Full Length Article

# Numerical and experimental analysis of the effect of adding water electrolysis products on the laminar burning velocity and stability of lean premixed methane/air flames at sub-atmospheric pressures

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HIGHLIGHTS

- Experimentally, $CH_4 + 10\%$ $H_2/O_2$ blend didn't strongly increase $S_L$ compared to $CH_4$.
- Experimentally, for $CH_4 + 25\%$ $H_2/O_2$ blend $S_L$ increases up to 14.8% compared to $CH_4$.
- Intrinsic instabilities were found for the $CH_4 + 25\%$ $H_2/O_2$ mixture.
- Intrinsic instabilities were not found for $CH_4$ and $CH_4 + 10\%$ $H_2/O_2$ mixtures.

ARTICLE INFO

ABSTRACT

This paper presents a study of $CH_4/Air/H_2/O_2$ premixed laminar flames using a ratio of $H_2/O_2$ equal to 2/1. This gas mixture represents the products of the electrolysis of water. To date, only numerical analysis has been carried out on these kinds of blends and using $H_2/O_2$ percentages of only up to 10% by volume. Furthermore, there have been no reports of experimental analysis or possible flame instabilities associated with the enrichment of the mix with $H_2/O_2$. In this study, the effect of adding water electrolysis products on the laminar burning velocity and stability of lean premixed methane/air flames is studied for different percentages of $H_2/O_2$ (10 and 25) in the lean combustion regime, which is relevant for most engineering applications. The results show that a higher addition of $H_2/O_2$ to methane increases the laminar burning velocity. In particular, at $\Phi = 0.75$, intrinsic instabilities are observed for mixtures with 25% of $H_2/O_2$ in methane. With the addition of $H_2/O_2$, the laminar flame thickness decreases and the growth rate increases. Hence, the addition of $H_2/O_2$ to a premixed natural gas flame enhances the hydrodynamic instabilities.

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## 1. Introduction

Lean premixed combustion is a proven method to control $NO_x$ below 10 parts per million (ppm) and CO below 15 ppm (both corrected at 15% $O_2$ on a dry basis) without requiring the cleanup of the exhaust gases [1]. However, the same conditions that favor ultra-low emissions (i.e., low reaction temperatures < 1900 K) also push the systems to operate near the stability limits, where noise, blow off and acoustic instabilities can seriously affect the

performance and reliability of the system. Conversely, the addition of both hydrogen and oxygen to the fuel can stabilize hydrocarbon flames with fuel concentrations well below the corresponding lean blow off limit of natural gas [1,2].

To obtain the maximum decarbonization benefits, the source of the hydrogen must be considered. The production of hydrogen by the electrolysis of water can be considered a renewable process if the electrical power used for the separation of the water elements is generated via renewable sources, such as wind, sun, geothermal reservoirs or hydropower. Modern electrolytic processes are able to attain conversion efficiencies up to 95% [3]; however, the global efficiencies of the system are not as high, with values between 55% and 75% [4]. Still, electrolysis is an approach that can minimize the carbon signature of the hydrogen produced. The addition of

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hydrogen to natural gas is already being explored in Europe as a mean to store excess renewable power [5].

While the hydrogen produced from electrolysis has been studied extensively, the concomitant product, i.e., oxygen, has not received much attention. One consideration is to add the produced oxygen to an oxidizer stream (e.g., air). The addition of oxygen to air (i.e., oxygen-enriched air) has proven to significantly improve the combustibility of hydrocarbons since it reduces the energy of ignition, increases the burning velocity and adiabatic flame temperature [6]. Thus, the simultaneous addition of $H_2$ and $O_2$ enhances the reactivity of the mixture. However, the literature offers a very limited number of studies regarding the stability of natural gas flames premixed with hydrogen and oxygen-enriched air [2,7].

In light of the discussion above, this paper provides a theoretical, numerical and experimental analysis of the effect of adding water electrolysis products, i.e., both hydrogen and oxygen, on the laminar burning velocity and stability of lean premixed methane/air flames.

## 2. Background

The effects of adding water electrolysis products to gasoline [8,9] and diesel engines [10] to improve the performance have been previously studied. Regarding the effect of adding water electrolysis products to natural gas, the studies are very limited. Within the extent of the literature review, only Uykur et al. [2] and Wang et al. [11] have numerically or experimentally analyzed $CH_4 + H_2/O_2$ mixtures and their effects on burning velocity and kinetics. The present study closes the existing gap regarding the effect of the simultaneous addition of $H_2/O_2$ to $CH_4$ on the burning velocity and flame stability of lean premixed laminar flames.

Uykur et al. [2] conducted a numerical analysis of the effect of adding 10–20% $H_2$ and 10% $H_2/O_2$ (ratio of 2/1) to methane on combustion characteristics. It was found that the same effect in the laminar burning velocity ($S_L$) was obtained by mixing methane with 20% $H_2$ or with 10% $H_2/O_2$. Furthermore, it was found that adding 10–20% $H_2$ to methane did not significantly improve the flammability limits, while adding 10% $H_2/O_2$ did slightly improve this property.

