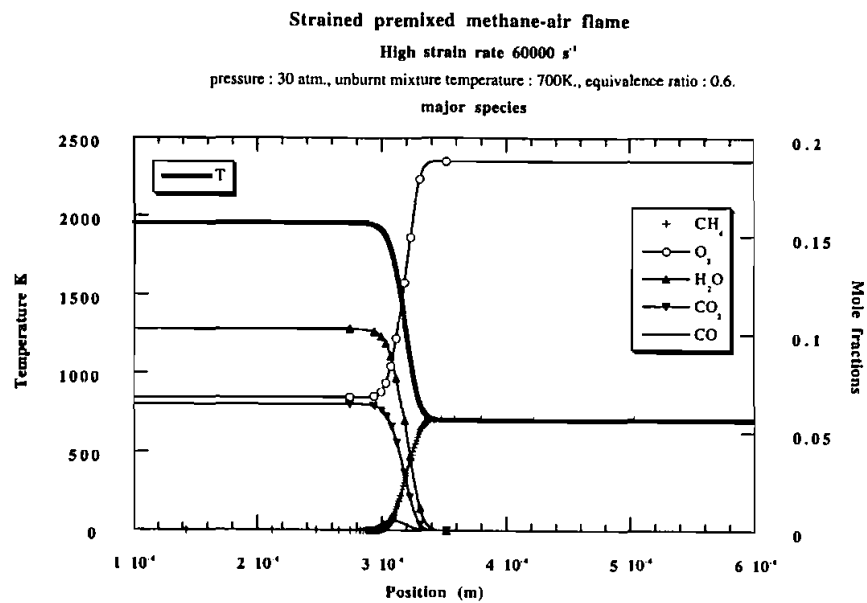


FIGURE 10a Structure of a strained methane/air flame: major species.

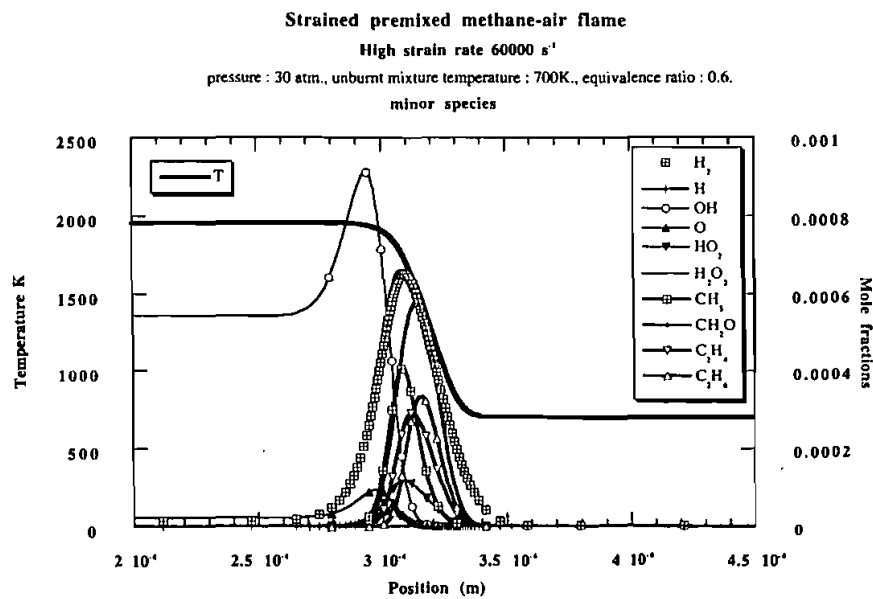


FIGURE 10b Structure of a strained methane/air flame: minor species.

