An experimental and kinetic study of syngas/air combustion at elevated temperatures and the effect of water addition

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ABSTRACT

Laminar flame speeds of premixed syngas/air mixtures were measured at various fuel equivalence ratios (0.6–3.0), H_2 content of the fuel, and preheat temperatures (298–500 K) using a spherically expanding flame configuration. The measured laminar flame speeds were compared with simulations using with three existing chemical kinetic models – GRI Mech 3.0, H_2/CO Davis Mechanism and San Diego mechanism. Reasonable agreement between computations and measurements was achieved at room temperature that validated the new experimental configuration. However, at higher preheat temperatures discrepancies between computed and measured values were large, especially for fuel rich mixtures. Addition of H_2O to two fuels (H_2/CO = 5/95 and 50/50) up to 40% in the fuel–air mixture was studied to understand the effect of moisture in coal derived syngas. For the H_2/CO = 5/95 fuel, flame speed was observed to increase with up to 20% H_2O addition and then to decrease with any further water addition. However, the higher H_2 content fuel (H_2/CO = 50/50) only showed a decrease in flame speed with an increasing water concentration in the fuel–air mixture. The different trends have been explained as a result of the competing chemical and physical (dilution and thermal) effects of H_2O addition on the syngas flames using sensitivity analyses and by analyzing reaction rates and radical concentrations.

1. Introduction

Synthetic gas (syngas) is a mixture of carbon monoxide and hydrogen in various compositions and can be derived from a number of sources such as coal, biomass gasification or from natural gas during steam reforming [1]. Syngas is a potential clean fuel and is already being used in high efficiency gas turbines in the Integrated Gasification Combined Cycle (IGCC) for power generation. The composition of syngas is dependent mainly on the fuel source from which it is derived and it can contain minor constituents such as CO_2, H_2O, NH_3 and H_2S. Variation in its composition can affect the performance of the system. Therefore, it was considered essential to study the burning properties of syngas over a wide range of compositions and with the presence of minor constituents. The accuracy of kinetic models is critical for the design of clean and efficient gas turbine combustors. Thus, comprehensive assessment of the validity of existing chemical kinetic mechanisms under conditions representative of the operating conditions of advanced gas turbines is required.

Numerous experimental and modeling studies have been reported for syngas. Experimental studies include determination of species profiles [2,3], ignition delay times [4–7] and laminar flame

speed measurements [8–17]. Several mechanisms have been developed that can be used to represent the chemical kinetics of CO–H_2 and air mixtures, among which are the GRI Mech 3.0 by Smith et al. [18], the H_2/CO Mech by Davis et al. [19], the San Diego mechanism by Petrova and Williams [20], a reduced reaction mechanism for methane and syngas combustion in gas turbines by Slavinskaya et al. [21], and another H_2/CO kinetic mechanism by Frassoldati et al. [22]. Recently two other mechanisms have been developed by Li et al. [23] and Sun et al. [8] with the latest reaction rate constants.

Laminar burning speed is an important parameter that is commonly used for validation of chemical kinetics. Laminar burning speeds of syngas have already been investigated extensively at normal temperature and pressure, using spherical bombs [8,10,11,13,17], counter-flow twin flame configurations [12,24] and Bunsen burners [15,25]. Considerable accuracy in the prediction of the laminar burning speeds has been achieved for normal temperature and pressure conditions. However, anomalies have been reported at higher temperatures and pressures that are closer to the conditions that exist in gas turbines or burners. Natarajan et al.'s [15] studies of CO–H_2 combustion for fuel lean conditions showed that at high temperatures the measured laminar flame speeds have large discrepancies with numerical simulations using GRI Mech 3.0 and H_2/CO Davis Mechanism. Furthermore, few studies (except [26–28]) have considered the effects of minor species

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such as H_2O, which could alter the ignition and combustion behavior of syngas.

The motivation of the present paper is to extend flame speed measurements of syngas for various H_2–CO compositions (5% H_2–95% H_2), equivalence ratios (0.6–3.0), and high temperatures (298–500 K) using a spherical flame configuration. In addition, the effect of addition of H_2O to the fuel mixture at high temperature was investigated. Difference in trends of laminar flame speeds with increasing water percentage, for fuels containing H_2/CO in different ratios has been discussed.

