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Effect of preheating on rich combustion for syngas production: experiments and modeling of burner-stabilized flames

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Abstract

The production of syngas from hydrocarbons is an important part of the hydrogen infrastructure. Previous experimental studies have shown that rich mixtures of methane, heptane, and ethanol are successfully converted to syngas in non-catalytic heat recirculating reactors but existing models fail to predict the efficiency of the conversion. Although the reactor designs vary, the primary feature is the recirculation of exhaust heat to preheat the reactants such that the flammability limits are extended and the burning rate is increased. Detailed modeling of these reactors, however, is complicated by the complex heat transfer processes. To understand modeling deficiencies, we are beginning an investigation of the conversion of rich mixtures of methane in a relatively simple flat flame burner with externally preheated reactants. The goal of this initial investigation with methane is to determine the similarity between the two burners that we use (an actively cooled McKenna burner and a ceramic matrix burner) and the computational model and to determine the effect of preheating on the expansion of the operating conditions of the ceramic burner. Experimental data include species measurements and operating range determinations. Additionally, we present results from computational modeling of the process for comparison with experimental data.

1 Introduction

Significant research on porous, or filtration, reactors has shown that these devices have large operating ranges and provide an effective means of converting rich hydrocarbon mixtures to syngas [1-5]. Experimental demonstrations of a wide variety of fuels including methane [1, 5],

heptane [2], and ethanol [3] suggest that these reactors are very robust and have many potential applications. Optimization of their design, though, is hindered by the complexity of the physical and chemical processes that take place in the reactor. Heat is recirculated from the high temperature zone through the porous matrix to the unburned reactants, and the amount of preheating is a function of the porous solid characteristics and the local fluid dynamics [6]. Direct observation of the reaction front is virtually impossible because it is embedded within the pores of the matrix. In addition, operation of these reactors is often at equivalence ratios well beyond those normally used for validation of chemical kinetics mechanisms [2, 3].

To further our understanding of rich filtration combustion, we are investigating the importance of various processes in these reactors. In the present study, we focus on the effect of preheating, without the complexity of the porous solid, on the combustion of rich methane/air mixtures. A flat flame is particularly attractive for this study because both experimental and computational methods are well-established. The McKenna burner is the best known of the standard flat flame rigs and we use this for our tests with reactants at ambient conditions. This burner, though, cannot withstand reactants at the high levels of preheat that are necessary for the current study, so we constructed a ceramic burner for experiments with high preheating. The results show the importance of significant levels of preheating on the extension of the operating range of these burners.

2 Method

Experiments and computations were conducted on burner-stabilized rich methane flames. A McKenna burner was used for experiments with no reactant preheating, and a ceramic matrix (Mullite) burner was used for experiments with preheating, both of which can be seen in Figure 1. The preheating in the ceramic burner is accomplished by heating the air with an inline heater and mixing with methane downstream of the heater. The temperature of the mixture was monitored with a thermocouple placed near the entry to the ceramic matrix. For the experiments, a flame was deemed flat by qualitative observation, and all the data presented here are measurements from flat flames. Experimental measurements included exhaust species, which

were obtained by using a Gas Chromatograph with small-diameter quartz probe placed ~1 cm above burner surface.

Computations were performed using Cantera's burner stabilized flame code [7]. The energy equation was enabled and the kinetics mechanism GRI3.0 [8] was used. It is important to note the differences between the two burners and the model when comparing results. In the model, a temperature boundary condition is set to the desired reactor inlet temperature, and the energy equation is solved in the gas phase. The temperature gradient at the surface represents an energy loss from the gas phase. A McKenna burner has an efficiently cooled surface, so the constant temperature boundary condition in the model is reasonable. However, the ceramic burner that we use is not actively cooled. Heat from the reaction is removed by radiation from the burner surface and by conduction. Some of the heat from the reaction zone is conducted upstream through the ceramic, thus preheating the incoming reactants. The model that we have used does not include these heat transfer mechanisms, so it is not expected that the modeling results will necessarily agree with the experimental results from the ceramic burner. Equilibrium calculations were also performed for constant pressure, adiabatic conditions using Cantera.

