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Lean heptane and propane combustion in a non-catalytic parallel-plate counter-flow reactor

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ABSTRACT

Small scale lean combustors, in conjunction with mechanical and electrical conversion devices, have the potential to meet increasing portable power needs. This study examines the combustion of lean premixed fuels in a mesoscale, heat-recirculating parallel-plate counter-flow reactor. The reactor utilizes internal heat recirculation to produce temperatures in excess of the adiabatic flame temperature, known as superadiabatic combustion. As a result of these elevated temperatures, burning velocities above adiabatic values and extension of flammability limits, including stable operation at very lean equivalence ratios, may be attained. Combustor stability and emissions of carbon monoxide, nitrogen oxides, and unburned hydrocarbons have been previously studied for lean combustion of methane in the counter-flow reactor. This study examines lean combustion of two increasingly complex fuels, propane and heptane, due to the logistical importance of both gaseous and liquid fuels. Stability ranges and emissions measurements are presented for the counter-flow reactor operating on these fuels, and the results are compared to previously measured emissions for lean methane. Stability ranges of the counter-flow reactor operating on lean propane and heptane are also compared to those in the rich regime. Further analysis of the results for lean methane, propane and heptane combustion utilizes peak reactor wall temperatures as an indicator of favorable operating conditions, and highlights the importance of superadiabatic operation for achieving large turn-down ratios and low levels of emissions.

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1. Introduction

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Demands for small scale and portable power sources are increasing for a wide variety of applications, including propulsion and electricity generation. Hydrocarbon fuels currently have higher energy density than even the most advanced batteries, therefore combustion-derived power continues to be preferable for many applications. The use of small scale combustors in conjunction with mechanical or electrical conversion devices, such as microturbines or thermoelectrics, has been proposed to address the increasing need for portable power [1–3]. Lean reactant mixtures are favorable because they can permit increased levels of fuel conversion and thermal efficiencies as compared to reactant mixtures nearer to stoichiometric. In order to achieve a given firing rate, however, increased volumetric throughput is required as reactants become progressively leaner. Therefore, the optimal equivalence ratio (ϕ) for combustor operation is dependent upon the application. In order for a combustor to be widely applicable, a broad operating range is desirable. As shown in a previous study, stable combustor operation over a broad range of equivalence ratios and inlet

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velocities (*u*) is necessary to achieve a wide range of firing rates while minimizing pollutant emissions [4].

One technique for extending conventional flammability limits and increasing burning speed is the use of heat recirculation to increase the enthalpy in the reaction zone above that of adiabatic conditions. This is accomplished by using hot combustion products to preheat reactants. As a result, peak temperatures in the combustion zone can be significantly superadiabatic, or in excess of the adiabatic temperature predicted by chemical equilibrium.

Various heat-recirculating combustor designs have been proposed and studied, and all operate on the principal of transferring heat through a solid. In some cases, the combustor consists of a solid matrix, such as a reticulated foam or packed bed of spheres [5–9]. In others it consists of parallel channels [10], such as the reactor used in this study. Theoretical analyses and experimental studies of channel reactors have shown that superadiabatic burning velocities and broadened flammability limits can be achieved [11–15]. Figure 1 is a schematic showing the operating principle of the reactor used in this study, known as a counter-flow reactor [16], which features a simple geometry of adjacent straight channels. The counter-flow reactor design is distinguished from many other heat-recirculating reactors by its lack of a predetermined reaction zone or combustion chamber. As a result, operation over a broad







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Fig. 1. Counter-flow reactor utilizes heat transfer from hot combustion products through the channel walls to preheat reactants and achieve superadiabatic combustion.

range of operating conditions can be obtained, as well as stable, non-propagating flame fronts which permit continuous operation over long time spans.

Previous studies on the counter-flow reactor have demonstrated its ability to convert fuel-rich reactants of methane [17], propane [18] and heptane [19] to hydrogen-rich syngas. Recently, this same reactor has been operated with fuel-lean mixtures of methane [4]. In the current study, increasingly complex fuels are studied: propane and heptane. The latter, being a liquid fuel, has importance as a single component surrogate for logistical fuels [20]. Furthermore, the demonstration of fuel-lean liquid combustion in the counter-flow reactor is an important demonstration of its fuel-flexibility for field applications because liquid fuels are generally safer and easier to transport than gaseous fuels.

