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## A CATALYTICALLY SUSTAINED MICROCOMBUSTOR BURNING PROPANE

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### ABSTRACT

The performance of a catalytically sustained microcombustor was investigated. The reactants for combustion were air and propane, which were mixed together and delivered to the end of a bayonet-style, heat recirculating combustor having a reaction volume measuring approximately  $0.25 \text{ mm}^3$ . Platinum wire in the shape of a coil was used as an active surface to catalyze the exothermic reactions. Measurements of temperature and reactant flow rate were made during the study. Results show that propane can be burned in a self-sustaining fashion in the microcombustor with heat release rates as low as 1.0 Watt.

### INTRODUCTION

Combustion processes play an important role in modern society for generating both heat and, through conversion, electrical power. New technological developments are calling for miniaturization of energy systems including combustion-based power production. To date, little has been accomplished in developing practical combustion systems in the microscale regime. What makes this area attractive is the energy density of hydrocarbon-based fuels: stored liquid fuels when burned with oxygen from the atmosphere have several times the energy density of today's batteries. In fact, this energy density is on the order of 50 kJ/gm, while that of a conventional battery is less than 1 kJ/gm. Because of this, it is of great interest to study the feasibility of developing microscale combustion systems as potential replacement for conventional batteries. The goal of this study is to investigate the behavior of a microscale heat recirculating combustor to determine the feasibility of developing a heat source for various small-scale energy applications.

A number of experimental challenges exist in

miniaturizing combustion. First, quenching must be prevented in order to create a self-sustaining microcombustor. Quenching will come in the form of both free radical destruction at the walls of the combustor and in heat loss to the surrounding structure. Consideration of radical destruction leads to the idea of employing active surfaces to promote reactions rather than hinder them. Second, heat loss from several mechanisms could lead to quenching of the exothermic reactions by reducing the temperature to such a point that self-sustaining reactions are no longer possible. In order to mitigate this effect, careful thermal management of the combustor is needed. Heat recirculation and thermal insulation are two methods being considered.

This paper describes a microcombustor having a total reaction volume of less than  $0.25 \text{ mm}^3$  and capable of burning propane. This study continues the work of Peterson and Vanderhoff [1] where a vacuum insulated microcombustor was investigated. In this prior work, hydrogen was used as the fuel and heat release rates as low as 200 mW were observed. This work continues the basic idea of a heat recirculating microcombustor where propane is used as the fuel. As suggested in the earlier work, the present investigation shows that heat loss is the fundamental mechanism dictating the ultimate size of the combustor when catalytic surfaces are used for preventing free radical quenching. This paper will first cover past work on microcombustion. Then a description of the experimental methodology will be given with an emphasis on the construction of the microcombustor. Finally, results will be given on the combustion of propane-air mixtures for a range of flow rates and stoichiometries.

## BACKGROUND:

Because microscale combustion requires minimizing the heat loss to the surroundings, heat recirculation becomes an important consideration. This particular area of the combustion literature is extensive and will not be reviewed here. It is, however, important to acknowledge the contributions made by Weinberg [2,3] to the field. He has developed both theoretical and experimental techniques to study heat-recirculating combustors and has put forth a large number of designs to implement the concept. Our microcombustor uses a counterflow, bayonet-style configuration to recover some of the thermal energy in the product stream. The bayonet configuration allows the hot end of the device to be thermally isolated from the surrounding structures kept at, or near, room temperature.

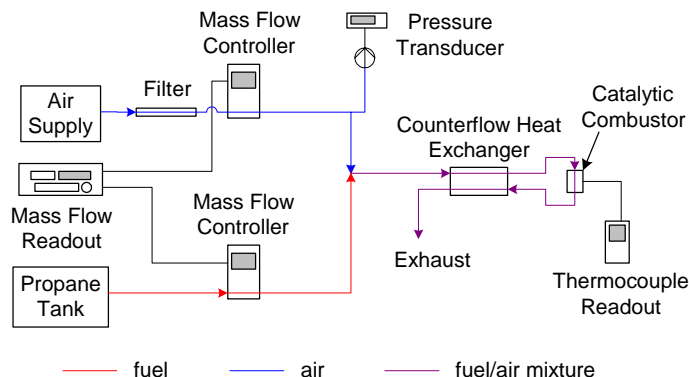
Peterson and Vanderhoff, as mentioned earlier, experimentally demonstrated a combustor with a reaction volume of approximately  $0.2 \text{ mm}^3$ . This was accomplished using a platinum catalyzed reaction involving hydrogen and air. The work also demonstrated the dependence of reaction temperature on heat loss from the reaction zone. To minimize heat loss, Peterson and Vanderhoff isolated the test section in a vacuum chamber. This reduced the heat loss by convection and conduction. Computational modeling of the heat loss from microscale bayonet-style heat exchangers has also been accomplished by Peterson [4]. The work investigated the primary mechanisms of heat loss from elevated temperature devices and identified the major heat loss routes as a function of characteristic size.

