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Proceedings of the Combustion Institute 34 (2013) 719–726

Proceedings of the Combustion Institute

www.elsevier.com/locate/proci

The effects of water dilution on hydrogen, syngas, and ethylene flames at elevated pressure

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Abstract

This work investigates experimentally and numerically the kinetic effects of water vapor addition on the burning rates of H_2, H_2/CO mixtures, and C_2H_4 from 1 atm to 10 atm at flame temperatures between 1600 K and 1800 K. Burning rates were measured using outwardly propagating spherical flames in a nearly constant pressure chamber. Results show good agreement with newly updated kinetic models for H_2 flames. However, there is considerable disagreement between simulations and measurements for H_2/CO and C_2H_4 flames at high pressure and high water vapor dilution. Both experiments and simulations show that water vapor addition causes a monotonic decrease in mass burning rate and the inhibitory effect increases with pressure. For hydrogen flames, water vapor addition reduces the critical pressure above which a negative pressure dependence of the burning rate is observed. However, for C_2H_4 flames, the burning rate always increases with pressure. The results also show that water vapor addition has the same effect as a pressure increase for H_2 and H_2/CO flames, shifting the reaction zone into a narrower window at higher temperatures. For all fuels, water vapor addition increases OH formation via $H_2O + O$ while reducing the overall active radical pool for hydrogen flames. For C_2H_4, the additional HO_2 production pathway through HCO results in a dramatic difference in pressure dependence of the burning rate from that observed for hydrogen. The present work provides important additions to the experimental database for syngas and C_0–C_2 high pressure kinetic model validations.

Keywords: Water dilution; Laminar burning rate; Elevated pressure; Syngas; Ethylene

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1. Introduction

In order to reduce NO_x emissions and achieve CO_2 capture from combustion in advanced fuel-flexible gas turbines, near limit combustion of high hydrogen content (HHC) syngas, natural gas, and shale gas at lower flame temperatures is desirable. Steam addition and exhaust gas recirculation (EGR) are frequently utilized to reduce the flame temperature. Additionally, advanced gas turbines are typically operated at high pressure. Typical oxygen blown coal derived syngas may contain up to 15% water vapor as well as H_2, CO, N_2, and CO_2 [1] while natural gas and shale gas contain CH_4, and varying amounts of C_2H_6, C_3H_8, and C_4H_{10}. The high hydrogen content in HHC fuels, high pressure, low temperature, and water vapor addition create challenges such as flashback, autoignition, and dynamic instabilities [2]. Understanding the kinetics of C_0–C_2

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http://dx.doi.org/10.1016/j.proci.2012.06.065

combustion with water vapor dilution at high pressure is central to developing robust computational tools for optimizing the design of advanced turbines. The fuels examined in this study are chosen to aid in the development of a hierarchical kinetic model that can be validated with practical fuels at applied conditions. Hydrogen and carbon monoxide, the major components of syngas, also form the base of hierarchical combustion chemistry, while ethylene is examined as it is an important intermediate species formed during higher carbon number alkane oxidation for those species found in natural gas and shale gas.

Previous studies of HHC fuel combustion have focused on hydrogen at low [3,4] and high pressure, [5] and hydrogen/carbon monoxide mixtures at high pressure [6,7]. In contrast to lower pressure observations, high pressure studies have shown the existence of a negative pressure dependence for the burning rate and significant deficiencies in model predictions [5,7]. Analyses of the high pressure results show that HO_2 + radical pathways play an important role in controlling radical branching/termination. An updated high pressure hydrogen oxidation model [8] was developed with particular attention paid to improving understanding of the HO_2 + radical reaction pathways.

While some studies [9–12] have evaluated the effects of water vapor dilution on burning rate, few [13,14] have considered elevated pressure conditions. For example, Das et al. [11] investigated water vapor dilution of hydrogen/carbon monoxide flames at atmospheric pressure. The results showed a large thermal (heat capacity) effect, as well as a chemical kinetic increase in OH production by $H_2O + O = 2OH$. Mazas et al. [12] investigated the effects of water on oxygen-enriched methane flames at atmospheric pressure, identifying a thermal effect, as well as a chemical kinetic effect principally due to the high collisional efficiency of water for $H + O_2(+M) = HO_2(+M)$. Mazas et al. found the chemical effect was more pronounced at lower flame temperatures and with lower oxygen contents. High pressure H_2 flame speeds with water vapor dilution [13,14] show a thermal and chemical inhibition due to termolecular reactions, but the experimental results have large uncertainties.

The goal of this paper is to examine the chemical kinetic effects of water vapor dilution on the mass burning rates of H_2, H_2/CO, and C_2H_4 flames from 1 to 10 atm at flame temperatures from 1600 K to 1800 K, and to provide validation targets for C_0–C_2 high pressure kinetic mechanism development. Mass burning rates are measured using outwardly propagating spherical flames in a high pressure heated combustion chamber. Then, chemical kinetic predictions are evaluated and the effects of water dilution on high pressure reaction pathways are analyzed.

2. Experimental methods

2.1. Experimental apparatus

Experiments were conducted in a high pressure constant volume heated spherical bomb (Fig. 1). The spherical combustion chamber has an inner diameter of 20 cm, allowing for a relatively large range of data that is minimally affected by unsymmetrical confinement induced fluid motion [15]. The combustion chamber is placed inside an oven under PID control based on the temperature in the combustion chamber. An electric fan inside the oven minimizes the temperature nonuniformity there to within 5 K.

A magnetically assisted passive pressure release system is used to release the high pressure generated after ignition and quench the flame, similar to [16]. When the pressure in the chamber rises above a specified pressure, the chamber is connected to a nitrogen tank, increasing the volume by a factor of 13 so that the pressure in the system remains within safe limits. This release occurs after the flame passes outside the viewing window. High speed Schlieren photography is utilized to image the flame propagation as in [16]. From the images, the flame front is located using an edge detection program and the flame radius, R_f, is determined. The flame radius data are smoothed using a moving average and the flame front velocity, dR_f/dt, is calculated by taking the time derivative.

Mixtures of hydrogen (99.9%), carbon monoxide (99.99%), oxygen (99.5%), helium (99.995%), argon (99.999%), and water vapor were prepared using the partial pressure method. Compressed carbon monoxide was stored in an aluminum cylinder with brass fittings to eliminate production of iron-carbonyl [2]. Before filling the combustion chamber, deionized liquid water was vaporized in a separate vaporization chamber. All gases including water vapor pass through electrically heated tubing before entering the combustion chamber. The tubing and vaporization chamber are heated to the same temperature as the oven using separate K-type thermocouples and resistance heaters. To prevent condensation at the pressure gauge outside the oven, the chamber is initially filled with helium. To reduce uncertainty, a portion of the major diluent flows through the pressure gauge tubing into the combustion chamber, pushing any fuel or oxidizer that may have been pushed into this tubing back into the chamber. The diluents and data range for analysis were adjusted for each set of conditions to minimize the effects of ignition transient behavior, cellular instabilities and wrinkling, and buoyancy [4].

Experiments were conducted at an initial pressure from 1 to 10 atm and initial temperature of 393 ± 2 K with an equivalence ratio of 0.85. This equivalence ratio was chosen because lean premixed combustion is typical of future syngas gas