2. Experimental method

The major components of the facility used for the experiments include a spherical combustion chamber placed inside a high temperature oven, a high voltage spark generator, and a Schlieren optical system for recording the flame propagation inside the chamber. A schematic of the experimental facility is shown in Fig. 1. The spherical combustion chamber is made of stainless steel with an inside diameter of 36 cm. The chamber is fitted with two diametrically opposed electrodes made of tungsten wire to provide a spark at the center of the chamber. The gap between these two electrodes is adjustable and the voltage between the two electrodes is controlled by the high voltage spark generator to provide the minimum ignition energy. Two quartz windows, with high durability against pressure and temperature shocks, are mounted on diametrically opposite ends to provide access for viewing the flame. The combustion chamber is placed in a customized oven that can be preheated to a maximum temperature of 650 K using a precise temperature controller. A Z-type Schlieren imaging system has been set up to visualize flame propagation using a high-speed digital camera with a maximum capture rate up to 6600 frames per second.

At the start of an experiment, the chamber was first vacuumed and then the reactant mixture was prepared by filling the chamber with gases at the appropriate partial pressures depending on the desired mixture composition and test pressure. The prepared mixture was allowed to lay undisturbed for 30 min to allow for any disturbances to settle and to ensure complete mixing of the fuel and air by diffusion. A triggering circuit was used to simultaneously trigger the recording on the camera and the spark igniter. For the experiments with water vapor addition, water was

preheated in a chamber to vapor phase before being added to the fuel mixture in the combustion chamber. Several thermocouples were installed inside the heated water chamber and on the pipelines between the water chamber and the combustion chamber to ensure the temperature were above the boiling point of water.

Commercial grade compressed air was used for the experiment with 99.5% purity. H_2 and carbon monoxide were graded at 99.8% and 99.5% purity respectively. A recent study by Chaos and Dryer [31] has attributed the deviations between experimental results from different groups to the presence of iron pentacarbonyl $(Fe(CO)_5)$ impurities in carbon monoxide. Therefore, carbon monoxide stored in aluminum containers was used in the experiments to ensure the absence of iron pentacarbonyl, a flame inhibitor, as shown by Rumminger and Linteris [30], that develops over time in carbon monoxide stored in steel containers.

3. Data processing

Similar to previous measurements of flame speeds [32,33] for gaseous fuels using a spherical combustion chamber of the same size, flame radius measurements have been limited to a range of 8 mm < r < 30 mm. The lower limit was chosen to avoid disturbances caused by the transient ignition process and the upper limit was to ensure that the pressure increase inside the chamber was negligible. The lower limit was determined experimentally in a manner similar to that employed in the studies of liquid fuels, which was described in a previous paper [34]. The upper limit of 30 mm was imposed on the radius of flame measurements in accordance with past flame speed measurement in a similar configuration [32,35–38]. This value corresponds to 16.67% of the maximum radius and just 0.46% of the total volume of the chamber, which ensures that the pressure rise over this radius is less than 0.7%. Additionally, Burke et al. [39] has shown that for flame radius less than 40% of the maximum, the flame speed is affected by less than 1% due to pressure increase in the chamber. Chen et al.'s [40] theoretical study of a spherically propagating methane/air flame also showed that for a ratio of flame radius to maximum radius lesser than 0.2, the effect of compression induced flow is almost negligible.

Under these assumptions, the local stretched flame speed and flame stretch is given by the following quasi-steady expressions proposed by Strehlow and Savage [41],
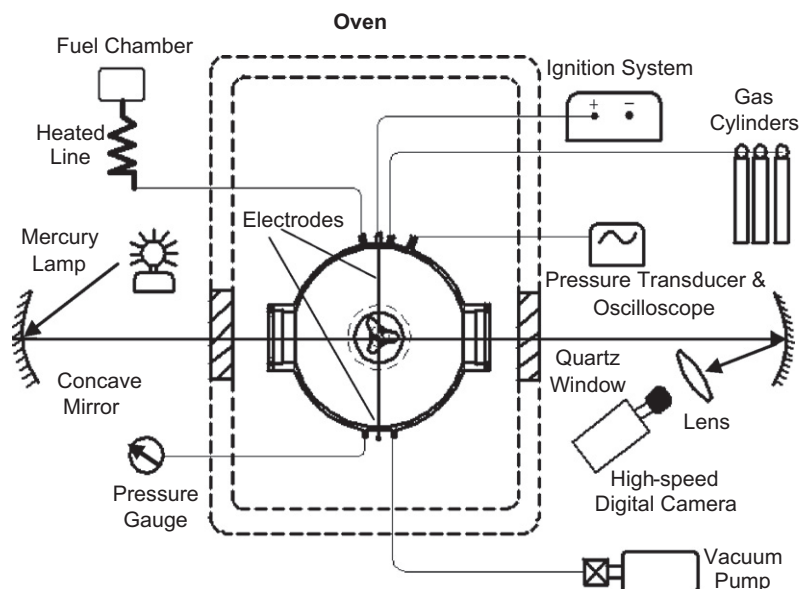

Fig. 1. A schematic of the experimental facility.