Laminar burning velocity measurement of H_2/CO mixtures at elevated pressure using Heat Flux Method

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Abstract

The present work reports the first results of laminar burning velocity measurement of 50:50 and 85:15 % (by volume) H_2/CO mixtures with O_2:N_2 and O_2:He oxidizers using the heat flux method at pressure up to 9 atm for equivalence ratio from 0.5 to 1.0. The heat flux method creates a 1-dimensional adiabatic stretchless flame which is an important prerequisite for measurement of laminar burning velocity. This technique is based on balancing the heat loss from the flame to the burner with heat gain to the unburnt gas mixture, in a very simple way, such that no net heat loss to the burner is observed. Instabilities were observed in H_2 flames with N_2 as the bath gas at elevated pressure under lean combustion regime.

1 Introduction

Integrated gasification combined cycle (IGCC) is one of the most recent technologies that utilize syngas to reduce NO_x emissions without compromising efficiency in power production. IGCC enables the conversion of coal and biomass to a gaseous synthetic fuel gas (syngas) in the gasifier. Syngas is a mixture of hydrogen (H_2), carbon monoxide (CO), carbon dioxide (CO_2) and steam (H_2O). In combination with the gasification, the removal of CO_2 is made possible leaving increased levels of H_2. This technology of carbon capture and storage (CCS) has been very promising. Usage of high hydrogen content syngas not only reduces production of certain pollutants, it also increases the scope of utilizing its high energy content in the form of a fuel. High H_2 fractions in combination with other gases like CO is the present trend of syngas that requires a great deal of research. Gas turbines used in such technology operate at conditions of high pressure, temperature and lean fuel-air mixture. Hence, studying syngas combustion behavior at such conditions has become important.

The laminar burning velocity (S_L) defines the rate at which the unburnt mixture is consumed in the propagating laminar flame. This parameter is considered one of the most important entities in assessing many phenomena like flame quenching, flashback stabilization etc. in burners and combustors in systems like mentioned above. Along with its importance in designing combustors, this parameter also plays a key role in validating chemical reaction mechanisms. In theory, this property can be obtained when a flame is 1-dimensional, adiabatic and stretchless. There are a number of methods that can be employed in measuring this quantity. Some of them are the counterflow flame method, spherically propagating flame

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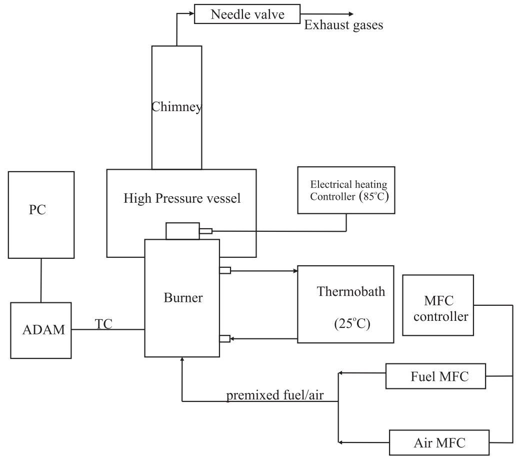


Figure 1: Schematic of the experimental setup

method and conical flame method. The heat flux method (HFM) [1] is a relatively new method and has proven to be accurate for measuring S_L since it creates a 1-dimensional, adiabatic and stretchless flame. The main principle of the heat flux method is to stabilize a flat flame with unburnt gas velocity such that the heat loss by the flame is compensated by heat gain by the unburnt gases. This relative heat flux is reflected in the burner plate temperature that decides the adiabatic state of the flame. The main focus of this work is to get insights into combustion of H_2/CO fuel mixtures at higher pressure and determine laminar burning velocity at lean conditions using the heat flux method.

In the recent work of Sun et al. [2] high pressure laminar flame speed measurements were reported to validate a chemical reaction mechanism for H_2/CO (from 1% H_2 to 50 % H_2 in the mixture) mixtures using a dual-chamber apparatus that generates an outwardly propagating premixed flame. A study of Natarajan et al. [3] demonstrates a conical flame in determining S_L at elevated pressure and temperature for similar fuel mixtures. A number of previous studies have also been highlighted. The present work focuses on applying an experimental method that produces stretchless flames that are sensitive to the adiabatic state. Flames of Syngas (50:50 %) and high-H_2 syngas (85:15 %) in the lean combustion regime at elevated pressure in combination with a variety of oxidizers will be studied. The laminar burning velocities of these mixtures have not been reported earlier.

