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NUMERICAL INVESTIGATION ON LAMINAR BURNING VELOCITY OF HYDROGEN-METHANE/AIR MIXTURES: A REVIEW

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Abstract

The laminar burning velocity of hydrogen/methane-air mixture is very crucial in the design and analysis of internal combustion engine and performance of combustion system if hydrogen is used as fuel. The aim of the present work was to critically analyze the validity of the available correlation to determine laminar burning velocity of hydrogen-methane-air mixture and the effect of hydrogen addition to the mixture. From the analysis, it can be said that the addition of hydrogen does change the global concentration of hydrogen-methane blend, yet the correlation is only valid for H<14 % volume. This implies that complex interaction between flame instability, chemically kinetic reaction and mass diffusivity on the global blend hydrogen-methane concentration is the main factor leading to the changes in laminar burning velocity and flammability of the fuel blending concentration.

Keywords: Laminar burning velocity, Hydrogen-methane blends, Flame instability, Chemical kinetic reaction, Le Chatelier's Rule.

1. Introduction

Nowadays, hydrogen/hydrocarbon blends are receiving substantial attention as alternative fuels in power generation and stationary systems as well as automotive industries. Hydrogen, known as an environmental friendly gas which produces only water when it is burn, has a very high combustibility and high flame speed with wide flammability limits [1-4] (LFL = 4.0% by volume, UFL = 76.5% by volume [5]). Hence, it is considerably advantaged using hydrogen to improve performance, extend operability ranges and reduce pollutant emissions of lean combustion in both stationary [6-8] and mobile [9-11] systems when it is added to hydrocarbon fuel, i.e., methane, diesel and gasoline. Such partially replacement of

Nomenclatures

A	Normalized stream tube cross-sectional area, m
CH_3	Methyl radical
C_t	Dimensionless constant
F_s	Second adjustable parameter
L_t	Turbulence macro-scale, m
M	Total mass flow rate, kg/s
NO_x	Nitrogen oxide
r	Flame radius, m
R_c	Combustion rate, J/kg
R_{min}	Minimum mass fraction among the fuel, oxygen and products
S_l	Laminar burning velocity, m/s
S_{l_CH4}	Laminar burning velocity of pure methane, m/s
S_{l_H2}	Laminar burning velocity of hydrogen, m/s
S_{l_LC}	Laminar burning velocity of Le Chatelier's Rule, m/s
S_t	Turbulent burning velocity, m/s
u_t	Turbulent intensity, m/s
v	Kinematic viscosity of unburned mixture, kg/m.s
X_{H2}	Mole fraction of hydrogen

Greek Symbols

ρ	Mixture density, kg/m^3
ρ_u	Unburned mixture density, kg/m^3
Γ	Turbulent diffusion coefficient for mass and/or energy
ϕ	Equivalence ratio

Abbreviations

LFL	Lower Flammability Limit
NTP	Normal Temperature & Pressure
UFL	Upper Flammability Limit

hydrocarbon fuel with hydrogen could be a first step toward the large-scale economical introduction of hydrogen into the energy infrastructure for stationary and for transport systems.

However, since the combustion properties of hydrogen differ in many aspects from those of hydrocarbon fuels gas, the allowable fraction of hydrogen in gas mixtures may be limited by the deteriorating performance of gas combustion equipment such as spark-ignited engines, burners and turbines to hydrogen-enriched fuel. Laminar flame speed is one of the most important physiochemical properties that characterize the combustion of a fuel. It embodies the fundamental information on diffusivity, reactivity, and it directly determines the rate of energy released during the combustion of a given combustible mixture. As reported by Law and Kwan [12], the flame speed or the rate of flame propagation is controlled by the diffusion of atoms and radical from the burned gas, and the rate at which these active species react with the unburned fuel.

Simulations of the premixed hybrid flames have been performed extensively. Most of these computations were carried out with laminar premixed flame codes such as CHEMKIN by Kee et al. [13] or COSILAB by Rogg [14], utilizing detailed kinetic schemes. Di Sarli and Benedetto [15] used the CHEMKIN PREMIX code

with the GRI kinetic mechanism to calculate the laminar burning velocities of hydrogen methane/air mixtures at NTP conditions. The air ratio and the fuel composition were varied from lean to rich and from pure methane to pure hydrogen, respectively. They investigated the effects of radical interactions, and came out with a correlation for the laminar burning velocity at different values of equivalence ratio and hydrogen content in the fuel. The results showed that the hydrogen addition enhances the methane to slightly reactive in lean mixtures, while at rich conditions, a strong inhibiting effect of the hydrogen substitution by methane was observed.

