

LAMINAR BURNING SPEED AND FLAME STRUCTURE OF SYNGAS/OXYGEN/HELIUM AND SYNGAS/AIR/EDG PREMIXED FLAME

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ABSTRACT

Experimental studies have been performed in conjunction with a novel differential based multi-shell model to investigate the flame structure and measure laminar burning speed of H₂/CO/oxidizer/diluent premixed flames at high pressures. The laminar burning speed of the combustion process is extracted from the pressure rise during flame propagation. This paper focuses on synthetic gas (syngas) as the fuel, which is a mixture of H₂ and CO, and investigates the effect of dilution with helium and EDG (Extra diluent gases), with a composition of 14% CO₂ and 86% N₂. In this experiment three different EDG concentration (0%, 5% and 10%) has been used. The experiments were performed in two constant volume spherical and cylindrical chambers. The cylindrical chamber was set up in a Z-shaped Schlieren shadowgraph system equipped with a high speed CMOS camera, capable of taking pictures up to 40,000 frames per second, that was used to study the stability of the flame. The experiments have been done for laminar burning speeds of smooth H₂/CO/O₂/He and H₂/CO/air/EDG flames over a wide range of temperatures (298K up to 500K), pressures (from sub-atmospheric up to 6atm), equivalence ratios (0.6-3) and three different hydrogen concentration of 5%, 10% and 25% respectively. Extra diluent gases (EDG) lower the laminar burning speeds but do not have significant effect on the stability compared to syngas/air in the stoichiometric case. Substitution of nitrogen in the air with helium, exactly with the same percentage, increases the range of temperature and pressure of stable flame as well as the laminar burning speed.

INTRODUCTION

Synthetic gas, also known as syngas, is primarily a mixture of hydrogen and carbon monoxide gas along with various other higher-order hydrocarbons. Syngas is considered an alternative fuel since it can be created through various sources such as biomass gasification, reactions that involve natural gas and coal, as well as the recycling of stationary turbine byproducts. With the advent of integrated gasification combined cycle (IGCC) technology, syngas can be created from coal cleaner with lower emissions as well as creating carbon free fuel. Thus, the development and research pertaining to syngas fuels are becoming more relevant amid growing concerns about pollutants and carbon emissions.

Syngas is considered as a strong candidate to replace many fuels currently in use, therefore it is imperative to fully understand and characterize how syngas behaves in various conditions. The laminar burning speed adequately characterizes

a fuel and provides a good indicator of how a fuel performs. It is widely used and contains information about a mixture's exothermicity, diffusivity, and reactivity. It is also important to study the laminar burning speed in a high pressure environment as well as with different diluents, since those are normally gas turbine-relevant conditions. One typical form of diluent is the inert gas used in exhaust gas recirculation (EGR) technique commonly used in automobile engines, which is primarily a mixture of carbon dioxide and nitrogen and other products of combustion.

There is a wide assortment of literature on the laminar burning speed of syngas fuels with and without diluent. Hassan et al. [1] measured the laminar burning velocities of various hydrogen to carbon monoxide ratios (3:97, 5:95, 10:90, 25:75, 50:50), sub-atmospheric to elevated pressures (0.5-4 atm), atmospheric temperature, and wide equivalence ratio (0.6-5.0) in a spherical combustion chamber. Sun et al. [2] used a dual-cylindrical chamber to extract laminar burning speed data at atmospheric temperature from many different H₂/CO ratios (1:99, 5:95, 25:75, 50:50), elevated pressures (up to 40 atm), and equivalence ratios (0.5-5.0). Sun et al. also replaced nitrogen with helium as the diluent in order to increase the stability of flames, which allowed them to obtain data for much higher pressures. Burke et al. [3] provided an in-depth study that explored the differences between experimenting in a spherical chamber versus a cylindrical chamber, and recommended that researchers using the constant-pressure methodology of extracting laminar burning speed restrict their data to less than 30% of the radius of the inner cylindrical wall. Vu et al. [4] compared the effects of CO₂, N₂, and He as diluents on the cellular instabilities in syngas flames in a cylindrical chamber at elevated pressures for a 50:50 hydrogen to carbon monoxide ratio, and found that He suppresses instabilities best and reduces the laminar burning speed the least. However, Vu et al.'s range of equivalence ratios was limited to slightly lean to slightly rich (0.8-1.4). Burbano et al. [5] used the burner method to extend the data on the effects of CO₂ and N₂ dilution on laminar burning speed and stability over a wider equivalence ratio (0.6-4.3). Lapalme and Seers [6] investigated the effect of initial temperature (up to 450K) as well as carbon dioxide and methane dilution on the laminar burning velocities of syngas flames, as well as provided a correlation based on their data. Han et al. [7] measured laminar burning velocities for various CO₂ diluent percentages (10%-40%) at elevated temperatures and pressures for equivalence ratios of $\phi = 0.8$ and $\phi = 1.0$ using a dual-cylindrical setup. Askari et al. [8] measured the laminar burning speeds of H₂/CO/air flames using a new differential-based multi-