2 High Pressure Experiments

The heat flux method makes use of the fact that the heat loss from the flame can be compensated by adding heat to the unburnt gas mixture. A schematic representation of the experimental setup is shown in Fig. 1. A perforated plate is fitted on a burner head (Fig. 2). The burner head is maintained at a temperature higher than the unburnt gas temperature. This gives a heat transport from the burner head to the burner plate and subsequently to the unburnt gas mixture. The balance in heat flux is reflected in the temperature profile of the burner plate.

The unburnt gas mixture flows through the plenum chamber of the burner which is maintained at 25 °C using a water thermal bath. The burner head is jacketed with a water thermal bath at 85 °C. Eight Copper-Constantan thermocouples are soldered at different radial positions on the perforated burner plate with holes of 0.3 mm diameter and 0.4 mm pitch (Fig. 2(right)). The plate has a thickness of 1 mm. The burner is placed in a high pressure cell (HPC) made of C45 steel designed for pressures

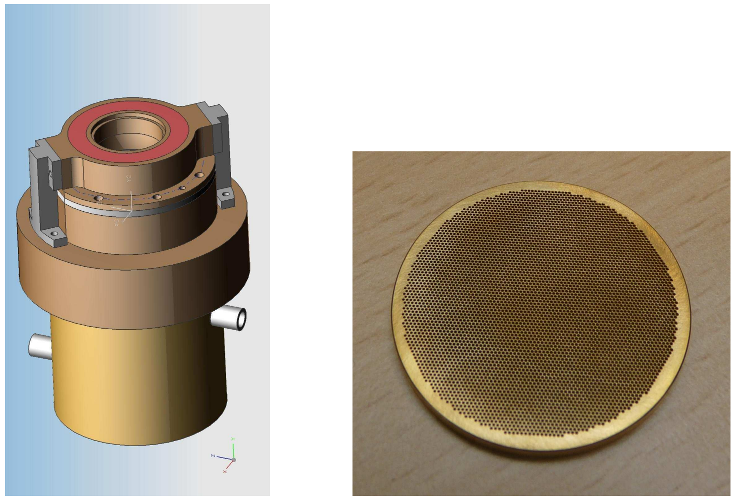


Figure 2: Burner and plate

up to 10 bars. A chimney is placed on top of the vessel through which burnt gases are guided out. A stainless steel connection pipe followed by a needle valve is connected to the exhaust of the chimney. The pressure in the vessel is controlled by this needle valve. When the unburnt gas velocity is higher than the adiabatic burning velocity (super-adiabatic) the heat gain by the gas is larger than the heat loss from the flame. The situation is opposite in case of a sub-adiabatic flame. The only measurement required in this technique is the temperature profile of the burner plate. The radial profile of temperature on the burner plate close to the adiabatic burning velocity is fitted by the method of least squares to a parabolic profile. The coefficients of such polynomials are plotted against sub and super adiabatic flow velocities. The adiabatic state is reached for zero value of the polynomial coefficient. The maximum error estimate for laminar burning velocity associated with this technique was 2 cm/s. Errors associated with equivalence ratio were less than 0.025 for all experiments. A detailed description of the concept, principle and procedure of a standard experiment using this method is discussed by Bosschaart and de Goey [1].

Table 1: Measurement conditions

Fuel	H_2/CO (50:50 %)	H_2/CO (85:15 %)
Oxidizer	O_2/N_2 (15:85 %)	O_2/N_2 (15:85 %)
	O_2/N_2 (10:90 %)	
	O_2/He (10:90 %)	
		O_2/He (11:89 %)
		O_2/He (12:88 %)
	O_2/He (12.5:87.5 %)	O_2/He (12.5:87.5 %)
ϕ	0.6 - 1.0	0.5-0.7
T (°C)	25	25
P (atm)	1 - 9	1 - 8

This method was initially applied to CH_4/air flames up to 5 atm [4]. The results agreed well with reported data available in the literature. Tests were then planned for H_2/CO mixtures. In doing so, a number of revisions had to be made in order to ensure an accurate and safe working environment. Since, these mixtures contained carbon monoxide, special attention was paid towards the safety aspects including a detection system and tested high pressure lines. Also, a special N_2-flushing line was introduced to keep the fuel line flushed when not in use. This ensured that no harmful gases could leak into the laboratory.