Hu et al. [16] studied the laminar burning velocity and the characteristics of premixed methane-hydrogen/air flames. In their work, sensitivity analysis and flame structure were performed. It was found that the laminar burning velocity increases with the hydrogen fraction and the peak value of the laminar burning velocity shift to the rich mixture side. Additionally, the laminar burning velocity of hydrogen-methane/air premixed flames has been experimentally measured at different values of equivalence ratio and fuel composition [17, 18]. The reported results lead to a clear conclusion that the laminar burning velocity of hydrogen/methane flames increase with increasing hydrogen fraction in the mixture. As far as the authors' concern, these simulation and chemical kinetics analysis specially focused on methane-rich flames [19-24] or hydrogen-rich flames [25], and few literatures reported on the combustion mechanism in the case of hydrogen addition over wide range of hydrogen fractions.

Previous studies in engines and flame showed that NOx concentration of methane combustion was increased as hydrogen was added especially at large hydrogen fraction [26, 27]. This could be regarded as the increase of combustion temperature when hydrogen was added. Thus, the objectives of this paper will focus on the numerical investigation on the validity of available correlations exists in determination of laminar burning velocity with various hydrogen fractions and equivalence ratios at room temperature and atmospheric pressure. This study perhaps, would give some additional understanding and data on methane-hydrogen/air mixture flames as well as provide new guidance to low emission combustion.

2. Numerical Analysis

Most works relating to calculate the laminar burning velocities of hydrogen-methane/air mixtures were carried out by means of simulations in one-dimensional, planar, adiabatic, steady, unstreched, laminar flame propagation. For instance, the Sandia PREMIX module of the CHEMKIN package was used by Di Sarli and Benedetto [15], Hu et al. [16], Wang et al. [28], and Halter et al. [29], together with the use of COSILAB code [30]. Both CHEMKIN and COSILAB adopted GRI-Mech 3.0 mechanism using a hybrid time-integration/Newton iteration technique to solve the steady-state mass, species, and energy conservation equations. GRI 3.0 consists of 325 elementary chemical reactions with associated rate coefficient expressions and thermochemical parameters for the 53 species. It includes a detailed combustion reaction mechanism for hydrogen. The GRI 3.0 mechanism has been validated by large experimental data for methane, ethane, carbon monoxide and hydrogen. Laminar burning velocity S_L was obtained from the continuity equation as shown in Eq. (1).

$$M = \rho_u S_L A \tag{1}$$

M is the total mass flow rate, ρ_u is the unburned mixture density and A is the cross-sectional area of the stream tube encompassing the flame normalized by the burner area. Cross-sectional area, A was assumed to be constant along the spatial coordinate and equal to unity.

Further, the AutoReaGas 3D code was also adopted by other researchers [31-33] to investigate the laminar burning velocities calculation, incorporating the conservation equations for mass, energy, and momentum by finite volume formulation. The turbulent flow field was described by the third model. Combustion reactions were considered as single-step conversions from reactants to products and the volume-based combustion rate, R_C, to be included in the mass conservation equation, was computed as shown in Eq. (2) from Bretislav et al. [34].

$$R_C = C_t \rho \frac{S_t^2}{\Gamma} R_{\min} \tag{2}$$

where C_t is a dimensionless constant which represents the main adjustable parameter, ρ is the mixture density, Γ is the turbulent diffusion coefficient for mass and/or energy, R_{min} is the minimum mass fraction among those of fuel, oxygen, and products. Equation (3) is the turbulent burning velocity S_t was expressed through the Bray [35] correlation.

$$S_t = 1.8 u_t^{0.412} L_t^{0.196} S_l^{0.784} v^{-0.196} \tag{3}$$

where u_t is referring to the turbulence intensity, L_t the turbulent macro-scale, S_l the laminar burning velocity, and v the kinematic viscosity of the unburned mixture. Further details of the code are reported by Van den Berg et al. [36]. A quasi-laminar modification is used for the initial laminar combustion rate. The effects of pressure, temperature and flame front wrinkling on the laminar burning velocity are described by a second adjustable parameter F_S which relates $S_{l,eff}$ to the flame radius r and to the theoretical laminar speed as described in Eq. (4) by Bakke [37].

$$S_{l,eff} = S_l (1 + F_S r) \tag{4}$$

In this study, a summarized correlation on laminar burning velocity has been listed in Table 1.