shell model over a wide range of temperatures (298K up to 617K), pressures (from sub-atmospheric up to 5.5atm), equivalence ratios (0.6-5) and three different hydrogen concentration of 5%, 10% and 25% respectively. They concluded when the initial pressure increases, the propensity tendency for the flame to destabilize takes place earlier due to a significant decrease of the flame thickness and enhancement of hydrodynamic instability. Laminar burning speeds for smooth flames were measured using the pressure rise method and Power law correlations have been developed for laminar burning speeds of smooth $H_2/CO/air$ flames.

This present study investigates the effect of diluent type on the stability and laminar burning speeds of $H_2/CO/O_2/He$ and $H_2/CO/air/EDG$ flames which extends available data and creates new data in literature. The effect of EDG (extra diluent gases) addition (5% and 10%) to $H_2/CO/air$ on flame structure will be studied in a wide range of temperatures, pressures and equivalence ratios. After the flame structure study, laminar burning speeds of $H_2/CO/air/EDG$ and $H_2/CO/O_2/He$ will be reported over a wide range of temperatures, pressures, equivalence ratios and three different hydrogen concentration of 5%, 10% and 25% respectively. It will be shown that laminar burning speeds of $H_2/CO/O_2/He$ cover a higher range of temperatures and pressures (compared to $H_2/CO/air$ mixtures) which can be useful for the validation of chemical kinetics mechanisms at high temperatures and pressures.

EXPERIMENTAL FACILITIES

Experiments have been performed using a spherical vessel for laminar burning speed measurement and a cylindrical vessel in a Schlieren system to study flame shape and structure. The cylindrical chamber is 13.5 cm in diameter and 13.5 cm in length. The cylindrical vessel is equipped with Quartz windows that are sealed to the chamber with two high temperature Parofluor O-rings. There are two band heaters to increase the initial gas temperature of cylindrical vessel up to 500 K. Both vessels are fitted with two extended automotive spark plugs, and K-type thermocouples to measure the temperature of the inside gas mixtures. The spark energy has been tuned to be sufficiently close to the minimum ignition energy to minimize the effect of spark discharge on flame expansion [9]. Figure 1 shows the general configuration of the experimental set up. The spherical vessel is made of stainless steel which can withstand pressures up to 400 atm. The spherical vessel is constructed from two hemispheres with a diameter of 15.24 cm. It is located in an oven which can heat up the vessel up to the initial temperature of 500 K. The pressure rise inside the spherical vessel was measured using a Kistler high sensitivity pressure sensor [10]. To be able to take optical recordings of the combustion event the cylindrical vessel was installed in a Schlieren Shadowgraph system equipped with a high speed CMOS camera, capable of taking pictures up to 40,000 frames per second [11,12]. The vessel was filled by the method of partial pressures using a manifold supply system comprised of valves, high accuracy pressure transducers, pipes connected to the respective mixture constituents, and a vacuum pump. A gas chromatography (GC) system was used to verify the composition of premixed fuel inside the vessel. A data

acquisition system was utilized to record the pressure-time data as well as the flame propagation images. A LabView program has been written to find the exact initial composition and to initiate the combustion process. After filling the chamber with fuel, oxidizer, and diluent the system is given at least 5 min to make sure that the mixture is quiescent in the chamber. At each operational condition experiments were done using both chambers. First, the cylindrical chamber was used to study the shape and structure of the flame and then the same experiments were performed in the spherical chamber to collect the pressure rise data. Only the portion of the pressure rise data where the flame is completely smooth, laminar, and spherical has been used to calculate the laminar burning speed. In this work each experiment was carried out at least three times at each working condition to ensure that the confidence level of the experiments was above 95% [13]. More information about experimental facilities can be found in other studies [10–12,14–16].

