The Effect of Hydrogen Enrichment on CH₄-Air Combustion in Strong Dilution.

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**Abstract**

Lean premixed combustion is a solution to reduce flame temperatures and pollutant emissions, especially NOx. This will strongly use for the future internal combustion engines. But the main drawbacks are the decrease of the global burning speed and the misfire. The aim of this study is first to verify or not is the addition of an amount of hydrogen can extend the lean operating limit of air-methane combustion with dilution and maintain the flame velocity at the same value without dilution. For that, the effect of adding hydrogen to methane-air-N₂ mixtures has been investigated. The laminar burning velocity for different N₂ dilution amounts (from 0 - 30%) and for different hydrogen additions (from 0 to 30%) has been determined at atmospheric pressure and ambient temperature. Experiments were carried out in a stainless steel combustion chamber. Flame speeds were determined by using classical shadowgraph technique. The images were recorded by a high-speed camera. A computational method based on a detailed chemical kinetic scheme (GRIMECH 3.0) was used to provide the expansion factor and to calculate the laminar burning velocities for comparison with experimental results. As hoped, by increasing hydrogen percentage in the mixture leads to an increase of the laminar burning velocity, but the absolute increasing due to the hydrogen addition of the laminar burning velocity decreases as the dilution with N₂ become higher.

1. Introduction

A way to reduce pollutant emissions, especially NOₓ is the lean combustion due to the lower combustion temperatures. However, lean premixed combustion has two limitations. The first is the instability of the flame and the misfire. The second limitation is the lower laminar burning velocity.

A strategy to overcome these problems is the addition of an amount of hydrogen, while hydrogen has a high laminar burning velocity and a wide flammability limits. Experimental studies [1] have shown that increasing the hydrogen content in the mixture was found to be responsible for the increase in the laminar burning velocity and for a reduction of the flame dependence on stretch. Ilbas et al. [2] found that if Hydrogen content increases in the Hydrogen-methane mixtures, the flame speeds and thus the burning velocities also increased dramatically and that the flammable regions were widened with hydrogen content increased in the mixture. Other investigations [3] showed that a small amount of hydrogen addition in turbulent premixed methane-air flames introduces slight linear modifications on the flame reactivity, in increasing the flame stability.

Akansu et al. [4] investigated on the utilization of natural gas-hydrogen mixtures in internal combustion engines and found that HC, CO₂ and CO emissions value decrease with increasing hydrogen percentage. The NOₓ emissions values generally increase with increasing hydrogen content, but, if a catalytic converter, an EGR system or lean burn technique are used, NOₓ emission values can be decreased to extremely low levels.
The laminar burning velocity is a fundamental parameter that characterizes both laminar and turbulent premixed combustions, within the wrinkled thin laminar flamelet regime. An experimental technique to determine the laminar burning velocity and the corresponding Markstein length is the spherical expanding flames used by [5, 6]. The stretch rates for the spherical expanding flames, ignited from a central ignition point, are well defined. A linear relation between the stretch rate and the laminar burning velocity proposed by Calvin [7] is used to extract the value of the unstretched laminar burning velocity. For the present experimental investigations, spherical expanding flames are studied by using shadowgraph technique. Two methodologies allowing the obtaining of the laminar burning velocity are tested in the present paper: the linear extrapolation on zero stretch rate and the analysis method [8]. A computational method based on a detailed chemical kinetic scheme (GRIMECH 3.0) [9] was used to calculate adiabatic flame temperatures and laminar burning velocities. The objective of this work is to determine the effect of hydrogen addition on the laminar burning velocity and to verify if it extends the lean operating limit of air-methane premixed combustion in dilution conditions.

2. Experimental details

In this study, experiments were carried out in a stainless steel combustion chamber. The combustion chamber has an inside volume of 24.32 liters. Two opposite and transparent windows (diameter 10.5cm) provide an optical access. The gases were mixed through an inside electric fan. Two tungsten electrodes (diameter 0.8mm), linked to a conventional capacitive discharge ignition system, are used to form the spark gap (2.8mm) at the centre of this chamber. The electrodes are slightly tilted with respect to the visualization plane; this decreases the display of disturbances due to the ignition in the images recorded. The volumes of methane and air are introduced inside the chamber with thermal mass flow meters, previously calibrated with the gases used in these experiments. In addition to the inside fan, this provides a perfect homogenous mixture. Before filling the chamber with gases, the vacuum was created in the chamber, and after introducing the different volumes of gases, the chamber is at atmospheric pressure, verified with a piezoelectric pressure transducer. Illumination was provided by a continuous Stabilite 2017 Argon ion Laser, with maximum output power of 6W. A parallel light was created by two plan-convex lenses, having 15 mm and 70 mm diameter and a 25 mm and 1000 mm focal length respectively. After the passage through both lenses and inside the combustion chamber, the beam is displayed on a screen; the maximum beam diameter can not exceed 70 mm (the diameter of the second lens). The evolution of the flame surface was observed by using shadowgraph method. The shadowgraphs are recorded by a high speed video camera operating at 6000 frames/s. Measurements, limited to flame having diameters less than 60 mm, imply that the total volume of burned gas was less than 0.5%, therefore the total chamber pressure can be considered constant. To avoid ignition disturbances, a minimum of flame radius greater than 7 mm was considered.
3. Laminar characteristics extraction

