The Effects of Hydrogen Enrichment on Propagation Speeds in Adiabatic Flat and Cellular Premixed Flames of $CH_4 + O_2 + CO_2$

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1 Introduction

CO_2 diluted oxy-fuel combustion is currently considered as one of the promising technologies facilitating CO_2 separation and eventual sequestration. Modern concepts of Integrated Gasification Combined Cycle systems also include production of a syngas rich with hydrogen. Mixing with natural gas then yields complex mixtures composed of hydrogen, methane, oxygen and carbon dioxide. The goal of the present study was investigation of the effects of hydrogen enrichment on propagation speeds in adiabatic flat and cellular premixed flames of $CH_4 + O_2 + CO_2$. Experimental measurements of the adiabatic burning velocity of laminar flat flames of $CH_4 + O_2 + CO_2$ and $C_2H_6 + O_2 + CO_2$ were reported [1, 2]. Adiabatic stabilization on flat flame burners is attractive for laminar premixed planar flame studies since it facilitates comparison with theoretical models. Therefore a Heat Flux method was used to determine flame propagation speeds under conditions when the net heat loss of the flame is zero. Under specific experimental conditions the flames become cellular; this leads to significant modification of the flame propagation speed [2, 3]. Experimental observations in methane, ethane and propane flames have been summarized recently [4]. These results received substantial attention; technical questions raised by the reviewers and readers of these papers [2-4] called for further quantification of the flames studied. In the present work it was attempted to address the following issues: (1) limiting propagation speed of the cellular flames when approaching room temperature of the burner surface, and (2) correlations of the propagation speeds and mean cell diameter with the amount of hydrogen in the fuel.

2 Experimental and Modeling Details

The experimental set-up for stabilizing an adiabatic flame using the Heat Flux method has been described elsewhere [1-3]. The burner consisted of the burner head mounted on a plenum chamber. The 2 mm thick burner plate perforated with small holes was attached to the burner outlet. The diameter of the burner mouth was 30 mm. The burner head had a heating jacked supplied with thermostated water to keep the temperature of the burner plate constant. In the present experiments this temperature was varied from 303 to 368 K. Pure methane and two mixtures containing 15 and 35 vol. % of hydrogen in the fuel were studied in the present work. In the following parameter α is defined as the mole fraction of hydrogen in the methane + hydrogen mixtures: $\alpha = H_2/(CH_4+H_2)$. The oxygen content in the oxidizer $D = O_2/(O_2+CO_2)$ was chosen equal to 0.3155 to be compatible with the previous studies [1-4].

A detailed C/H/N/O reaction mechanism for the combustion of small hydrocarbons was used for the modeling [5]. The current version of the mechanism (Release 0.5) consists of 1200 reactions among 127 species. The

CHEMKIN - II collection of codes, including transport properties from Sandia National Laboratories, were used. Multi-component diffusion and thermal diffusion options were taken into account. Adaptive mesh parameters were GRAD = 0.05 and CURV = 0.5; total number of grid points was typically 350 – 400.

3 Results and Discussion

Propagation speeds in (CH_4 + H_2) + O_2 + CO_2 flames with dilution ratio D = 0.3155 at atmospheric pressure and standard temperature of the fresh mixtures (298 K) for different hydrogen content in the fuel are shown in Figs. 1 - 3. In agreement with the previous observations [2-4] temperature of the burner plate and mixture composition define the appearance of a flame. Lowering the temperature of the burner plate extends the range of equivalence ratios over which the cellularity was observed. Increasing the temperature of the burner plate eliminates this instability. When this temperature was fixed at 368 K, the flame stabilized closer to its surface, making the flame perfectly flat [1, 2]. Then the adiabatic burning velocity in flat flames could be determined.

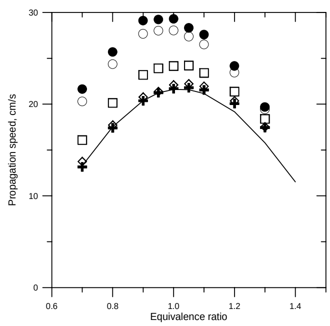

Fig. 1. Propagation speeds in CH_4 + O_2 + CO_2 flames. Crosses: the burner plate temperature = 368 K, diamonds: T = 343 K, squares: T = 323 K, open circles: T = 303 K, solid circles: extrapolated to 298 K, line: adiabatic burning velocity.