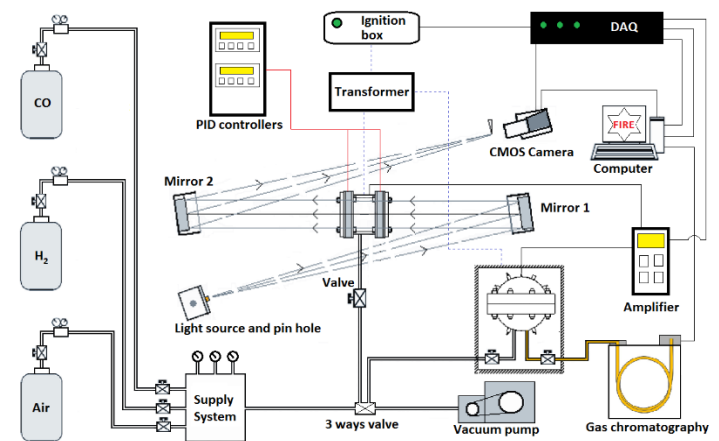


Figure 1 Three external boundary condition types

EXPERIMENTAL RESULTS AND DISCUSSION

Experiments with extra diluent gases (14% CO_2 + 86% N_2) have been done with two different volumetric percentages of 5% and 10%. The initial conditions of the experiments were fixed at atmospheric initial temperature of 298 K, initial pressures of 0.5, 1, and 2 atm, equivalence ratios of 0.6, 1, 2 and 3 and three different hydrogen concentration of 5%, 10% and 25%. Figure 2 shows the snapshots of the expanding spherical flame with changing volumetric percentages of diluents at hydrogen concentration of 25%, initial temperature of 298 K, initial pressure of 2 atm, equivalence ratio of 3 and different flame radius. As it can be seen in Figure 2 addition of EDG increases the flame stability. This behaviour is much more obvious at $\phi \geq 2.0$. The reason is due to increase the flame thickness which promotes the flame stabilities as shown in Figure 3. Figure 4 shows a series of snapshots of $H_2/CO/O_2/diluents$ (N_2 and He) flames for equivalence ratio of 2.0, initial pressure of 2 atm, initial temperature of 298 K and different hydrogen concentration of 5, 10 and 25%. The onset of cellularity is shown for the case of nitrogen as diluent. As it can be seen the tendency for cell formation is increased with increasing hydrogen

concentration in the fuel blend. In this case flame becomes much more sensitive to instability which causes the cellularity takes place in smaller radius. As it obvious from Figure 4 substituting nitrogen with helium makes the flame much more stable and smooth by delaying the cellularity. This can be explained by the increase of Lewis number and flame thickness comparing to nitrogen as shown in Figure 5 which weakens the impact of thermo-diffusive and hydrodynamic instabilities, respectively. As it can be seen in Figure 6 the critical Peclet number for helium addition is less than nitrogen case which means that the instability happens later.