Table 1. Correlations on Laminar Burning Velocity.

Correlation	Reference	Remarks
$M = \rho_u S_L A$	Continuity Equation	M is total mass flow rate ρ_u is unburned mixture density A is cross sectional area was assumed to be constant
$S_t = 1.8 u_t^{0.412} L_t^{0.196} S_l^{0.784} v^{-0.196}$	Bray [35]	S_l is the laminar burning velocity F_S is second adjustable parameter r is radius
$S_{l,eff} = S_l (1 + F_S r)$	Bakke [37]	S_l is the laminar burning velocity $S_{l,eff}$ is quasi-laminar modification S_{l_LC} the laminar burning velocity a function of equivalence ratio and hydrogen mole fraction X_{H2} refers to hydrogen mole fraction S_{l_H2} laminar burning velocity of hydrogen S_{l_CH4} is laminar burning velocity of pure methane

3. Results and Discussion

Figure 1 shows the experimental and measured laminar burning velocity for hydrogen-methane/air mixtures at ambient condition. The computational analysis using numerical calculation mentioned in Section 2.0 are compared with literature data from Yu et al. [38], Law and Kwon [39], Hu et al. [16], Tanoue et al.[40], Hu and Huang et al. [41], Takahashi et al. [42], Sun et al. [43] and Dowdy et al. [44]. It can be said that calculated S_l is in good agreement with experimental data, except a minor error depicted when $S_l > 2$ m/s at equivalence ratio of $\phi = 1.0$ and 1.2.

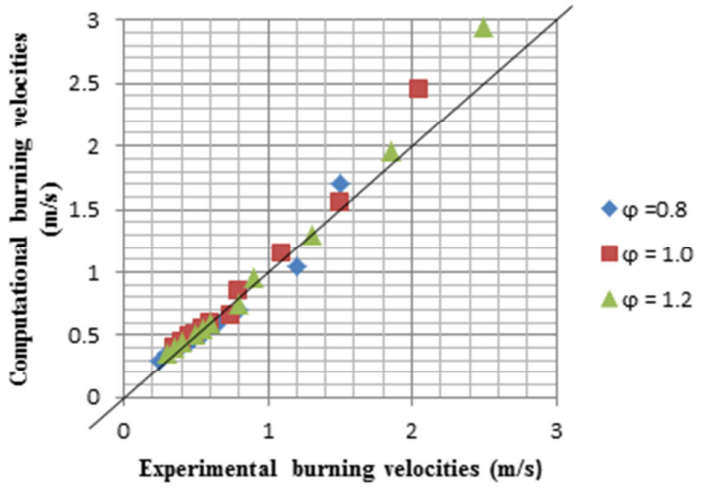

Fig. 1., Comparison between Computed Burning Velocities with GRI-Mech 3.0 and from Experiment for Methane-hydrogen-air Mixtures [45].

It worth to note that until now, correlations for evaluating the laminar burning velocity of hydrogen-methane/air mixtures as a function of equivalence ratio and fuel composition have been proposed by Yu et al. [38] and El-Sherif [24], however it was only for the low hydrogen content as illustrated in Fig. 2. Figure 2 shows the experimental and calculated data as a function of equivalence ratio. Three sources of experimental data were used for the comparison; Coppens et al. [46], who used a heat flow method in order to determine the laminar burning velocity, Halter et al. [29] who used closed vessel method while slot burner method is used by Boushaki et al. [30] work. Bare in mind, only Boushaki et al. [30] work varied the hydrogen concentration between 0% to 30% in volume. The Le Chatelier's Rule-like formula has been used in order to obtain a correlation of laminar burning velocity of hydrogen-methane/air mixtures valid even at intermediate and high hydrogen content and at different value of equivalence ratio. The Le Chatelier's Rule-like formula expressed as shown in Eq. (5).

$$S_{l\_LC}(\phi, X_{H2}) = \frac{1}{X_{H2} / S_{l\_H2}(\phi) + (1 - X_{H2}) / S_{l\_CH4}(\phi)} \tag{5}$$

S_{l_LC} is the laminar burning velocity as calculated using Le Chatelier's rule-like formula as a function of equivalence ratio and hydrogen mole fraction. X_{H2}

refers to hydrogen mole fraction; S_{l_H2} and S_{l_CH4} are laminar burning velocity of hydrogen and pure methane respectively.