3 Results

Experimental data include measurement of species in the exhaust and preheat temperature. The species data are presented as yields defined as:

$$H_2\ Yield = \ 100 \times \frac{2 \times \dot{N}_{H_2}}{4 \times \dot{N}_{CH_4}}$$

$$CO\ Yield = \ 100 \times \frac{\dot{N}_{CO}}{\dot{N}_{CH_4}}$$

Where $\dot{N}$ indicates the molar flow rate of a particular species

Data from the computational model are included for comparison. Equilibrium data are also presented as they provide insight into the thermodynamic characteristics of the methane/air system.

3.1 McKenna Burner Results

In Figure 2 the line and the shaded region indicate the conditions under which a flat flame was obtained on the McKenna burner. As the inlet velocity is increased, the range of φ that yields a flat flame decreased.

Figure 3 shows the H_2 and CO yields as a function of φ for experiments, computations and equilibrium calculations. The inlet velocity was held constant at 10 cm/s. The experimental results, computational results, and the equilibrium data all show excellent agreement in trend and very good agreement in magnitude. The yields are small near stoichiometric, and then rise quickly with increasing φ. The maximum value of H2 yield is ~25% and the maximum CO yield is ~65%, both at φ = 1.35.

In order to understand the effect on inlet velocity, we performed experiments with the Mckenna burner holding φ constant at 1.2 and varied the inlet velocity (Figure 4). Since equilibrium is based only on thermodynamic properties, the equilibrium values do not change with inlet velocity. Flame computations predict constant or slightly increasing yield with inlet velocity and show good agreement with equilibrium. The experimental results show no discernable trend except for an increase at the lowest value of inlet velocity. At higher inlet velocities, the yields of hydrogen and carbon monoxide

3.2 Ceramic Burner Results

To isolate the effect of the preheating on the reaction process, we studied a rich flat flame with preheated reactants. Computations using Cantera were performed to identify the level of preheating required to produce a stable solution for burner-stabilized methane/air flames with an inlet velocity of 10 cm/s. The results of these computations show that preheating is required for a stable flame with mixtures having φ near 1.5 and higher. As the φ increases beyond this value, higher levels of preheating are necessary; reactants having an φ of 2.5 must be preheated to more than 800K for a stable solution to be obtained.

Experiments were conducted on the ceramic burner with the inlet velocity held constant at 20 cm/s, and observations of flatness as a function of φ and inlet temperature are presented in Figure 5. As φ is increased, the inlet temperature required to produce a flat flame also increases. This trend is not surprising since the conventional flammability limits also with increase with initial temperature [9]. With unheated reactants, the maximum equivalence ratio that produced a flat flame was ~1.2. With reactants preheated to 700K, a flat flame was obtained at an equivalence ratio ~1.5.

Figure 6 shows the H_2 and CO yield as a function of φ for experiments conducted with the ceramic burner with the inlet temperature and velocity held at 300K and 20 cm/s, respectively. The experimental and computational results follow the trends of equilibrium showing a sharp increase in yield as the equivalence ratio is increased from stoichiometric. These results are very similar to those presented for the McKenna burner (Fig. 3) although it should be noted that the inlet velocities were different. These data suggest that, at least in this range of tested φ, the ceramic burner may be used as a surrogate for the McKenna burner in experimentation.

Conclusions/Future Work

An initial investigation of rich, preheated flat flames was undertaken in order to understand the effect of preheating on conversion of methane to syngas. It was found that under the relatively limited range of φ tested that the model agreed well with the experiments in yield trend for both the McKenna burner and the ceramic burner. It was also found that the two burners showed similar yields under a range of φ. The results provide some evidence that the ceramic burner may be used as a surrogate for a Mckenna burner, but more testing is required. Lastly, we found that the operating range of the ceramic burner was increased significantly by preheating the reactants.

Besides performing experiments with methane under an expanded the range of conditions, we also plan perform a similar set of experiments with larger fuels such as butanol and heptane.

These data, along with data obtained from a filtration reactor, will be used to help understand some of the more fundamental characteristics of heat recirculating reactors.