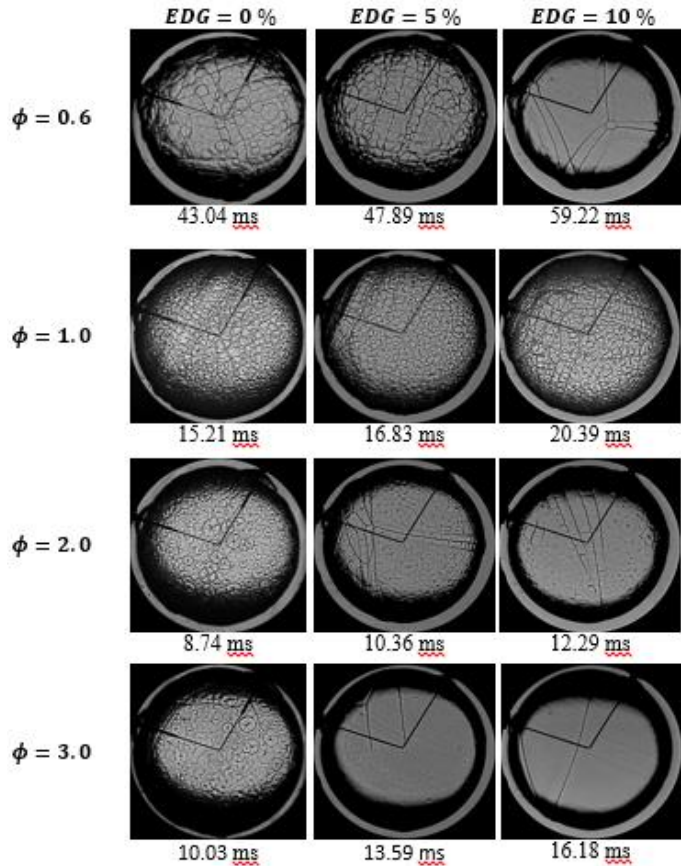


Figure 2 Snapshots of the H₂/CO/air/EDG flames for various EDG percentages and equivalence ratios at hydrogen concentration of 25%, initial temperature of 298 K, initial pressure of 1 atm

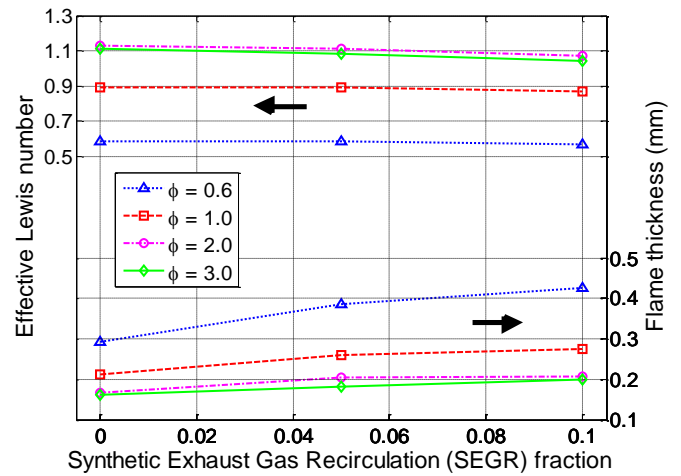


Figure 3 Effective Lewis number and flame thickness of the H₂/CO/air flames corresponding to the snapshots of Figure 2

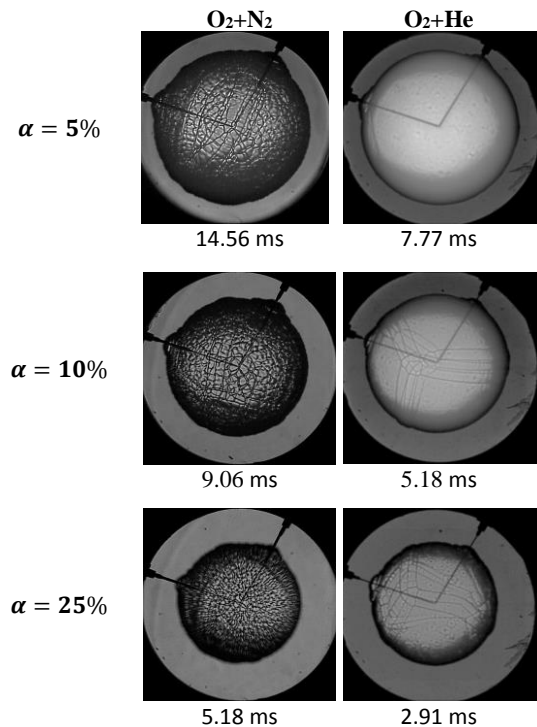


Figure 4 Snapshots of the H₂/CO/O₂/diluent flames for two different diluents of Nitrogen and Helium, various hydrogen concentration at equivalence ratio of 2, initial temperature of 298 K and initial pressure of 2 atm

