# Effect of Additional Diluents on Laminar Burning Velocities and Cellular instability in Outwardly Propagating Methane/Ethylene-Air Premixed Spherical Flame

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## 1. Introduction

The tight environment regulation as well as increasing energy consumption was caused by the overusing fossil fuels, global warming, and depletion of energy sources. One reasonable solution on the above problems is to reduce NO<sub>x</sub> emissions and to develop lean-premixed combustors. Burning lean hydrocarbon-air mixtures offers the potential of reduced NOx emissions according to the thermal mechanism [1], and lean flammable limit was greatly extended with enrichment of  $H_2$ [2-4]. In downstream interaction between lean premixed methane-air and hydrogen (or H<sub>2</sub>/CO syngas)-air flames, lean flammable limits were significantly extended while the extinction behaviors were complicatedly influenced, e.g. preferential diffusion of H and H<sub>2</sub> as well as complicated chemical effects of sharing the radicals such as H, O, and OH on extinction behavior in interacting flames [2-4]. The effects of additional reformate gas on flame extinction in CH<sub>4</sub>-air were examined and it was shown that enrichment of reformate gas greatly reduced NO formation [5]. Nonetheless, applying a variety of such low calorific fuels to a combustor requires to explore fundamental flame characteristics such as flame stability and instability, ignition and extinction, and pollutant emissions. To particularly grasp the flame instability, several studies have been conducted to estimate the effect of the diluents on the combustion [6-9]. For stoichiometric  $H_2/air$  mixture, diffusivethermal effects in cellular instability was suppressed greatly by helium addition to the mixture [10]. Vu et al. studied the effects of diluents, such as  $CO_2$ ,  $N_2$ , and He, on cellular instability in syngas-air premixed flames. The results showed that diffusional-thermal and hydrodynamic instability was suppressed by adding diluents to the syngas-air flame [11]. However, the effect of additional diluents on  $CH_4/C_2H_4/air$  premixed flame have been hardly found in the literature. Particularly to secure the fuel flexibility and to evaluate flame stability, Diluents (CO<sub>2</sub> and He)-added CH<sub>4</sub>/C<sub>2</sub>H<sub>4</sub>/air premixed flames were examined and unstretched laminar burning velocities, Markstein length, and cellular instability with diluents at elevated pressures (up to 4 atm) were evaluated in the current study.

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#### 2. Experimental setup

Experiments were conducted in a stainless steel, cylindrical constant volume chamber (CCVC) having 200 mm in i.d. and 220 mm in length, as shown in Fig. 1. For optical access, two quartz windows with 100mm in diameter and 40 mm in thickness were installed to mount oppositely. Opposed tungsten electrodes were connected to ignite combustible mixture with high voltage source (8  $\sim$  9 kV) at the center of the chamber. The electrodes were 0.5 mm in thickness and the gap between them was adjustable. Methane and ethylene with a purity of 99.99 % and air with that of 99.95 % were used, respectively. Fuels and air were supplied to the CCVC in a manner that the partial pressure of each reactant was adjusted by a pressure transmitter (AEP Lab DMM, -1-40 bar,  $\pm 0.5$  % FSO) to achieve the desired initial chamber pressure. To ensure complete mixing of supplied reactants to the CCVC and their guiescence, the experiment was conducted after 15 minutes elapsed. After the flame propagation was completed, the CCVC was ventilated to the laboratory exhaust system and purged with an air compressor to get rid of condensed water for the next experiment. The propagating spherical flame was visualized using a Schileren sysytem with a 300-W xenon light source, a pair of 150 mm diameter concave mirrors, and recorded using a high-speed digital camera (Olympus i-Speed3) operating at 10,000 frames per second with 528×400 pixels of resolution. The temporal chamber pressure during ignition and flame propagation was acquired by a dynamic pressure transducer (Kistler 601A, 0-250 bar) and a data logger (GRAPHTEC GL900).

### 3. Results and Discussion

For outwardly propagating spherical premixed flame, the burnt stretched flame propagation velocity,  $S_b$ , with respect to burned mixture is calculated by the instantaneous flame radius,  $R_f$ , measured from a Schlieren image, as  $S_b = dR_f/dt$ , where t denotes the time. The flame stretch rate, K, in an outwardly propagating spherical premixed flames is expressed as

$$K = \frac{1}{A}\frac{dA}{dt} = \frac{2}{R_f}\frac{dR_f}{dt} = \frac{2}{R_f}S_b \tag{1}$$



Figure 1. Schematic of experimental setup and schlieren system.



Figure 2. Measured and predicted Laminar burning velocities in  $CH_4/C_2H_4$ -air premixed flames diluted with  $CO_2$  (a) and He (b) at  $P_u = 1$  atm.