Wang et al. [11] carried out an experimental study of premixed laminar methane/oxygen/argon flames doped with hydrogen at low pressure. They measured the mole fractions of $CH_3$, $C_2H_2$, $C_2H_4$, $CH_2O$, $CH_3OH$ and $C_2H_2O$ with a synchrotron photoionization technique. They found that mole fraction of hydrocarbon intermediates species decreases remarkably when increasing the hydrogen fraction. This is attributed to the increase of H and OH radicals and their high diffusivity, which promotes the chemical reactions.

Schefer et al. [12] performed a study in a swirl burner to analyze the enrichment of methane with hydrogen. Their results show that the lean blowoff stability limit is extended with the addition of hydrogen to the fuel. Further analysis of OH-PLIF data show that moderate amounts of hydrogen enrichment significantly increased the concentrations of OH radicals, which are key species for the oxidation of hydrocarbons.

Okafor et al. [13] studied the effect of hydrogen addition on methane/hydrogen/air premixed flames at a pressure of 0.1 MPa and a temperature of 350 K. The results indicate that hydrogen flames are more unstable (wrinkled) than methane flames.

Law and Kwon [14] found that when hydrogen is partially substituted with hydrocarbons the laminar burning velocity drops when compared to hydrogen/air flames. Further analysis show the flame thickness increases with hydrocarbon substitution, which diminishes hydrodynamic instabilities.

Bell and Gupta [15] found that the lean flammability limit was extended towards leaner equivalence ratios as the amount of hydrogen was increased. Similarly, Yu et al. [16] studied the effects of hydrogen addition on the fundamental propagation characteristics of methane/air premixed flames in a venting duct. They found that flame front is smooth at the early propagation stage, and when $H_2$ addition is about 50% the flame front begins to wrinkle.

Oh and Noh [17] evaluated the laminar burning velocity of oxy-$CH_4$ flames. They found that $S_L$ of oxy-$CH_4$ flames is about 7.5 times faster than that of laminar premixed methane–air flames. Their experimental data indicates that the calculations of laminar burning velocity using Gri Mech 3.0 are not in agreement with the experimental data.

Zhang et al. [18] made a numerical study about the effects of oxygen enrichment on methane/air flames. They found that the combustion enhancement is due to the promoted formation of radicals of H, OH and O, and where $H + O_2 \iff O + OH$ and $OH + H_2 \iff H + H_2O$ are the dominant reactions.

## 3. Theoretical model

### 3.1. Laminar burning velocity

Different kinetic mechanisms have been developed to simulate the reaction kinetics by considering intermediate combustion reactions. A typical mechanism includes the reaction rates expressed in the Arrhenius form. In this study, the chemical kinetics were simulated using the Gri Mech 3.0 mechanism [19], which has been optimized to reproduce the kinetics of natural gas, with 53 chemical species and 325 elementary reactions.

To carry out the numerical analysis, the PREMIX subroutine of the CHEMKIN Pro ® software [20] is used. This subroutine simulates the laminar burning velocity of a flat, stationary, one dimensional and adiabatic flame model.

### 3.2. Intrinsic instabilities

Hydrodynamic instability, diffusional-thermal instability and buoyancy-driven instability are the three main types of intrinsic instabilities which can deform a flame (wrinkling) [21]. The last type is only present at very low burning velocities and equivalence ratios near the flammability limit. Hydrodynamic instability, also called the Landau-Darrieus instability, is caused by density changes across the flame, i.e., thermal expansion; hence, it is present in any kind of flame. Moreover, thermo-diffusional instability is generated due to non-equidiffusion, which is represented by a well-known parameter, i.e., $Le$. Thermo-diffusional instability could increase flame wrinkling or attenuate hydrodynamic instability, depending on the flame thickness, non-unity Lewis number, and global activation energy [22].

Mass diffusion has an important effect on flame stability. Eq. (1) shows the analytical expression for the laminar burning velocity for a premixed lean flame, derived by Mitani [23] considering a global reaction ($v'_F F + v'_O O \rightarrow P$):

$$s_L = \left[ \frac{2\lambda_b v'_F (v'_O/v'_F)^{n_O} \rho_b^{n_F+n_O} B T_b^{\beta_1} Y_{F,u}^{n_F+n_O} Le_F^{n_F} Le_O^{n_O}}{\rho_u^2 c_p W_F^{n_F+n_O-1} \beta^{n_F+n_O-1}} \right] G(n_F + n_O)^{1/2} e^{-\frac{\beta}{2\alpha}} \tag{1}$$

$$Le = \frac{D_{th}}{D_{i,j}} = \frac{\lambda}{\rho_u c_p D_{i,j}} \tag{2}$$

$D_{th}$ is the thermal diffusivity, $D_{i,j}$ is the mass diffusion, $\lambda$ is the thermal conductivity, $B$ is a pre-exponential constant, $\beta_1$ is an exponent that depends on the fuel composition, $\rho$ is the density,