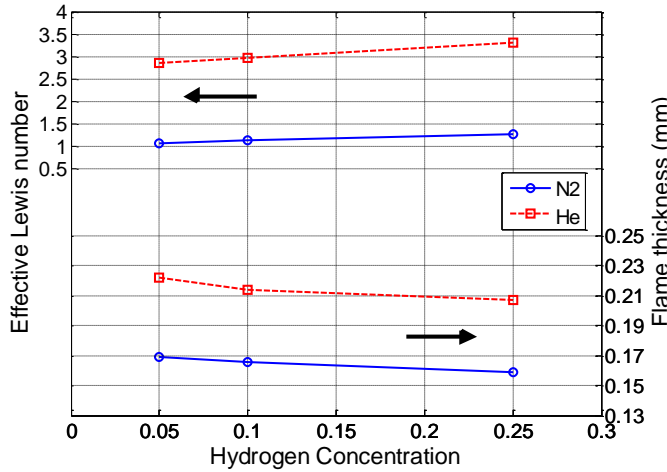


Figure 5 Effective Lewis number and flame thickness of the $H_2/CO/O_2$ /diluents (N_2 or He) flames corresponding to the snapshots of Figure 4

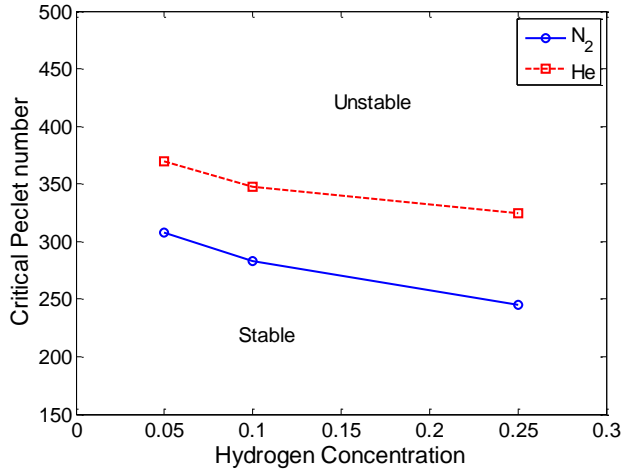


Figure 6 Critical Peclet number of the $H_2/CO/O_2$ /diluents (N_2 or He) flames for various hydrogen concentration at initial pressure of 2 atm, initial temperature of 298 K and equivalence ratio of 2.0

STRETCH EFFECT INVESTIGATION

Flame stretch in spherically expanding flames can be defined as:

$$\kappa = \frac{1}{A_f} \frac{dA_f}{dt} = \frac{2}{r} \frac{dr}{dt} \quad (1)$$

where κ is the stretch rate, A_f is the flame front area, t is time, and r is the flame radius. In order to study the effect of stretch, laminar burning speeds of $H_2/CO/O_2/He$ mixtures have been measured at different stretch rates and flame radii ($r > 4\text{cm}$) with similar unburned gas properties such as temperature, pressure and equivalence ratios. To perform these experiments, different tests have been arranged by changing the initial

temperature and pressure of the mixtures along specific isentropic lines. More information about this method can be found in previous publications [15,17]. Figure 7 shows the variation of laminar burning speed versus stretch rate for different equivalence ratios and unburned gas conditions at hydrogen concentration of 5%. As it can be seen the laminar burning speeds do not change for flame radii greater than 4 cm and stretch rates lower than 80 s^{-1} . Based on many studies in the literature [18–21] the stretch rates higher than 100 s^{-1} can have significant effect on the laminar burning speeds. Therefore, in this paper all the laminar burning speed data are reported for stretch rates of less than 80 s^{-1} .