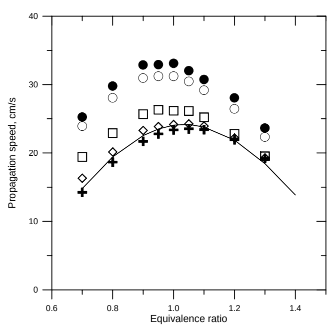

Fig. 2. Propagation speeds in (CH_4 + H_2) + O_2 + CO_2 flames with hydrogen content α = 0.15. Crosses: the burner plate temperature = 368 K, diamonds: T = 343 K, squares: T = 323 K, open circles: T = 303 K, solid circles: extrapolated to 298 K, line: adiabatic burning velocity.

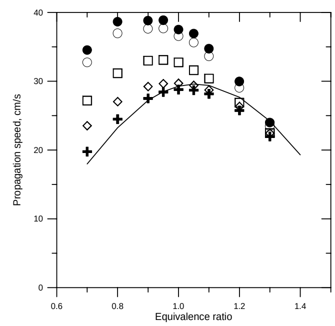

Fig. 3. Propagation speeds in (CH_4 + H_2) + O_2 + CO_2 flames with hydrogen content α = 0.35. Crosses: the burner plate temperature = 368 K, diamonds: T = 343 K, squares: T = 323 K, open circles: T = 303 K, solid circles: extrapolated to 298 K, line: adiabatic burning velocity.

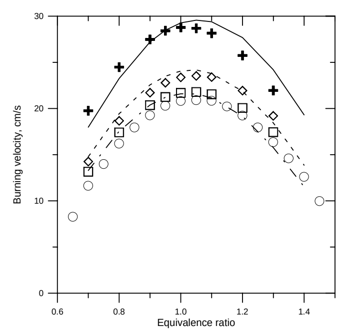

Fig. 4. Laminar burning velocities of (CH_4 + H_2) + O_2 + CO_2 mixtures. Symbols: experiment, lines: modeling. Crosses and solid line: α = 0.35; diamonds and dashed line: α = 0.15; circles [1], squares (present work) and dash-dot line: α = 0.

Figure 4 summarizes laminar burning velocities of (CH_4 + H_2) + O_2 + CO_2 mixtures with dilution ratio D = 0.3155 for different hydrogen content in the fuel. Earlier measurements in CH_4 + O_2 + CO_2 flames [1] are also shown. They were found in good agreement with the present results taking into account typical uncertainty of the burning velocity of about ± 1 cm/s. Also shown in Figs 1-4 are the calculated burning velocities which reproduce experimental variations with equivalence ratio and hydrogen content in the fuel, α, from 0 till 0.35.

Adiabatic propagation speeds of cellular flames are systematically higher than those in laminar flat flames; the difference is more pronounced at lower temperatures of stabilization. Decreasing the temperature of the burner plate led to increase of the propagation speeds. Due to the principle of the flame stabilization using the Heat Flux method it was impossible to stabilize the flames when the temperature of the burner plate is equal to the temperature of the fresh mixture (298 K). However, in the present work the lowest temperature was quite close (303 K). To extrapolate the measured propagation speeds toward standard conditions and thus to mimic freely propagating cellular flames the variations of the speeds with the temperature were approximated by the fourth-order polynomial functions. These extrapolated values are also shown in Figs. 1 – 3. The measurements presented in Figs. 1 – 3 thus cover a complete range of propagation speeds from laminar burning velocity to quasi-freely propagating flames. To quantify the flame acceleration due to cellularity, the ratios of the propagation speed extrapolated to 298 K to the laminar burning velocity in (CH_4 + H_2) + O_2 + CO_2 flames were determined and plotted in Fig. 5 for different hydrogen content in the fuel. One can conclude that within the experimental uncertainties all mixtures behave similarly; no influence of the hydrogen enrichment could be elucidated.