Sitzki et al. [5] have developed microscale excess enthalpy burners fabricated using a process called EFAB. The process can produce arbitrary 3-D structures by stacking hundreds of individually patterned layers. Using this technique, the researchers have built and tested both two-dimensional and toroidal versions of their burners that rely on heat recirculation for thermal management. The toroidal version is in the form of a “Swiss roll” where the hot region of the burner is isolated from the surroundings by the inward spiral of reactants and the outward flow of products.

## EXPERIMENTAL SECTION:

The following will describe the micro-combustor test set-up. The goal of this work was to reduce the size of a combustion chamber to less than one cubic millimeter while burning a propane/air mixture. Several mechanisms were used to aid in both the initiation and the sustaining of the combustion reactions. First, platinum was used as a catalyst to decrease both the activation energy of the fuel and the deactivation of radicals at the wall of the combustor. Second, heat recirculation from the exhaust gases was used to preheat the reactant mixture in a counterflow heat exchanger arrangement. This preheating increases the adiabatic flame temperature of the reaction, further decreasing quenching effects. Third, quartz tubing was used for the walls of the chamber. This reduced the heat loss using a low thermal

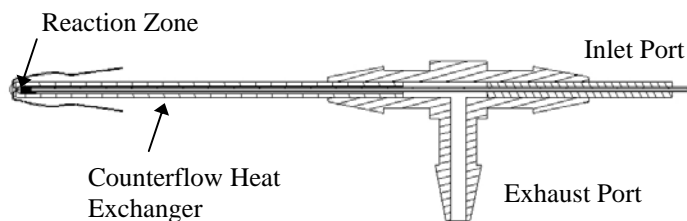
conductivity material while still allowing the system to run at elevated temperatures. It is theorized that these effects will be sufficient to counteract the losses experienced in the microscale regime resulting in a self-sustaining combustion reaction in chambers significantly smaller than the characteristic quench distance.



**Figure 1: Diagram of the experimental test set-up for measuring temperature and reactant flow rates.**

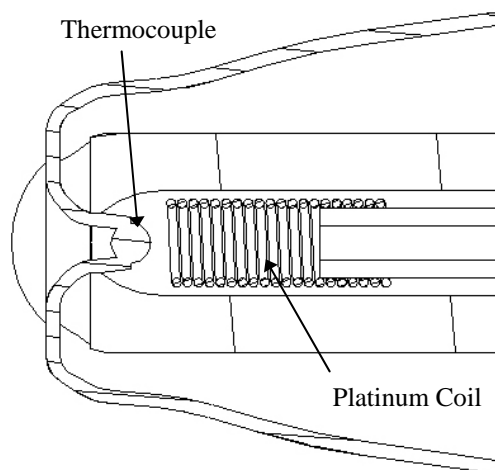
A schematic of the experimental set-up is shown in Fig. 1. The fuel was supplied by an MKS mass flow controller, model M100B11CP1BV, from a propane tank. The use of flow controllers was critical to the success of this study because of pressure variations (during operation) caused by a number of different mechanisms. These effects will be discussed later. Air was also supplied via an MKS mass flow controller, model M100B12CP1BV, from pressurized filtered building air. A Validyne pressure transducer, model DP15-42, was placed in the airline near the air/fuel mixing section to monitor the inlet pressure of the combustor.

The counterflow heat exchanger and combustion chamber were assembled as one system. Figure 2 shows a cross section of the device. The counterflow heat exchanger was first created by joining a 27-gauge stainless steel (SS) tube, O.D. of 0.4 mm, to a T-section fitting with epoxy such that one end projected out approximately two inches. A closed end quartz tube could then be slipped over the top of the stainless steel tubing. The reactant gas mixture is supplied through the T-section inside the SS tubing until it reaches the end of the quartz envelope, at which point the flow is forced back around the outside of the SS tubing to exit from the side port of the T-section.



**Figure 2: Sketch of the counter-flow microcombustor arrangement showing reactant inlet and product outlet along with the thermocouple at the hot end of the device.**

The combustion chamber was constructed in two steps. First, the platinum coil was prepared by wrapping 50- $\mu\text{m}$ -dia. platinum wire around the above-mentioned SS tubing, then sliding approximately 1 - 2 mm of the coil off the end. The quartz envelope was then prepared for assembly. First, one end of a 0.6 mm ID quartz tube was closed via an acetylene torch. A slot was then cut in the tip of the closed end perpendicular to the length of the tube such that a small opening was produced. This was accomplished by fastening the tube to a cutting fixture and using a diamond blade wire saw with a 250  $\mu\text{m}$  blade. A 75- $\mu\text{m}$ -dia. type-K thermocouple was inserted into the opening and cemented into place using Omegabond 300. A Tektronix thermocouple readout, model DTM 920, was attached to the thermocouple leads to display the temperature of the reaction zone. The prepared quartz envelope was slid over the top of the SS tube/platinum coil assembly and sealed in place using silicone adhesive. A close-up of the combustion region is shown in Fig. 3.