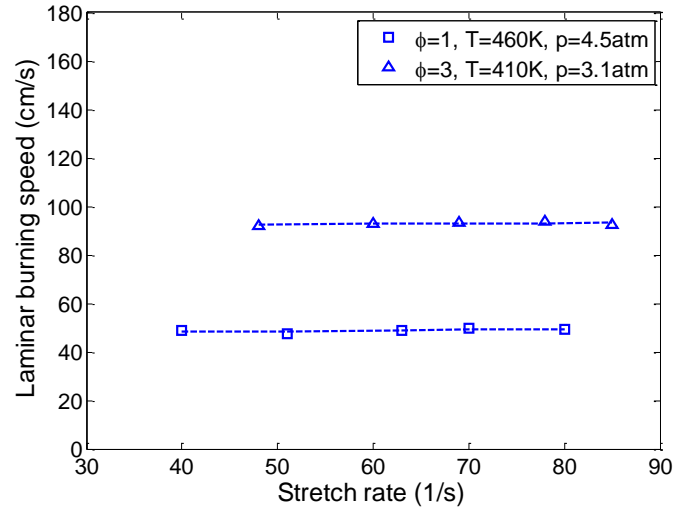


Figure 7 Laminar burning speed of $H_2/CO/O_2/N_2$ mixture versus stretch rates for two different equivalence ratios and unburned gas conditions at hydrogen concentration of 5%

BURNING SPEED THEORITICAL MODEL

The following model is for a spherical flame of one-dimensional geometry in the radial direction. The chamber is filled with a combustible mixture and will be ignited at time $t = 0$ at the center of the chamber by two extended electrodes. Upon ignition, an isotropic flame is created and begins to propagate outwardly in the radial direction. The following assumptions are made in the analysis of the combustion inside the vessel:

1. The unburned gas is initially at rest and has a uniform temperature, pressure and composition.
2. The pressure is presumed to be uniform within the chamber at each time step.
3. The burned gas in each shell will be at a different temperature and a different composition while it is in local thermodynamic equilibrium.
4. The gases are assumed to behave like an ideal gas.
5. The reaction zone has a negligible thickness.

The energy conservation equations for unburned, burned, and currently burning regions are respectively:

$$\dot{U}_u = -\dot{m}_b h_u + \dot{Q}_u - \dot{W}_u \quad (2)$$

$$\dot{U}_j = \dot{Q}_j - \dot{W}_j \quad , \quad j = b_1 - b_{n-1} \quad (3)$$

$$\dot{U}_{b_n} = \dot{m}_b h_u + \dot{Q}_{b_n} - \dot{W}_{b_n} \quad (4)$$

U is the internal energy, \dot{m}_b the mass burning rate, Q the energy transfer and W the work. In these equations the subscripts of j denotes the already burned shells, u and b denotes the unburned and burned gas conditions respectively, n and b_n denote the total number of shells in burned zone and currently burning shell, respectively. The dot sign on top of the parameters refers to the complete derivative with respect to time. Using thermodynamic relations [22] and concept of displacement thickness [23] Eqs. (2)-(4) expand into the following format:

$$\dot{T}_u^\infty = \frac{A_u^\infty \dot{p} + \frac{\rho_u^\infty h_u^\infty}{1-x_b} V_{dis_u} \dot{x}_b + \rho_u^\infty h_u^\infty \dot{V}_{dis_u} + \dot{Q}_u}{B_u^\infty} \quad (5)$$

$$\dot{T}_j^\infty = \frac{A_j^\infty \dot{p} + \rho_j^\infty h_j^\infty \dot{V}_{dis_j} + \dot{Q}_j}{B_j^\infty} \quad , \quad j = b_1 - b_{n-1} \quad (6)$$

$$\dot{T}_{b_n}^\infty = \frac{A_{b_n}^\infty \dot{p} + \left(m(h_u^\infty - h_{b_n}^\infty) - \frac{\rho_u^\infty h_u^\infty}{1-x_b} V_{dis_u} \right) \dot{x}_b}{B_{b_n}^\infty} + \frac{\rho_{b_n}^\infty h_{b_n}^\infty \dot{V}_{dis_{b_n}} + \dot{Q}_{b_n}}{B_{b_n}^\infty} \quad (7)$$

$$\dot{x}_b = - \frac{(C_u^\infty + \sum_k C_k^\infty) \dot{p} + (D_u^\infty + \sum_k D_k^\infty)}{\frac{F}{(E_u^\infty + \sum_k E_k^\infty)}} \quad (8)$$