Visual and photographic observations of the flames were performed to quantify their cellular structure. Figure 6 shows the number of cells observed in (CH_4 + H_2) + O_2 + CO_2 flames with hydrogen content α = 0.35. In agreement with the previous observations for different fuels, CH_4, C_2H_6 and C_3H_8, [2-4] increasing the temperature of the burner plate led to increase of the number of cells observed. Figure 7 shows the number of cells observed in (CH_4 + H_2) + O_2 + CO_2 mixtures at the burner plate temperature = 303 K for different hydrogen content in the fuel. Enrichment by hydrogen clearly affects appearance and the number of cells observed.

Mean cell diameter was computed by dividing the burner diameter (30 mm) by the square root of the number of cells. Mean cell diameters in lean (CH_4 + H_2) + O_2 + CO_2 flames (Φ = 0.8) with dilution ratio D = 0.3155 as a function of hydrogen content, α, are shown in Fig. 8. The trends observed at different temperatures of the burner plate look similar and show that hydrogen enrichment leads to decrease of the mean cell diameter.

4 Conclusions

The results obtained in the present work in (CH_4 + H_2) + O_2 + CO_2 mixtures are in good accordance with the previous observations for different fuels, CH_4, C_2H_6 and C_3H_8, [2-4]. The effects of hydrogen enrichment on propagation speeds in adiabatic flat and cellular premixed flames of CH_4 + O_2 + CO_2 have been experimentally studied and computationally confirmed for flat laminar flames. The enrichment by hydrogen leads to:
- the increase of the laminar burning velocities;
- the increase of the number of cells observed;
- the decrease of the mean cell diameter.

The flame acceleration due to cellularity was not affected by the hydrogen enrichment.

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References

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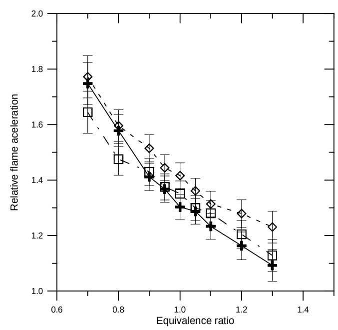

Fig. 5. Ratio of the propagation speed extrapolated to 298 K to the laminar burning velocity in (CH_4 + H_2) + O_2 + CO_2 flames. Crosses and solid line: α = 0.35; diamonds and dashed line: α = 0.15; squares and dash-dot line: α = 0.

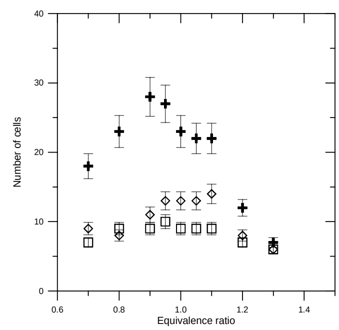

Fig. 7. Number of cells observed in (CH_4 + H_2) + O_2 + CO_2 mixtures. The burner plate temperature = 303 K. Crosses: α = 0.35; diamonds: α = 0.15; squares: α = 0.

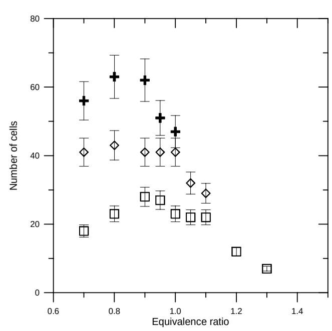

Fig. 6. Number of cells observed in lean (CH_4 + H_2) + O_2 + CO_2 flames (Φ = 0.8) with hydrogen content α = 0.35. Crosses: the burner plate temperature = 343 K, diamonds: T = 323 K, squares: T = 303 K.

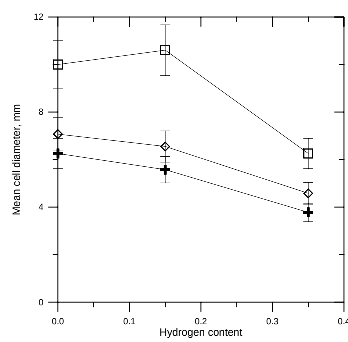

Fig. 8. Mean cell diameter in (CH_4 + H_2) + O_2 + CO_2 flames as a function of hydrogen content α. Crosses: the burner plate temperature = 343 K, diamonds: T = 323 K, squares: T = 303 K.