The experimental methodology was validated through comparing unstretched laminar burning speeds measured at various flame radius ranges using three kinds of extrapolation models with numerical ones from using Premix Code with GRI v-3.0 in methane-air premixed flames [12]. Based on them, the most reliable detailed reaction mechanism to methane/ethylene-air premixed flame was examined through comparison among GRI v-3.0,USC Mech II, and Sung Mech. These results showed that experimental unstretched laminar burning velocities using the nonlinear extrapolation model (in eqn. (2)) [13, 14] in range of 10-25 mm in flame radius in  $CH_4/C_2H_4/$  - air premixed flame were best-fitted to predicted ones with USC Mech II [15].

$$\ln(S_b) = \ln(S_b^0) - S_b^0 L_b \times (\frac{2}{R_f S_b})$$
(2)

Experimental results for Su<sup>0</sup> obtained with NM in diluents-added CH<sub>4</sub>/C<sub>2</sub>H<sub>4</sub> - air premixed flame were compared with numerical ones, and exhibited in Fig. 2. Here, the diluents volume fraction is defined by  $\Omega_{diluents} = X_{c_2H_4} / (X_{CH_4} + X_{c_2H_4} + X_{diluents}) \text{ where } X_i \text{ is the mole fraction of species } i (i = C_2H_4, CH_4, CO_2 \text{ or } H_2).$ As shown in Fig. 2, the maximum deviation between experimental and numerical  $S_u^0$  appears at the equivalence ratio ( $\phi$ ) of 1.4. It is seen that, at fuel-rich condition, the errors with simulation were larger than those with fuel-lean codition. Further investigation colud be required to clarify the reason on the discrepancy between experiment and simulation. The results using the OPPDIF [16] and PREMIX codes [17] were also compared because adiabatic unstretched laminar flame speed cannot obtained due to high thermal conductivity (for He) and high radiative heat loss (for CO<sub>2</sub>) in He- and CO<sub>2</sub>-added premixed flames. As shown in Fig. 3, both predicted unstretched laminar burning speeds in He- and CO<sub>2</sub>-added premixed flames are reasonably in agreement with experimental ones at  $0.6 \le \phi \le 1.0$ . However, in such lean cases, the predicted results with OPPDIF are better-fitted to experimental data. However, in rich cases ( $\phi = 1.2$  and 1.4), both numerical results underpredict experimental data appreciably. The deviation increased in increase of  $\Omega_{co_2}$ , e.g. 59 % for OPPDIF at  $\phi = 1.4$  and  $\Omega_{He} = 0.1$  in CO<sub>2</sub>-added flames and 37.27 % for OPPDIF and 43.16 % for PREMIX code at  $\phi = 1.4$  and  $\Omega_{\mu} = 0.1$  in He-added flames. Note that this tendency is very similar to that in CH<sub>4</sub>-air premixed flame [12]. Further detailed error analysis is required and will be a future work. Markstein length cahracterizaes the effect of local heat release of a propagating flame on cariation in the surface topology along the flame and the associated local flame front curvature. If the burnt Markstein length  $L_b > 0$ , the flame speed decreases with flame stretch (thereby-stabilized), and vice versa [1, 18, 19]. Burnt Markstein length (extrated from NM) as a function of diluent



Figure 3. Marstein length against diluents volume fraction in CH<sub>4</sub>/C<sub>2</sub>H<sub>4</sub>-air premixed flames diluted with CO<sub>2</sub> and He for  $\phi = 0.8$  and 1.2 at  $P_u = 1$  atm (left) and at various initial pressures for  $\phi = 0.8$  (right).



Figure 4. Effective Lewis number against  $\phi$  in CH<sub>4</sub>/C<sub>2</sub>H<sub>4</sub>-air premixed flames diluted with CO<sub>2</sub> (a) and He (b)

concentration  $\Omega_{diluents}$  in diluents-added CH<sub>4</sub>/C<sub>2</sub>H<sub>4</sub>-air mixture at  $P_u = 1$  atm for  $\phi = 1.4$  are demonstrated in Fig. 3. The tendency of  $L_b$  increase (decrease) with increasing diluents volume fraction (equivalence ratio), implying that increase of additional diluents volume fraction stabilizes the flame. The results also show that the Markstein lengthe in He-added cases are lower than those in CO<sub>2</sub>-added cases. Formation of cellular structure over flame surface is attributed to imbalance of mass and thermal diffusion and hydrodynamic instability in case that buoyancy effects are negligible. Figure 4 shows variations of  $Le_{eff}$  against equivalence ratio in CH<sub>4</sub>/C<sub>2</sub>H<sub>4</sub>-air premixed flames diluted with CO<sub>2</sub> (left) and He (right) at various diluents volume fractions. The results show that effective Lewis numbers are larger than unity at  $0.6 \le \phi \le 1.0$  for all diluents volume fractions and diffusional-thermal instability can be thus suppressed sufficiently. Then, cellular instability is addressed to hydrodynamic effects, which is mainly affected by by flame thickness,  $l_f$ , and



Figure 5. Effects of CO<sub>2</sub> addition on cellular instability in CH<sub>4</sub>/C<sub>2</sub>H<sub>4</sub>/air premixed flame at  $\phi = 0.8$ .

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thermal expansion ratio,  $\sigma$ , as identified in the Baroclinic torque term in vorticity equation, and is investigated in Figs. 5 and 6. As both diluents volume fraction increase, the effective Lewis numbers are always larger than unit. Thus the cellular formations are not addressed to diffusive-thermal instability. In both CO<sub>2</sub>- and He-added cases, hydrodynamic instabilities are suppressed because  $\sigma(l_f)$  decreases (increases) in increase of CO<sub>2</sub> volume fraction.

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