$, k = u, b_1, \dots, b_{n-1}, b_n$

Eqs. (5)-(8) form a set of nonlinear ordinary differential equations which contain $n + 3$ unknowns: $p(t)$, $x_b(t)$, T_u^∞ and $T_{b_i}^\infty$ ($i = 1$ to n). Given experimental pressure as a function of time, they can be solved numerically using CVODE method to find burned mass fraction and temperature distribution. Finally, the laminar burning speed is calculated as:

$$S_u = \frac{m \dot{x}_b}{\rho_u^\infty A_f} \quad (9)$$

More information about model can be found in our previous publication [8].

BURNING SPEED MEASUREMENT

All laminar burning speed data are reported in regions where $r > 4$ cm and stretch rate of less than 80 s^{-1} . In these regions the effects of stretch and spark energy discharge are negligible. More information about burning model can be found in our previous paper [8].

Figure 8 to Figure 11 show the dependence of the laminar burning speed on the equivalence ratio, pressure, diluent, and hydrogen content. Figure 8 suggests that the maximum flame speed occurs around an equivalence ratio of 2 for a diluted

syngas mixture that is mostly carbon monoxide (5% H_2 + 95% CO) at an initial pressure of 2 atm. The laminar burning speed of the lean syngas mixture is quite low because of the additional 5% EDG. Figure 9 shows the negative pressure dependence of the laminar burning speed at a given equivalence ratio of 3, while Figure 11 shows the positive effect of increasing hydrogen content on the laminar burning speed. Figure 10 contrasts the effects of diluents on the laminar burning speed, and also highlights the profound effect of replacing nitrogen with helium. Adding more initial diluent decreases the laminar burning speed, as expected.

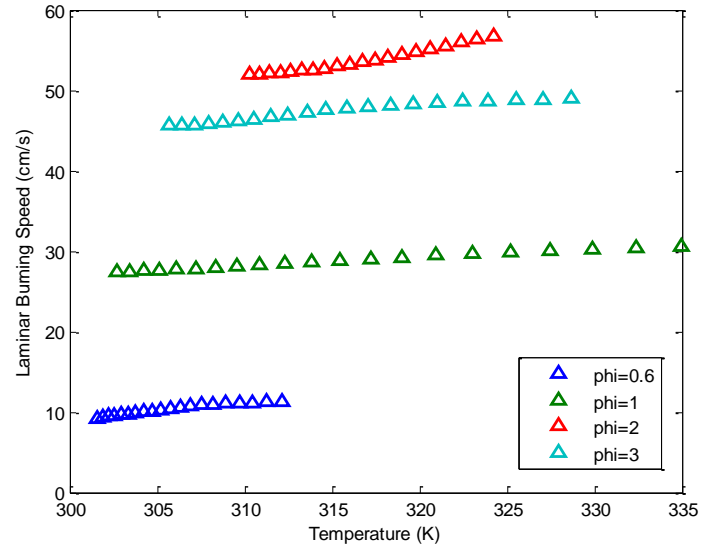


Figure 8 Laminar burning speed of $H_2/CO/oxidizer/diluent$ mixture along isentropes at different equivalence ratios

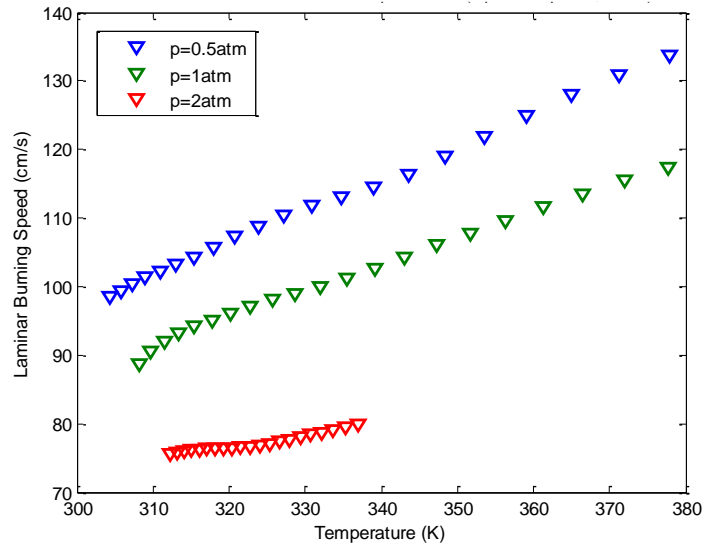


Figure 9 Laminar burning speed of $H_2/CO/oxidizer/diluent$ mixture along isentropes at different pressures