Counter-flow combustor stability and emissions measurements of carbon monoxide (CO), nitrogen oxides (NO_x) and unburned hydrocarbons (UHCs) are analyzed in this study for lean operation on propane and heptane. Stability is compared for reactor operation in the lean and rich regimes for both fuels. Reactor channel wall temperatures are compared to adiabatic equilibrium temperatures to determine superadiabicity. The results of lean methane, propane and heptane combustion in the counter-flow reactor are examined to understand the influence of superadiabatic operation on operating range and emissions.

2. Materials and methods

2.1. Experimental apparatus

Figure 2 shows the counter-flow reactor, which is the same design used in previous reforming [17–19] and lean combustor [4] studies. It contains four 4 mm high parallel channels constructed of 1 mm thick silicon carbide (SiC) walls. The reactor is 17.3 cm long, with a 91.5 mm long main section and two 40.8 mm long reactor heads. Inlets, outlets, end plugs which prevent the mixing of combustion products and reactants, and two 6.4 mm wide sections of SiC porous foam which act as flow straighteners and flame arresters are located in the reactor heads. Two alumina walls spaced 33.6 mm apart enclose the channels. B-type thermocouples (TCs) are inserted into the channels and sit flush with the channel wall in order to prevent flame holding. Premixed fuel and air are provided to the reactor using mass flow controllers for propane and air, and a pump for heptane. Heptane is vaporized prior to



Fig. 2. Four-channel counter-flow reactor with dimensions and internal components indicated.

entering the reactor. A detailed description of the heptane vaporization system used in this study is available in Belmont et al. [19].

Concentrations of carbon monoxide (CO), nitrogen oxides (NO_x) , and unburned hydrocarbons (UHCs) were measured in the reactor exhaust using Rosemount Analytical analyzers. Samples were drawn through an uncooled quartz probe with a 2 mm inner diameter and inert tubing, and dried before analysis. A previous study [18] showed slightly lower product species concentrations from outside channels, and minimal variation of composition within a single channel except near the alumina walls. In consideration of these findings, exhaust samples in this study were taken from the center channels at the middle of the channel exit. Therefore, the data produced in this study is expected to be representative of interior channels in scaled up reactors.

2.2. Experimental method

The start-up procedure began by burning a near-stoichiometric mixture of the fuel of interest at the reactor outlets at an inlet velocity of 50 cm/s. The reaction fronts propagated upstream in the channels once the reactor heads had warmed sufficiently, and stabilized downstream of the porous SiC flow straighteners. Operating conditions were then gradually adjusted to the reference conditions for each fuel, and were maintained at those values until temperatures stabilized. References conditions for propane and heptane were $\phi = 0.41$ and u = 125 cm/s.

The warm-up phase took approximately 45 min. Experiments included the determination of stable operating conditions and the exhaust gas concentrations of CO, NO_x and UHCs for each fuel over a range of inlet velocities and equivalence ratios. Test conditions were considered stable if combustion was sustained in the main reactor section for 10 min at constant temperatures. Reactor wall temperatures were monitored in each of the four channels throughout the experiments. Gas temperatures were not measured directly due to the tendency of thermocouples inserted into the channel to act as flame holders. Peak wall temperatures were recorded at stable reactor conditions and showed less than ±5 °C variation during that time and while exhaust gas samples were taken. Reactor temperatures were limited to less than 1300 °C in order to avoid damage to the reactor.

Following the warm-up phase, equivalence ratio or inlet velocity was adjusted to the desired test values. Inlet velocity was specified at standard conditions of 25 °C and 1 atm. Emissions results were obtained over a range of inlet velocities with equivalence ratio held constant at ϕ = 0.41, and over a range of equivalence ratios with inlet velocity held constant at 125 cm/s. Emissions concentrations in parts per million (ppm) are reported as measured on a dry basis.

Each operating point was tested twice. Total uncertainty was calculated at each point as the root-sum-square of the contributing uncertainties. The uncertainty of emissions results is due to repeatability of measurements as calculated using a student-t distribution, the calibration gases and the analyzers. Average uncertainties of CO, NO_x and UHC measurements for propane and heptane are ±13, 2 and 16 ppm and ±28, 2 and 1 ppm, respectively. Inlet velocity uncertainty is due to uncertainties of the mass flow controllers for air and propane, the pump for heptane, and the channel cross-sectional area. Equivalence ratio uncertainty is due to contributions from the flow controllers and pump. Average uncertainties of inlet velocity and equivalence ratio are ±10 cm/s and ± 0.03 for propane, and are ± 10 cm/s and ± 0.04 for heptane. Temperature uncertainty is attributed to contributions from repeatability of measurements, the thermocouples, and the data acquisition system. Average temperature uncertainties for propane and heptane are estimated to be ±16 °C and ±20 °C, respectively.

1056

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