Acknowledgements

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List of Figures

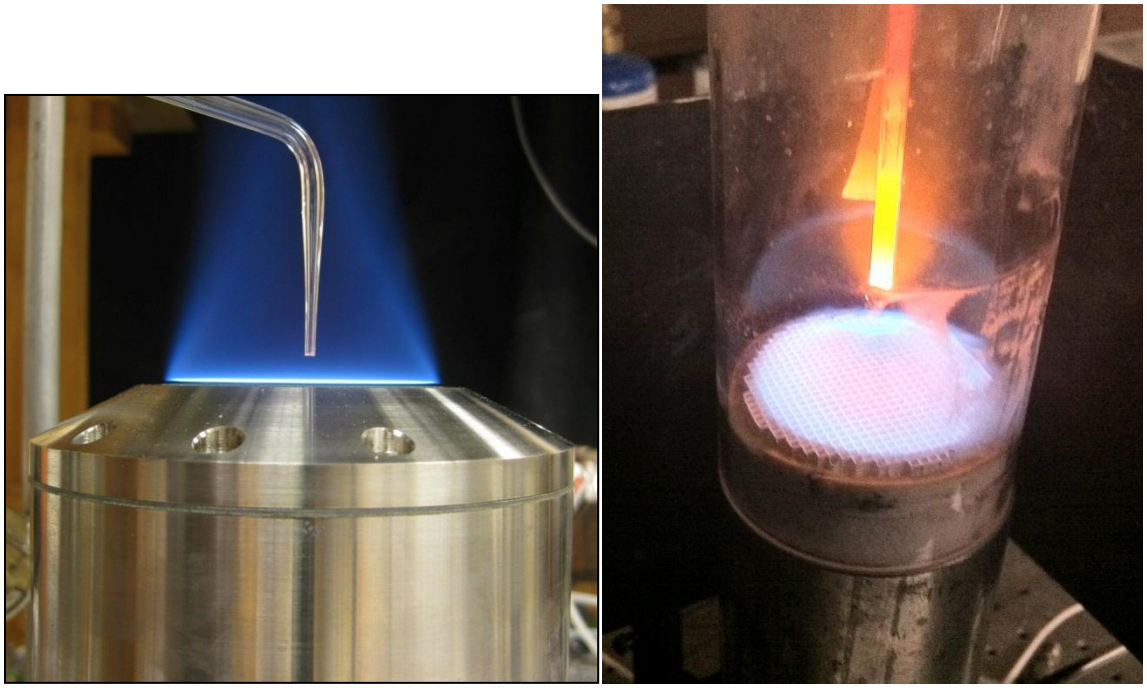

Figure 1: (a) McKenna burner, (b) Ceramic burner

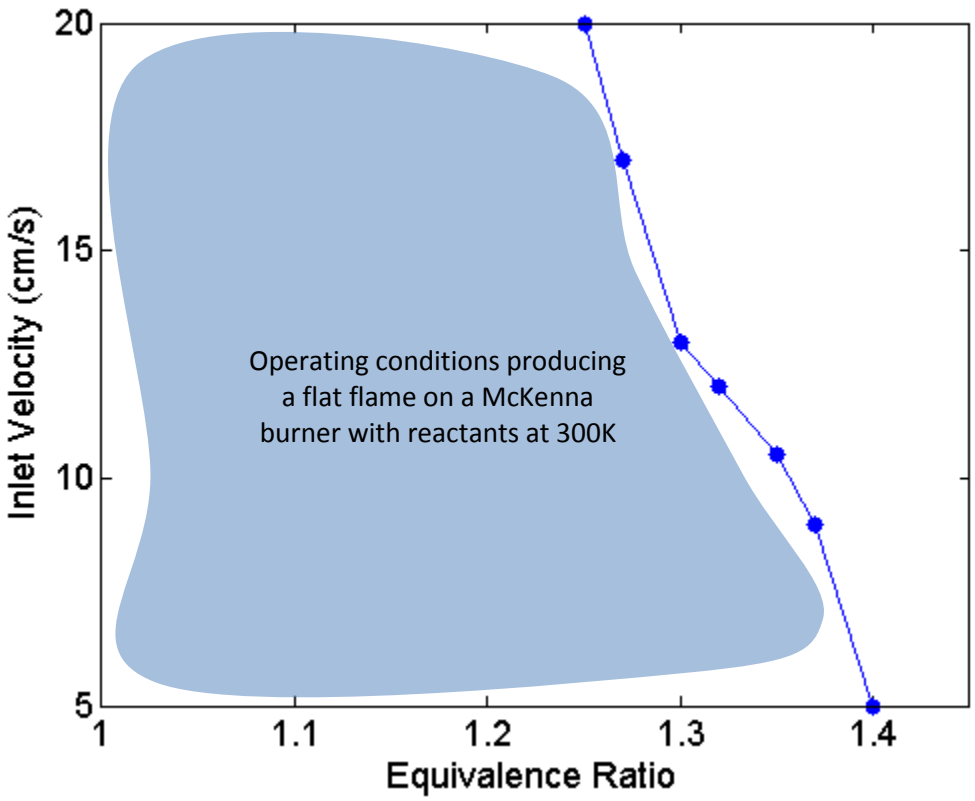

Figure 2: Operating conditions producing flat flames on McKenna burner