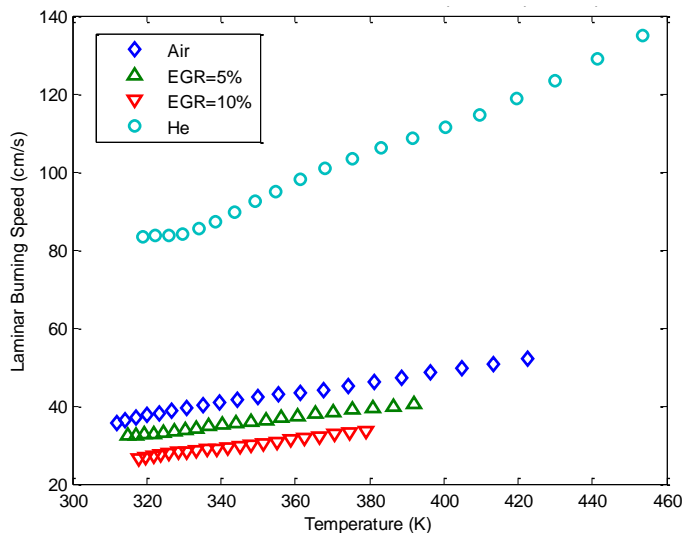


Figure 10 Laminar burning speed of H_2/CO /oxidizer/diluent mixture along isentropes at different diluents

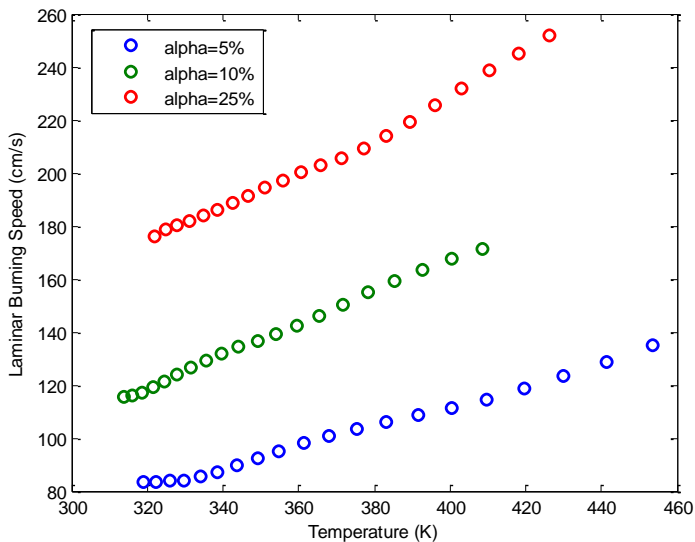


Figure 11 Laminar burning speed of H_2/CO /oxidizer/diluent mixture along isentropes at different hydrogen fractions

SUMMARY AND CONCLUSION

Experiments on syngas fuels were carried out in constant-volume chambers for hydrogen percentages of 5%, 10%, and 25%, initial pressures of 0.5 atm, 1 atm, and 2 atm, equivalence ratios of 0.6-3.0, and different diluents at ambient temperature. Measurements and snapshots were taken on syngas/air mixtures that were diluted with EDG percentages of 5% and 10% as well as syngas/oxidizer mixtures with helium (with the same volume percentages as oxygen and nitrogen in air). Extra diluent gases (EDG) decreases the laminar burning speed while increasing stability. Helium diluent increases the laminar burning speed given the same percentage by volume. Substitution of nitrogen in the air with helium, exactly with the same percentage,

increases the range of temperature and pressure of stable flame as well as the laminar burning speed. All data presented were in regions of negligible stretch.

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