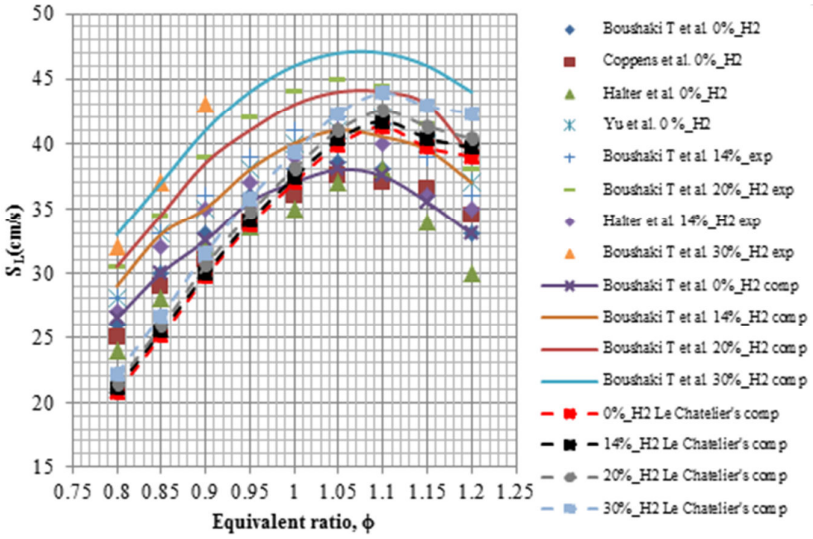

Fig. 2., Experimental and Calculated Data as a Function of Equivalence Ratio, ϕ.

It can be said that experimental data is in good agreement with calculated both using numerical equation and Le Chatelier's formula only at H < 14 % volume. When H > 14% volume was applied, the data scattered only at lean to stoichiometric concentration, none to rich concentration. Yet, similar trend can be observed; when the hydrogen content increases, the laminar burning velocities also increase. On the contrary, at rich conditions when dealing with high hydrogen (and then H radicals) content, the intermediate radical (methyl radical, CH_3) has suppressed the important H and OH radical being formed, reducing H and OH radical in reaction zone. This leads to the slower ignition and affects the S_l as well as pressure development.

In addition, the experimental result from Boushaki et al. [30] shows very high laminar burning velocity compared to the computational result, as the flame starts to lose its triangular shape due to flame instabilities. Knowing the behaviour of the hybrid mixtures laminar burning velocity is very complex, and without a detailed investigation on its global concentration, the laminar burning velocity may be obtained from the values of the individual constituents at the same conditions by varying the hydrogen content in the blend [15]. This implies that the complex interaction of flame instability, chemically kinetic reaction and mass diffusivity on the global blend hydrogen-methane concentration should be explored more in order to follow Le Chatelier's rule-like formula.

4. Conclusions

A review was carried out to investigate the validity of available correlations in determination of laminar premixed hydrogen-methane/air flames. Le Chatelier's

law was applied unstretched laminar burning velocity at adiabatic flame temperature. The main conclusions are summarized as follows:

- Laminar burning velocity is increased with the increase of hydrogen fraction. In the case hydrogen fraction less than 14 % volume, the correlation adopted follows Le Chatelier's rule but not when H > 14% volume. This is due to enhancement of chemical reaction with hydrogen addition at lean to stoichiometric because of the increase of H, O and OH concentrations in the flame as hydrogen is added. When hydrogen fraction is larger than 14 % volume, a decrease in laminar burning velocity is with the increase of hydrogen fraction.
- The standard format is followed A strong correlation exists between laminar burning velocity and equivalence ratio of hydrogen-methane/air blends. An exponential pattern is observed when fuel is increased.
- It can be said that complex interaction of flame instability, chemically kinetic reaction and mass diffusivity on the global blend hydrogen-methane concentration should be explored more in order to follow Le Chatelier's rule for rich mixtures hydrogen-methane blends.

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