Figure 3: H_2/CO flames in high pressure environment.

In addition to this, it was also observed that carbon monoxide when stored in iron or Ni based cylinders or stainless steel flow lines, some of the gas converts into compounds like carbonyls [5]. When the mixture is lighted, the flame emits a pale, off-white color. This indicates that the mixture is not pure anymore. Hence, special care was taken to install brass and copper lines. It was also ensured that all the components could withstand pressures as high as atleast 10 bar. Mass flow controllers were calibrated specially for high pressure.

3 Results and Discussion

The tests were focused on 50:50 % and 85:15% H_2:CO mixture (by volume). In industrial conditions, these mixtures are burnt with air. This results in very high burning velocity. The heat flux method is best operational with a burning velocity not higher than 80 cm/s with the above mentioned plate dimensions. Hence, oxidizers were diluted with Nitrogen (N_2) and Helium (He). Table 1 summarizes all the experiments that were performed for the present work.

H_2/CO mixtures were introduced to the system first at atmospheric conditions. Figure 3 (left) shows a flame enclosed in the high pressure system. A flame obtained in heat flux method is 1-dimensional and does not exhibit any kind of instability/disturbance. Initially, the burner head was heated with a electrical heater [**?**]. With each experiment special attention was paid to ensure proper conduction of heat from the heater to the burner head by applying a highly conducting paste. To avoid acoustical instabilities that resulted from improper conduction, a new burner design was proposed and implemented in which water heating was incorporated. Due to better contact and conduction, this problem was solved. Atmospheric results of 50:50% H_2:CO are represented in Fig. 4. The results are well in agreement with Mclean et al. [6] and Sun et al. [2]. We also notice that the laminar burning velocity obtained with 10:90% O_2:He is comparable to that obtained by air. This observation has been well described by Galmiche et al. [7]. Figure 5(left) depicts the flames at pressures up to 5 atm with diluted oxidizers - 10:90% ; 15:85% O_2:N_2 ; 10:90% O_2:He (by volume) at equivalence ratio, ϕ = 0.7, 0.8, 1.0. With higher pressures the flame showed cellular/structural instabilities (Fig. 3(right)) with O_2:N_2 oxidizers due thermo-diffusive and hydrodynamic instabilities. Similar behaviour has been noticed by Sun et al. [2] and Natarajan et al. [3].

With time these experiments were repeated to gain experience and stability in operation. Since, it was observed that most stable and flat flames were obtained with O_2:He oxidizer, the rest of the measurements were carried out at different proportions of this oxidizer. Also, the objective was to reach lean conditions. During these measurements an equivalence ratio of 0.5 was achieved with the required fuel mixture. The picture in figure 3 (middle) shows a 50:50 % H_2:CO flame at 8.5 atm in a slightly super adiabatic state. The flames were highly sensitive as the pressure was increased. 1-1.5 cm/s change in

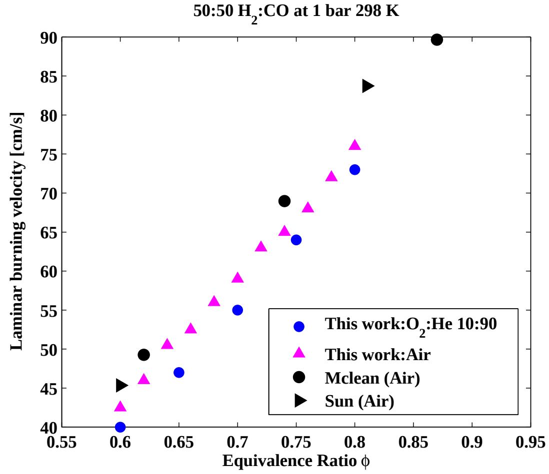
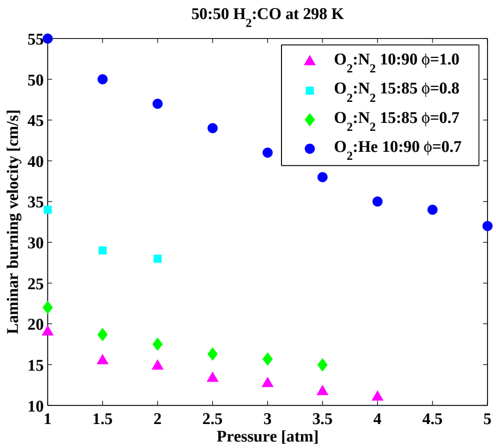