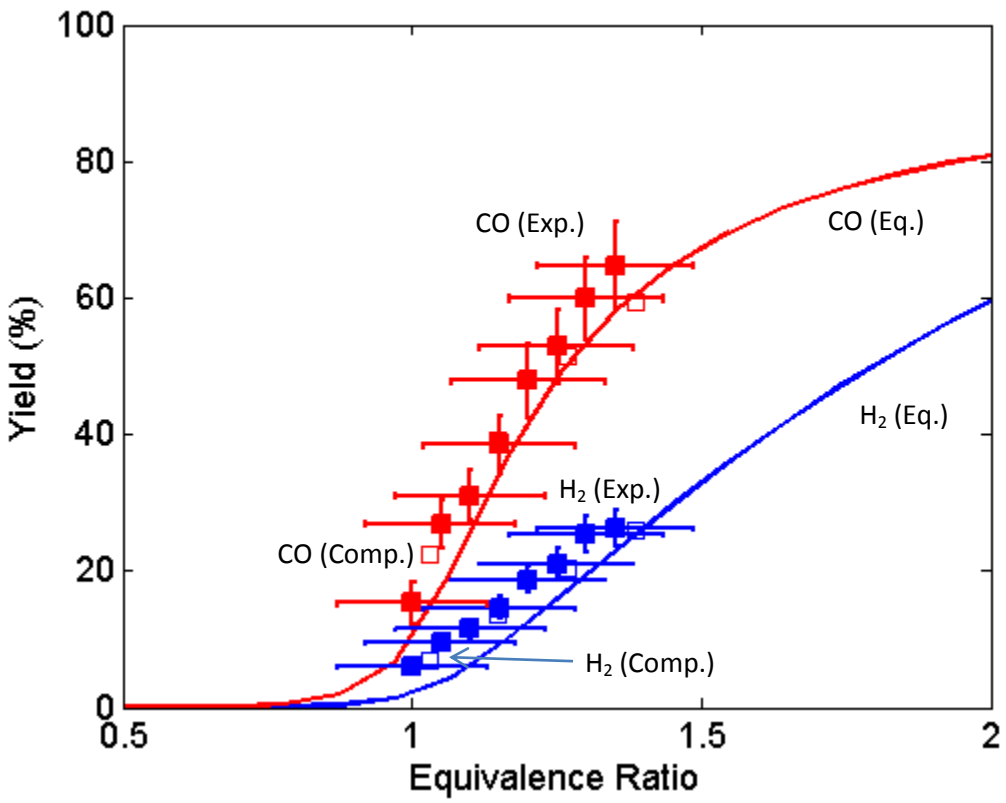

Figure 3: H_2 and CO Yield vs. Φ (inlet temperature = 300K, inlet velocity = 10 cm/s) – McKenna burner-stabilized flame experiments (Exp.), computations (Comp.), and equilibrium calculations (Eq.)

