

Development of a Pt Nanoparticle-based Microcombustor Power Device

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ABSTRACT

This work discusses design and development of a microcombustor-thermoelectric coupled device using catalytic combustion with platinum nanoparticles. Previously, our work successfully demonstrated room temperature ignition and control over catalytic activity of methanol-air mixtures with platinum nanoparticles. Current efforts are focussed on using our catalytic system to design a microcombustor power source by integrating it with thermoelectric generators. For this work, commercial thermoelectric generators were used to convert the temperature gradient created by catalytic combustion directly to electrical power. Specifically, a planar reactor assembly was designed and tested to optimize temperature gradients and therefore power output. Alternative fuels were explored for optimizing the microreactor performance. Key parameters that influence performance and contribute to the modeling efforts were identified.

Keywords: microcombustion, catalysis, nanoparticles, platinum, thermoelectrics

1 INTRODUCTION

Microcombustors coupled with thermoelectric devices have the potential to provide a prolonged power source for portable devices as opposed to conventional batteries due to significantly higher energy densities of hydrocarbon fuels [1], [2]. An additional, advantage of using hydrocarbons as an energy source is that recharging then can be easily achieved by simply adding more fuel for such integrated devices, making them desirable for remote operations. Current research is primarily focused on the design of microreactors to sustain combustion and maximize temperature gradients [3]. A number of noteworthy power devices based on microcombustion have been investigated [3]-[7]. The challenge however is to develop a device that performs reliably and surpasses with conventional battery energy density.

Our previous work, demonstrated a stable and repeatable room temperature ignition of methanol-air mixture using platinum (Pt) nanoparticles in sub-millimeter channels [8]-[10]. This work was conducted using Pt nanoparticles with average diameters of 8 nm deposited on cordierite substrates. Considering the tendency of

nano particles to sinter at relatively low temperatures, our system demonstrated stable catalytic combustion with temperatures greater than 500 °C. Furthermore, the temperatures can be controlled by altering the reactant flow rate and catalytic loading to achieve fuel conversion rates up to 60%. For this work, a new planar-faced reactor was designed to host the cordierite substrate and integrate the reactor with thermoelectric modules. Results indicated appreciable temperature gradients were achieved with a first generation design. Additionally, this work documents an exploratory study on alternative commercial fuels for the catalytic microcombustion.

2 EXPERIMENTAL

The details of experimental setup and procedure are described elsewhere, however, they are briefly discussed here. Pt nanoparticles were synthesized using colloidal synthesis method proposed by Bonnet et. al. [11]. The resulting 8 nm particles, verified using TEM imaging, were deposited on cordierite substrates with 800 μm channel width. The substrate channels were coated with Pt nanoparticles on all four sides using the draw-coating method. Substrates were cylindrical with 13.5 mm diameter and 19 mm length. Figure 1 provides a schematic of the power generation device. The substrates, surrounded by quartz wool, were housed in an aluminum reactor (external dimensions of 36 mm x 30 mm x 20 mm). The reactor exhibited planar faces for mounting thermoelectric modules. Thermoelectric modules consisted of two 2.5W Hi-Z Technology, Inc generators. Folded-metal computer heat sinks were used as cold junctions for the device. Temperature measurements were conducted using K-type thermocouples located at the locations indicated in Fig. 1. Near stoichiometric methanol-air mixture was supplied to the Pt-coated substrate using a glass bubbler.

