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Hydrogen assisted self-ignition of propane/air mixtures in catalytic microburners

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Abstract

A catalytic Pt-based microdevice is evaluated for the combustion of hydrogen and/or propane. It is found that in confined ceramic microchannels hydrogen/air mixtures self-ignite over a wide range of compositions. This discovery is capitalized to self-ignite propane/air mixtures with the assistance of hydrogen addition. It is shown that propane kinetically inhibits hydrogen catalytic combustion at low hydrogen fractions. The minimum hydrogen composition for self-ignition of propane/air mixture compositions is found to be relatively constant, irrespective of propane composition. The transient and steady state behavior of these systems is described, and the minimization of hydrogen usage and startup time are discussed.
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1. Introduction

Microburners are increasingly studied for the catalytic and non-catalytic portable production of heat and/or energy [1–6]. The energy produced can be utilized by various means. Examples include heat generation for remote use, such as for soldiers and space flights, thermoelectrics to produce electricity [7], and heat supply to microscale reactors carrying out endothermic reactions, such as ammonia decomposition or steam reforming [8–11], to produce hydrogen for portable fuel cells. Because hydrocarbons possess a significantly greater energy density than traditional metal acid batteries [4], hydrocarbon-based microburners are

an enticing prospective energy source for portable power applications, such as cell phones, laptops, portable electronics, and personal heaters. Most current prototypes depend on external heating to generate energy for ignition. The additional apparatus necessary to power such external heaters can negate mass advantages of microburners.

One concept for designing a microburner is to downscale conventional scale gaseous burners. However, homogeneous flames are typically quenched when confined in spaces with dimensions below 1–2 mm because of thermal and radical quenching at burner walls [12–14]. As the characteristic length scales of a reactor decrease, the surface area to volume ratio increases, leading to enhanced heat and mass transfer rates between the surface and fluid. As a result, thermal losses to the wall and radical adsorption followed by recombination into stable molecules rise. Despite these challenges, devices capable of self-sustaining

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homogeneous flames in channels with gaps smaller than 1 mm have been developed [1,2].

To complement the aforementioned gas-phase microburner experimental work, computational fluid dynamics simulations in our group were performed to analyze the stability of gaseous microburners [5,6,15,16]. One of the most important findings is that the thermal properties of materials of construction play a vital role in the overall thermal stability of microburners. The reactor walls not only account for heat losses through transverse conduction, but they are often responsible for the majority of the upstream heat transfer, which is necessary to preheat the feed to the ignition temperature. To compound this problem, large wall temperatures and oscillatory instabilities can occur under certain conditions eventually leading to mechanical failure. Even with optimum wall materials, homogeneous combustion allows external heat losses just in the range of free convection for methane/air mixtures and in the low end of forced convection for propane/air mixtures. It becomes clear that while gaseous microburners are feasible to operate, their operation is restrictive.

Combustion at the mesoscale has also emerged including the “swiss roll” heat-recirculating burner with gap sizes of the order of 3 mm, for homogeneous and heterogeneous combustion of propane with air [3,17]. A mesoscale electro-spray device was also developed to atomize liquid fuels by flowing the fuel through a capillary tube and applying a large electrical potential, leading to small droplets that were then mixed with preheated air and fed to a catalytic screen to be combusted [18–20].

We have recently discussed a fabrication protocol of catalytic micro-channel burners along with their performance [21]. Two reactor sizes were discussed, one with a 250 μm gap and another with a 1000 μm gap. These devices were fabricated from alumina, with multiple integrated thermocouples, to measure axial and transversal temperature profiles. Additionally, it was found that propane/air mixtures can be self-combusted in both reactors. The 250 μm gap size reactor exhibited reduced ignition temperatures compared to the 1000 μm reactor (e.g., 150 $^{\circ}\text{C}$ versus 235 $^{\circ}\text{C}$). Finally, the 250 μm gap size reactor achieved nearly complete conversion of propane under most ignited states, whereas the 1000 μm gap size reactor converted less of the propane (60–80%). Overall, confinement (decreased microburner size) leads to an increased performance due mainly to an enhanced mass transfer coefficient.

In this work, we first provide evidence of self-ignition of hydrogen/air mixtures at the small scale in ceramic microchannels with Pt catalyst over a wide range of equivalence ratios. Capitalizing on the self-ignition of hydrogen, a novel idea of igniting hydrocarbons by hydrogen-assisted

catalytic combustion at the small scale is explored. Finally, the synergism of mixture combustion is exploited.

2. Experimental overview

In this work, microburners consisting of a rectangular channel within a cylindrical alumina support are used. A schematic of such a microburner is shown in Fig. 1. The channels are 1 cm wide by 5 cm long with a channel gap of 250 μm giving a reactor volume of 125 μL . A cross-section of one of the reactors is shown in Fig. 1. The dark area in the center is the platinum catalyst. Posts are placed at the inlet in an offset pattern to ensure uniform flow through the channel. Thermocouples are also embedded in the support with the tips flush to the channel wall to obtain axial and transverse temperature profiles. Details of the fabrication are discussed elsewhere [21].

The flow rates of the gases are controlled using mass flow controllers supplied by MKS Instruments. To determine conversions and selectivities, the exhaust gasses are sampled with a probe, which is then sent to a gas chromatograph (GC). A Hayesep column, in conjunction with a molecular sieve with a thermal conductivity detector (TCD), and an HP-PLOT column with a flame ionization detector (FID) are used. Grade 2.0 compressed air, Grade 5.0 compressed nitrogen, and compressed hydrogen are used (Keen Compressed Gases). The propane is >99% pure (Scott Specialty Gases). In all experiments, the total volumetric flow rate at the entrance is kept constant at 2 SLPM, resulting in an average inlet velocity of approximately 13.3 m/s.

The temperatures, conversions, and selectivities are all reproducible to within a few percent over 10–15 h of operation. Deviations in the maximum reactor temperature in propane combustion

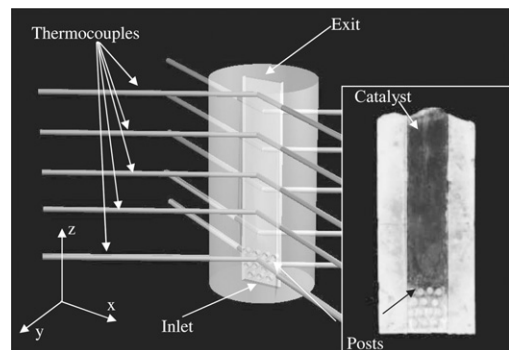


Fig. 1. Schematic showing the placement of the embedded reactor components. The inset is a cross-section of half of a reactor showing the orientation of the inlet posts.

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