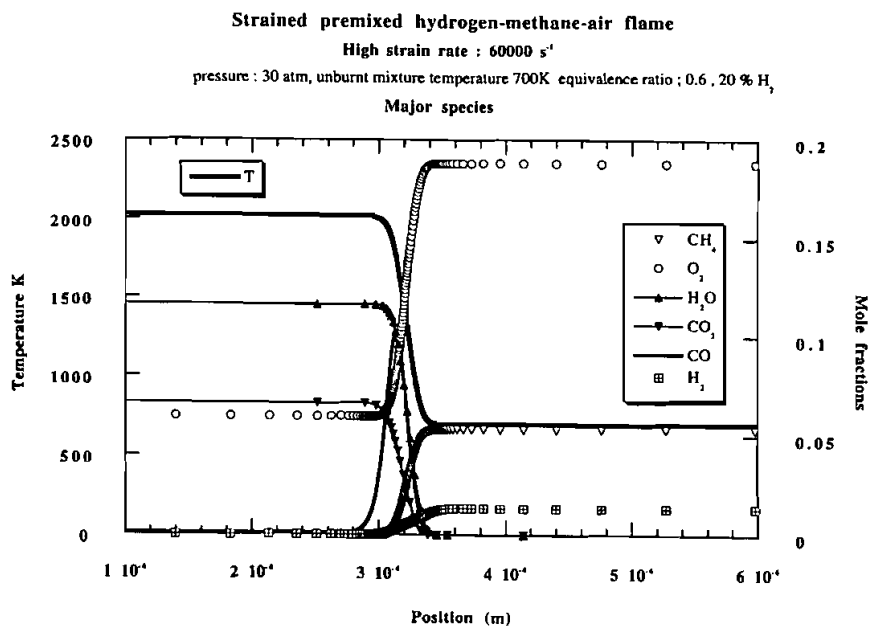


FIGURE 11a Structure of a strained methane/hydrogen/air flame: major species.

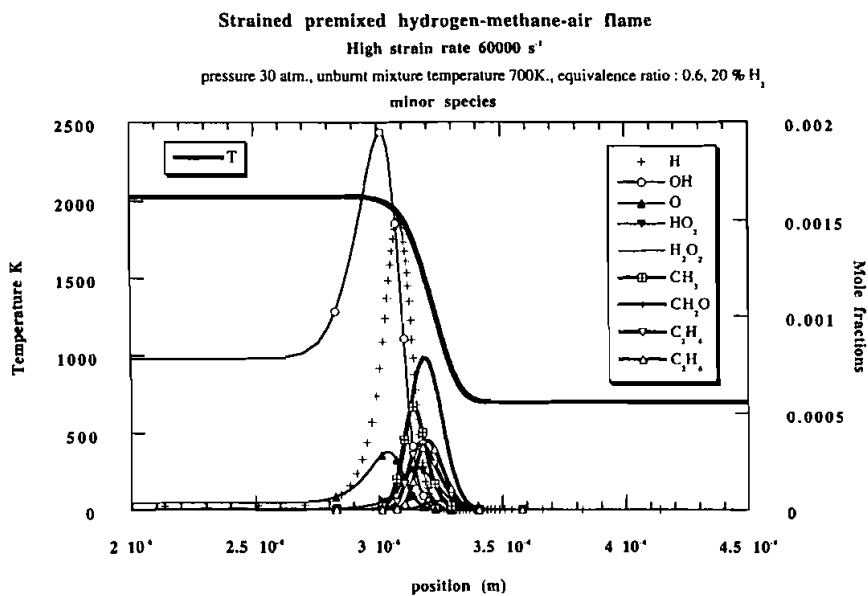


FIGURE 11b Structure of a strained methane/hydrogen/air flame: minor species.

The previous reasoning was performed at constant equivalence ratio. The higher resistance to stretch with hydrogen, demonstrated in this paper, can be used to shift the operating conditions to leaner flames compared to pure methane/air flames. With this new compromise,  $\text{NO}_x$  emissions can be reduced for constant CO and HC levels. It is also possible to choose a compromise which reduces  $\text{NO}_x$ , CO and HC simultaneously, but obviously in a smaller proportion.

A slight problem caused by hydrogen blending is the reduction of induction times, leading to an increased tendency towards knock. However, the increased values of the reaction rates of methane/hydrogen flames observed in Section 4 makes it possible and necessary to use smaller values of spark advance (Delorme *et al.*, 1997), which in turn reduces the probability of knock. So this problem should not be critical for practical applications.

## 6. CONCLUSION

The numerical calculations performed in this paper are compatible with experimental results and explain which combustion properties are responsible for the reduced pollutant emissions observed in premixed methane/hydrogen/air combustion. The simplification of the turbulent combustion problem to that of the stagnation point flame has been fruitful, although one should keep in mind that such an approach neglects the effects of curvature and non-stationarity. The use of hydrogen blending offers the possibility of reducing the pollution emission of natural gas combustion to even lower levels; practical applications of this possibility will of course depend on economic considerations and future pollution standards. Automotive applications have been emphasized in the text, but we would like to stress that other applications are possible, such as co-generation for instance.

### **Acknowledgements**

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