A mass-flow controller was used to deliver specified flow rates. Typical runs involved cycling flow of the fuel-air mixtures on and off, while measuring the substrate and other critical spatial temperatures, as indicated in Fig. 1. Of these, the temperature difference between the hot and cold side of the thermoelectric module was important. The greater the temperature difference the

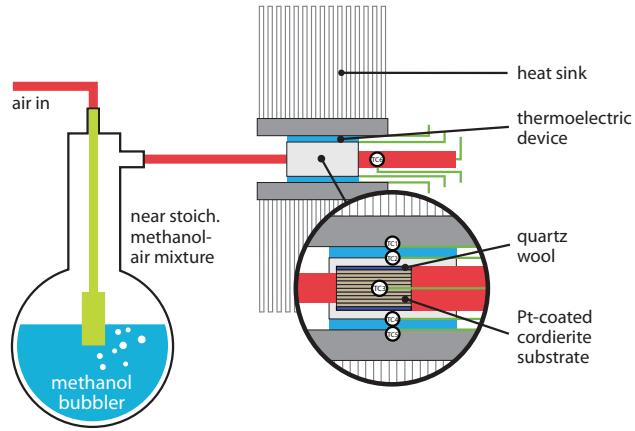


Figure 1: Experimental schematic of the microcombustion-thermoelectric coupled device. The vertical device orientation is for illustration purpose only. Actual device was oriented horizontally to obtain symmetric temperature gradients on either side of the reactor.

greater the power output.

For the alternative fuel study, a tubular aluminum reactor from the previous studies was used. For these studies, only the substrate temperature measurement was necessary to assess fuel performance. Fuels included methane, propane, butane and ethanol as commercial alternatives to methanol. Gaseous fuels were stoichiometrically premixed, whereas, ethanol was heated to 45 °C in the bubbler to achieve stoichiometric fuel-air mixture. Furthermore, ethanol required heating of the inlet line to prevent condensation.

2.1 RESULTS AND DISCUSSION

2.2 Alternative Fuel Study

Preliminary tests indicated none of the alternative fuels ignited at room temperature. The results were in contrast to the methanol-air mixtures studies from previous work. Preheating of the reactant line at approximately 200 °C was required to achieve ignition. Figure 2 provides substrate temperature history of three fuels: ethanol, propane and butane. Methane results are not included in Fig. 2, because catalytic combustion of methane was achieved only for the first cycle and failed to combust for the remainder of the experiment. Methane results are in agreement with its combustion chemistry.

Figure 2 provides representative results of multiple catalysis experiments conducted for each fuel. Stable catalytic combustion temperatures are achieved for propane and butane fuels with peak cycle temperatures remaining nominally constant. However, ethanol catalytic combustion behavior is noticeably erratic with multiple

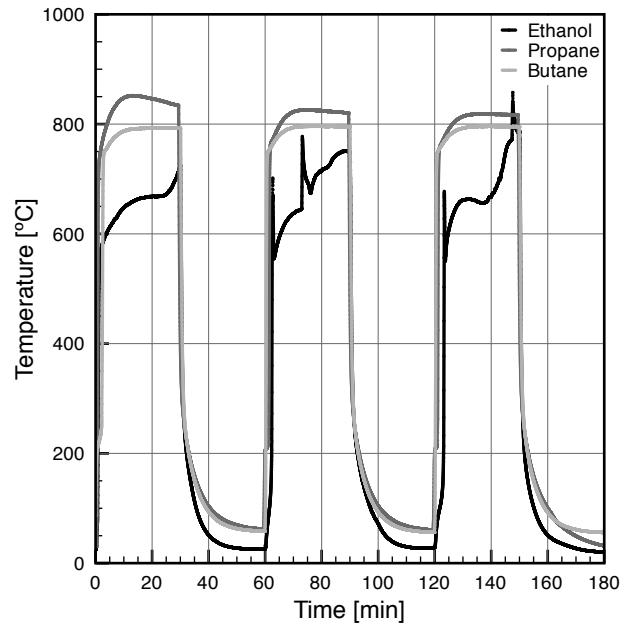


Figure 2: Catalytic temperature histories of ethanol, propane and butane flowing at 800 mL/min with two-side coated substrates. All tests involved 200 °C pre-heating of the inlet flow to achieve ignition

spikes in temperature at various stages of catalytic cycle. Similar ethanol behavior was observed over multiple experimental runs. Comparing the mean catalytic temperatures, the result indicate temperatures are not in agreement with the heating values of each fuel. For instance, butane posses the largest lower heating value (LHV), however the results point to more intermediate substrate temperatures in comparison. The trend suggests fuel conversion rate is a function of fuel type. Further analysis of exhaust products is required to elucidate the trends and the ethanol behavior. Overall, the study established methanol as a more desirable fuel for microcombustion, particularly for its ability to ignite at room temperature.