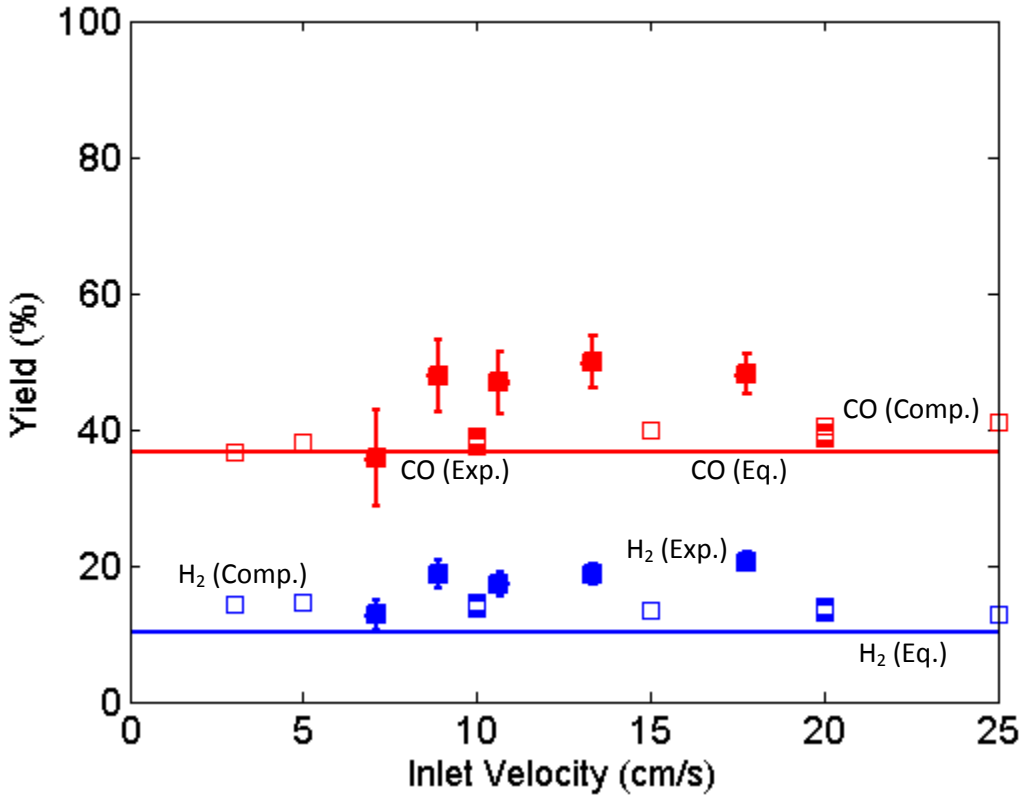

Figure 4: H_2 and CO Yield vs. inlet velocity (inlet temperature = 300K, Φ = 1.2) – McKenna burner-stabilized flame experiments (Exp.), computations (Comp.), and equilibrium calculations (Eq.)