Figure 4: Measurements at atmospheric pressure



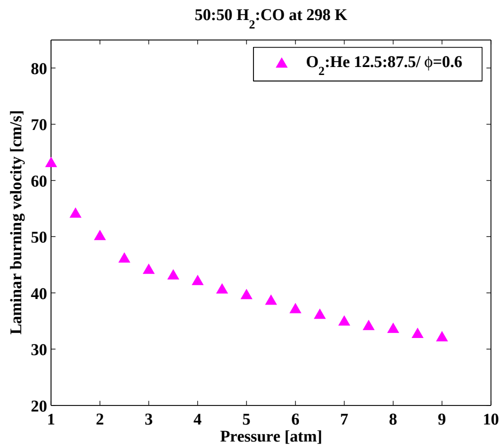
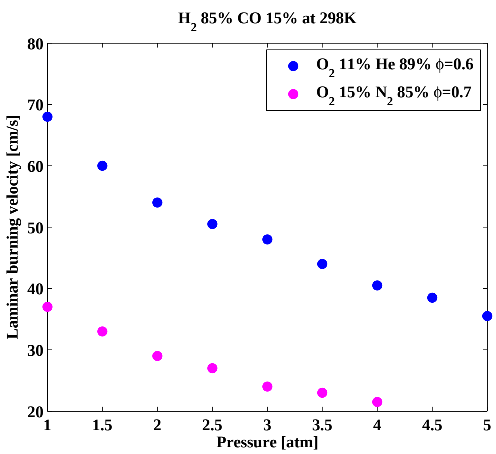


Figure 5: Laminar burning velocity of 50:50 % H_2:CO (by volume) mixture.



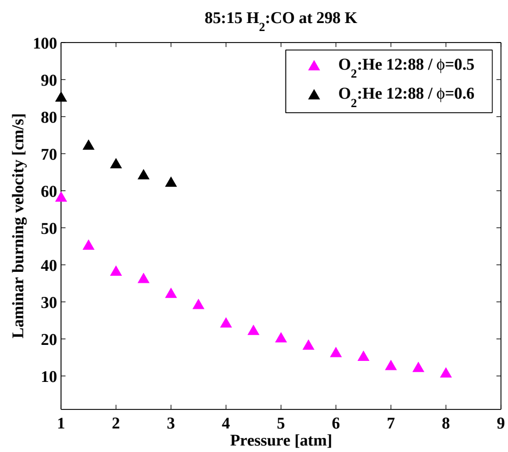


Figure 6: Laminar burning velocity of 85:15 % H_2:CO (by volume) mixture.

gas velocity from adiabatic state showed curved flames (super adiabatic) or nearly touched the burner plate and increased the plate temperature (sub adiabatic).

The tests were extended to 9 atm for 50:50 % (Fig. 5 (right) and 8 atm for 85:15 % H_2:CO flames(Fig. 6). Even at lean conditions there was high heat release that condensed water in the cell and a lot of heat from the hot burnt gases was absorbed by the walls of the high pressure cell (HPC). The oxidizers were consumed very fast in these experiments. Future experiments can be designed with the use of a burner plate with smaller hole diameter.

4 Conclusions

Stretchless adiabatic laminar burning velocities at elevated pressure were determined for 50:50 % and 85:15 % H_2:CO mixtures in combination with a variety of oxidizer compositions using the Heat Flux Method (HFM). Experiments at atmospheric pressure agreed well with recent results from literature. Further experiments were performed for elevated pressures up to 9 atm in steps of 0.5 atm, for a range of equivalence ratio from 0.5 to 1.0 at 25 °C. The 1-dimensional flames were stabilized in a high pressure environment with zero net heat loss which was reflected in the temperature profile of the burner plate.

5 Acknowledgement

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