**Figure 3: Close-up of the hot end of the microcombustor showing inner delivery tube, platinum coil, and thermocouple placement.**

A removable heating coil was constructed by wrapping heating wire around the outside of a 3 mm quartz tube. The wire ends were bent to create leads in order to attach

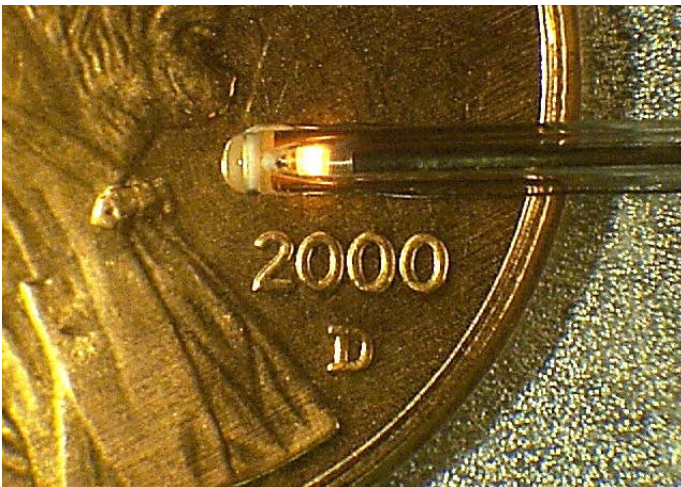
the coil to a power supply. The heating coil was placed over the closed end of the quartz envelope (without contacting) to surround the reaction zone. In addition, a Plexiglas shield was fabricated and placed around the test system to reduce the convective heat transfer effects of air circulation in the room.

The propane/air mixture ratio was initially set to stoichiometric to begin the experiments. Heating the combustion region to a range between 400 and 500  $^{\circ}\text{C}$  was sufficient to ignite the mixture on a properly prepared platinum surface. The external heating coil could then be removed. Stable operation of the combustor was determined by letting the reaction zone temperature, as measured by the embedded thermocouple, to arrive at a constant temperature. After this was achieved, the fuel and airflow rates were slowly adjusted to explore the rich and lean operation of the combustor. Fuel flow rates used in this experiment were set to 0.5, 1, 1.5, and 2 sccm. The airflow rate was adjusted either higher or lower than stoichiometric at a particular fuel flow rate in order to collect mixture ratio data. Most measurements were recorded while the temperature was constant and stable. However, at certain fuel flow rates and at the rich end of the mixture ratio range some instability was observed. This will be explained in more detail later.

There were several experimental details investigated in this research. One such detail was the ignition of the reaction. It was found to be very difficult to ignite the surface catalytic reactions with the heating coil if the platinum wire was not prepared first in an open-air flame, i.e. without the quartz envelop in place. This indicates that there is a necessary surface preparation in order to facilitate thermal ignition. Surface preparation in this work consisted of producing a surface reaction in the atmosphere via a flame before inserting the coil assembly into the quartz envelope.

Thermocouple placement was also an important issue in this experiment; first in fastening the thermocouple to the quartz envelope and then in its placement. The fastening of the thermocouple was of concern because of the high temperatures at which the system operated. Omegabond 300 was operable at the elevated temperatures, but its thermal expansion was different enough from quartz that separation occurred in early experiments. For this reason, a thermocouple-embedding scheme, as described above, was developed and implemented to remove thermal expansion effects. Thermocouple placement was important in that the desired temperature to be measured was the temperature of the reaction zone. To accomplish this, the thermocouple was placed inside the tip of the quartz envelope. One concern with this thermocouple location was the effect of radiation on the thermocouple measurement. A thermal model was developed to aid in characterization of the path of heat transfer from the reaction zone to the thermocouple. It was found that the radiation heat transfer from the platinum surface was negligible compared to the convective heat transfer of the hot gases flowing over the thermocouple bead.

Flow rate measurements were of critical importance in this investigation as well. When first running the experiment, it was found that the size and temperature of the reaction zone significantly affected the flow regime. When combustion was initiated, the temperature of the combustion region would significantly increase, causing an increase in pressure in the upstream section, which would in turn cause a change in the volumetric flow rate and mixture composition. Mass flow controllers as described above were used because they regulate flow on a mass basis, thus nullifying the varying effects of temperature and pressure on the propane/air mixture. Inserting mass flow controllers into the air and fuel lines allowed for variation in upstream pressure while maintaining consistent mixture flow rates.