After the spark, the flame is ignited and propagates spherically. The radius of the flame growth versus time is presented in Fig. 1.

As mentioned before, the small flame radius represents a distortion due to the ignition, and once the radius becomes greater than 7 mm the flame assumes a perfect spherical geometry. For each mixture, the experience was performed twice and the values reported are a simple average of both values obtained.

A Matlab program was used to detect the luminous front and for image analysis. Instantaneous flame front radius is obtained from image post processing (background subtraction). Even for high dilution levels, the error induced by assuming a spherical shape for the flame front is very low. Thanks to the high speed recording, the mean flame front temporal evolution is obtained an example is presented in Fig. 2.
Fig. 2. Shadowgraphs of the flame propagation for CH₄-air mixture with 20% H₂ addition and 20% N₂ dilution.

An example of flame front detection is also presented in Fig. 3, one can note that even for diluted mixtures, the flame front can be assumed spherical.

![Flame front detection](image)

Fig. 3. Flame front detection for CH₄-air mixture with 20% H₂ addition and 20% N₂ dilution.

The relation between the change in laminar flame speed and stretch, including strain rate and curvature effects, proposed by Calvin [7] is:

\[ V_s = V_{50} - L_b \times K \tag{2} \]

With \( V_s \) the stretched flame speed, calculated by:

\[ V_s = \frac{dR}{dt} \tag{3} \]

\( V_{50} \) the unstretched flame speed

\( L_b \) : Markestein length for burned gas, it corresponds to a measure of the flame response to the stretch

\( K \) : the total stretch rating on the flame, which represents the fractional time rate of a flame surface element of area \( A \), defined as:

\[ K = \frac{1}{A} \times \frac{dA}{dt} \tag{4} \]

In case of spherically outwardly expanding flame front, this expression for flame stretch can be written as the following:

\[ K = \frac{2}{R} \times \frac{dR}{dt} \tag{5} \]
In these conditions, we can use the laminar flame speed and flame stretch linear relation Eq. (2).

**First methodology:**

$V_s$ was calculated after a 3rd degree polynomial fitting of the radius. The degree of polynomial was chosen on the basis of fitting errors and the linearity of the relation between laminar flame speed and stretch. The unstretched flame velocity $V_{S0}$ was obtained by a linear extrapolation to a zero stretch rate.

The unstretched laminar burning velocity based on unburned gas properties can be deduced from this relation:

$$u_{0} = \frac{\rho_b}{\rho_u} V_{S0} (6) , \rho_b \text{ the burned gas density and } \rho_u \text{ the unburned gas one}$$

The density ratio $\frac{\rho_b}{\rho_u}$ needed to extract $u_{0}$ was computed using the adiabatic flame calculation with Premix.

**The second methodology**

Another $u_{0}$ and $L_b$ were determined by using the Analytic method given by Burluka et al. [8] and based on least squares on calculating $u_{0}$ and $L_b$ from the radius time record.

In this methodology, the rays used are not those obtained after polynomial fitting, but the original rays given by image analysis ($r_{original}$). That is done to have an idea about the impact of polynomial fitting on the results and to compare two different methodologies.

In the spherically expanding flames, as mentioned before, the total stretch rate acting across the flame surface can be written as:

$$K = \frac{1}{A} \frac{dA}{dt} = 2 \frac{dR}{dt} = 2 \frac{dA}{dR} = 2 * V_s (7)$$

The substitution of this equation with eq. (2) and the integration with respect to time gives :

$$r_{original}(t) - r_{original}(t_0) + 2*L_b*\ln\left(\frac{r_{original}(t)}{r_{original}(t_0)}\right) = V_{S0} *(t-t_0) (8)$$

The correspondent least squares function is:

$$\Psi(L_b, V_{S0}) = \sum_{i=1}^{N} \left( t_i - t_0 - \frac{r_{original} - r_{0 \text{ original}}}{V_{S0}} - 2 * \frac{L_b}{V_{S0}} * \ln \left( \frac{r_{original}}{r_{0 \text{ original}}} \right) \right)^2 (9)$$

Where $N$ is the number of radius considered.