2.3 Power Generation Device

Multiple aluminum reactors were fabricated with varying dimensions and substrate locations. Substrates with four Pt-coated sides were used for these experiments. The temperatures at the hot-side of the thermoelectric module and the time response were used as metrics for comparison of reactor performance. Furthermore, three different commercial heat sinks were tested to maximize the temperature difference. Highly effective heat sinks (with maximum cooling) had detrimental effect on the combustion due to quenching of the catalytic reactions. Multiple exhaust configurations were explored to maximize heat transfer through the reactor. Uniform surface

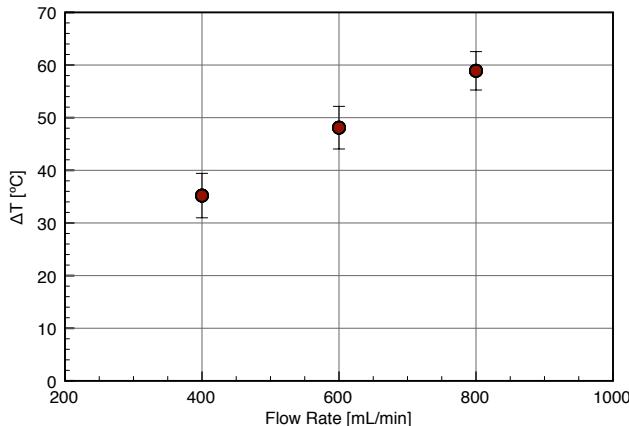


Figure 3: Average temperature differences between hot and cold side of the thermoelectric module as a function of methanol-air mixture flow rate. Each data point represents average values from two symmetric thermoelectric modules over multiple cycles and runs. Error bars represent a single standard deviation.

temperatures were achieved for the aluminum reactor; eliminating the need for thermal spreaders used in other designs. The final design was tested for temperature difference over the thermoelectric modules for multiple reactant flow rates.

Figure 3 presents data from stoichiometric methanol-air mixtures at three different flow rates. The temperature difference represents the mean difference in temperature between the hot and cold sides of the thermoelectric modules. Each data point represents results from three different experimental runs. Figure 3 shows the temperature difference increases with increasing flow rate. This trend is expected considering the increased heat release as function of increased reactant flow rate. With the power output from a thermoelectric module directly proportional to the temperature difference, the maximum electrical power output can be expected with 800 mL/min reactant flow rate.

Since electrical power output from the thermoelectric modules were not obtained, theoretical power output based on device performance specification can be estimated. In addition, based on previous fuel conversion data and LHV of methanol, the system efficiency can be calculated. Estimates suggest initial device efficiency of $\sim 0.1\%$. While higher flow rates are possible to increase the temperature difference, the higher flow rates also lead to higher catalytic temperatures that in turn increase nanoparticle sintering rates. The optimum flow rate is yet to be explored. On the other hand, improved thermal management using reactor redesign has the potential to dramatically benefit device efficiency. For instance, there is a large temperature difference be-

tween the substrate and the hot side of the thermoelectric module. A redesign that reduces this temperature differential can greatly improve efficiency. Efforts are currently underway to alter the substrate configuration to better match device assembly.

3 CONCLUSION

This work provides important experimental data on design of a microcombustion-thermoelectric power device using Pt nanoparticles as catalysts. While the efficiency of the current device is low, it is important to note that such a device with 1% efficiency can prove competitive with conventional battery technology, due to the high energy densities associated with hydrocarbon fuels. The results also contribute towards developing a theoretical model for further optimization of the design. More importantly, the results provide a viable template towards a highly miniaturized design for a microcombustion-thermoelectric coupled power device.

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