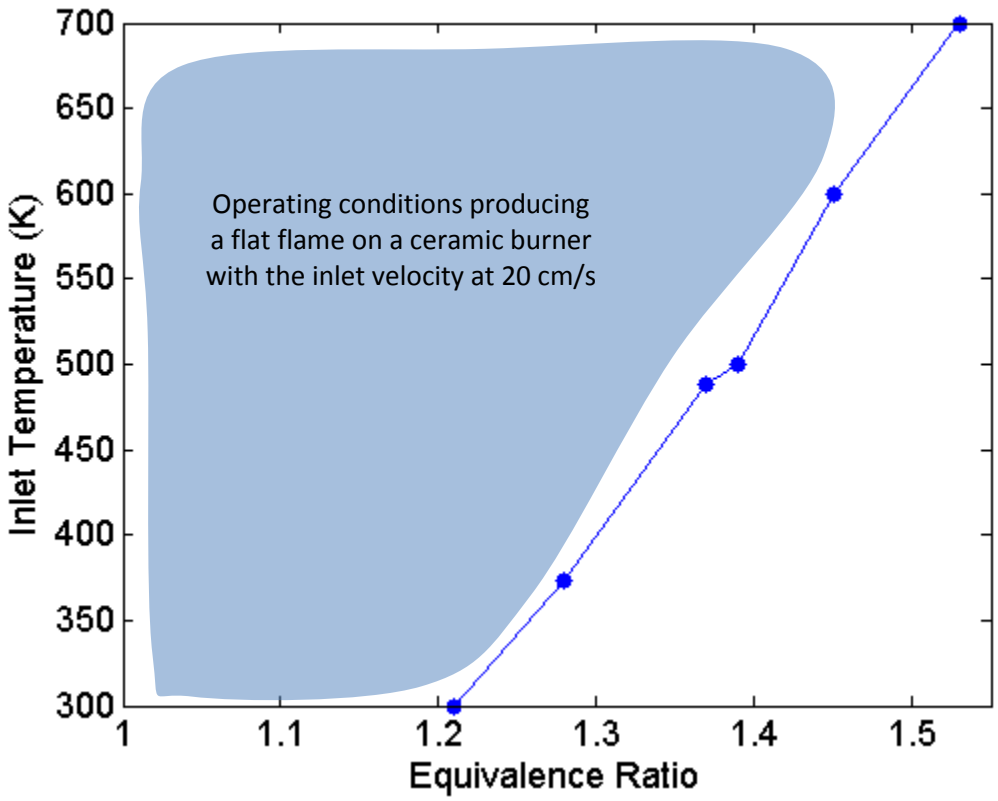

Figure 5: Operating conditions producing flat flames on ceramic burner

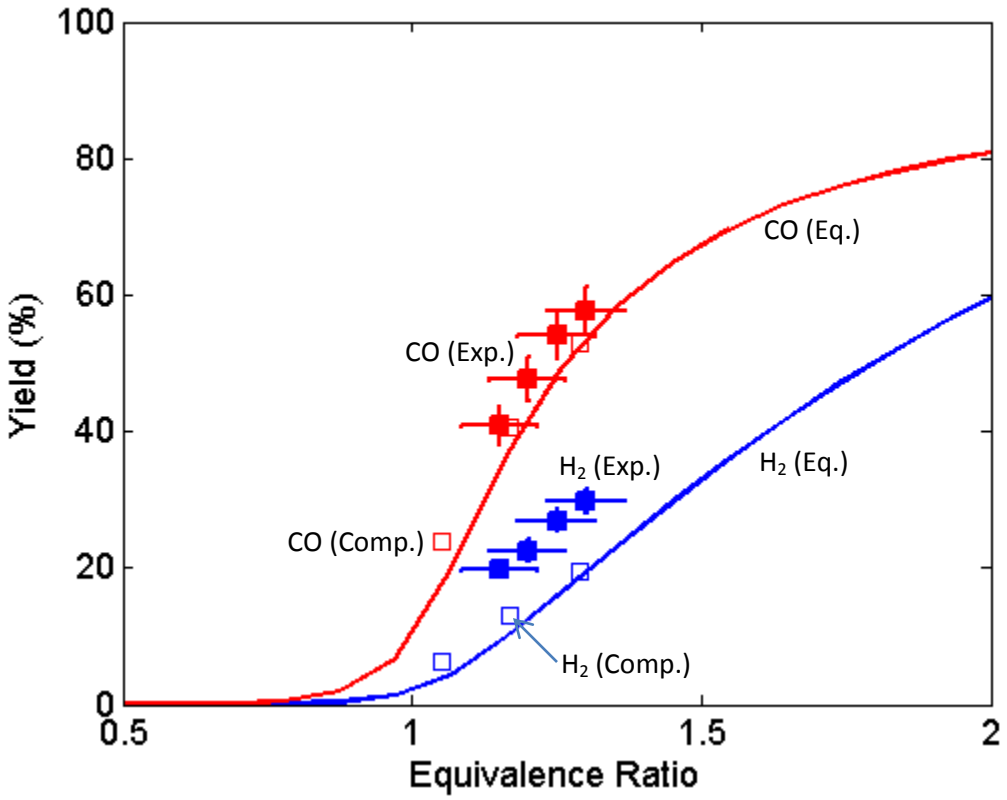

Figure 6: H_2 and CO yield vs. Φ (inlet temperature = 300K, inlet velocity = 20 cm/s) – Ceramic burner-stabilized flame experiments (Exp.), computations (Comp.), and equilibrium calculations (Eq.)