The condition of the minimum of this least squares function is:

$$\frac{d\Psi}{dV_{S0}} = \frac{d\Psi}{dL_b} = 0 (10)$$

This condition give a system that involves two equations and two unknowns ($V_s$ and $L_b$). We can obtain the values of $V_s$ and $L_b$ by solving this system.

The laminar burning velocity for different N2 dilution amounts (from 0 - 30%) and for different hydrogen additions (from 0 to 30%) was determined, all experiments are performed at an initial pressure of 1 atmosphere and an initial temperature of 300K.
4. Results and discussion

A determination of the flammability limit for CH$_4$-H$_2$-air-N$_2$ mixtures varying the H$_2$ percentage in the mixture (from 0% to 30%) was done using a visual criterion. Fig.4 shows that the addition of an amount of hydrogen can extend the lean operating limit of methane-air-N$_2$ mixtures. With 30% of H$_2$, and with the same ignition system described before, it is possible to extend the lean limits from 34% of N$_2$ dilution to 39% of N$_2$ dilution.

![Flammability limits for CH$_4$-H$_2$-air-N$_2$ mixtures as function of H$_2$% amount.](image1)

Fig. 4. Flammability limits for CH$_4$-H$_2$-air-N$_2$ mixtures as function of H$_2$% amount.

Fig. 5. is a selection of an experimental data after polynomial fitting, for CH$_4$-air mixture with 0, 10 and 20% of H$_2$ addition and 20% of N$_2$ dilution, that shows the variation of flame velocity with total stretch rate. The flame grows from right to left as the flame stretch is inversely proportional to the flame radius.

![Evolution of the laminar flame velocity as a function of stretch rate for mixtures with different hydrogen addition and with 20% of N$_2$ dilution.](image2)

Fig. 5. Evolution of the laminar flame velocity as a function of stretch rate for mixtures with different hydrogen addition and with 20% of N$_2$ dilution.

The effect of stretch on the flame velocity changes varying the percentage of hydrogen in the mixture. These results show a decrease of the dependence of the flame velocity of the stretch with the addition of hydrogen; while the slope of the curve velocity against stretch decreases with higher hydrogen addition in the mixture.

The laminar burning velocities, presented in Fig. 6., and obtained using Eq. (6) after a linear extrapolation are compared to the laminar burning velocities calculated by the analytic methodology. It can be concluded that these two methodologies are in a good agreement and give similar results.
Fig. 7 shows that the experimental results and the values of the laminar burning velocities calculated with Premix have the same trend with N$_2$ dilution and the values of the both methods are too close.

![Graph showing laminar burning velocities](image1)

![Graph showing laminar burning velocities](image2)

Fig. 6 and Fig. 7 - Comparison of laminar burning velocities obtained from two different methods and from Premix simulation.

A relative variation of the laminar burning velocity, was calculated as:

\[
r_{\text{inc}} = \frac{u_{10\%} - u_{0\%}}{u_{0\%}}
\]

(11) where \( n \) is the percentage of hydrogen addition and \( u_{10\%} \) is the experimental laminar burning velocity.

The results reported in Table 1 show that the relative variation is constant, with the same percentage of hydrogen addition, for different percentage of N$_2$ dilution.

<table>
<thead>
<tr>
<th>$r_{\text{inc}}$ for different mixtures</th>
<th>0% of H$_2$</th>
<th>10% of H$_2$</th>
<th>20% of H$_2$</th>
<th>30% of H$_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0% of N$_2$</td>
<td>0</td>
<td>0.096</td>
<td>0.154</td>
<td>0.232</td>
</tr>
<tr>
<td>10% of N$_2$</td>
<td>0</td>
<td>0.068</td>
<td>0.117</td>
<td>0.223</td>
</tr>
<tr>
<td>20% of N$_2$</td>
<td>0</td>
<td>0.099</td>
<td>0.145</td>
<td>0.224</td>
</tr>
<tr>
<td>30% of N$_2$</td>
<td>0</td>
<td>0.133</td>
<td>0.172</td>
<td>0.268</td>
</tr>
</tbody>
</table>

Table 1 – Variation of the laminar burning velocity as function of H$_2$ and N$_2$ additions

5. Conclusion

From these first results, we clearly proved that the flammability limits are extended for air-methane-N$_2$ flame by adding H$_2$. Moreover, the study of the laminar burning velocity indicates also that the addition of H$_2$ allows a faster combustion propagation of air-methane mixture in higher diluted atmosphere, but the initial value (without dilution) is not reached. These results are very promising: the addition of small quantity of H$_2$ can allow the engine working with higher Exhaust Gases rate.

References

2. M. Ilbas, A. P. Crayford, I. Yılmaz, P. J. Bowen and N. Syred, Laminar-burning