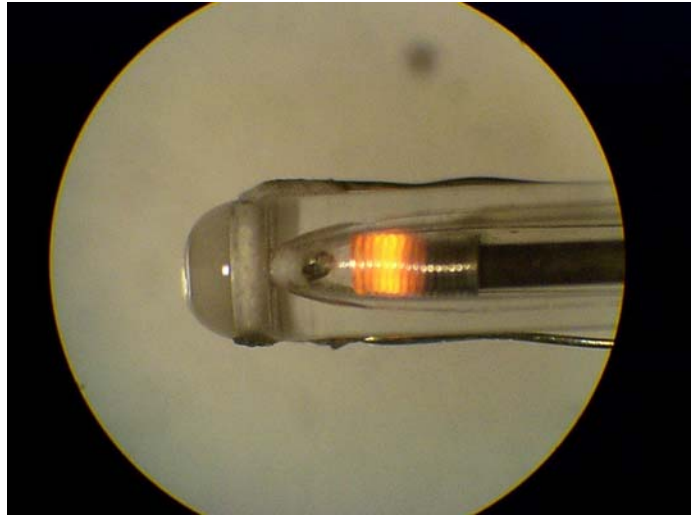
**Figure 4: Microcombustor operation on propane and air.**

## RESULTS AND DISCUSSION:

A picture of the operating microcombustor is shown in Fig. 4. The exothermic reactions appear to be taking place on the surface of the platinum coil. The thermocouple can be seen just to the left of the glowing coil. Figure 5 shows a close-up picture of the surface reactions at a low fuel flow rate. The brightness of the surface was found to be directly proportional to the fuel flow rate and dependent on the stoichiometric mixture ratio.

The test results are presented in Fig. 6 in the form of a measured hot end temperature vs. equivalence ratio. The curves show an increase in reaction temperature with increasing fuel flow rate. This increase does not appear to be linear, but rather to asymptotically approach some maximum temperature at higher rates of fuel flow. This may be caused by an increase in convective heat loss and decreasing diffusion of active species to the platinum surface as reactant flow increases. Also shown in the graph is an inverted U-shape

trend indicating a maximum reaction temperature exists for a given fuel flow rate. This maximum temperature occurs at slightly rich conditions. The surface area of the platinum coil, the amount of corrosion in the system, and the location of the thermocouple with respect to the reaction zone directly influence the measured maximum temperature of the reaction zone. The results also show a decrease in combustion temperature and flame stability toward the rich flammability limit.



**Figure 5: A close-up of the microcombustor at a low flow rate of propane and air.**

Some interesting observations were made while running the tests. For higher fuel flow rates, the reaction began to oscillate significantly at slightly rich mixture ratios. For example, at a fuel flow rate of 2 sccm, the temperature measurements began to oscillate  $\pm 150$  °C for an equivalence ratio of 1.3. Oscillations were also observed at 1.5 and 2.5 sccm. At the lower flow rate, they were not as pronounced, but at 2.5 sccm, the amplitude of the oscillations made data collection impossible. These oscillations were coincident with a change in location of the reaction along the coil. As the system would oscillate, the reactions seemed to work their way from the end of the platinum coil down to the stainless steel tube and then back again.

Lack of insulation around the quartz tubing also proved to be a source of system instability. Convective air currents on the quartz envelope caused a decrease in the reaction temperature and subsequently a decrease in the brightness of the platinum surface emissions. This is consistent with the notion that increasing the convective heat transfer from the quartz tube decreases the inner tube wall temperature and increases heat loss from the reaction zone.

Uncertainty in this experiment was calculated for both the reaction temperature and the mixture ratio. For the reaction temperature, uncertainty included specified values for the thermocouple and thermocouple readout as well as estimated contributions from the sampling rate and readability. It was found that the major contributor to uncertainty was that associated with the repeatability of the measurements. This was due primarily to the oscillations as mentioned earlier. At higher temperatures, the uncertainty in the thermocouple also played a significant role. The values of uncertainty ranged from  $\pm 12$  °C for stoichiometric mixtures to  $\pm 30$  °C at rich and lean conditions. Uncertainty in the oscillatory data was estimated to reach  $\pm 100$  °C for the higher flow rates.

For equivalence ratio, the uncertainty was calculated by taking into consideration the uncertainty specified for the flow controllers. It was found that at low flow rates, the specified uncertainty was a larger percentage of the actual flow rates, thus significantly increasing uncertainty for lean mixtures. Uncertainty of the equivalence ratio was calculated to be  $\pm 0.30$  when airflow rates were low and  $\pm 0.03$  when airflow rates were high.

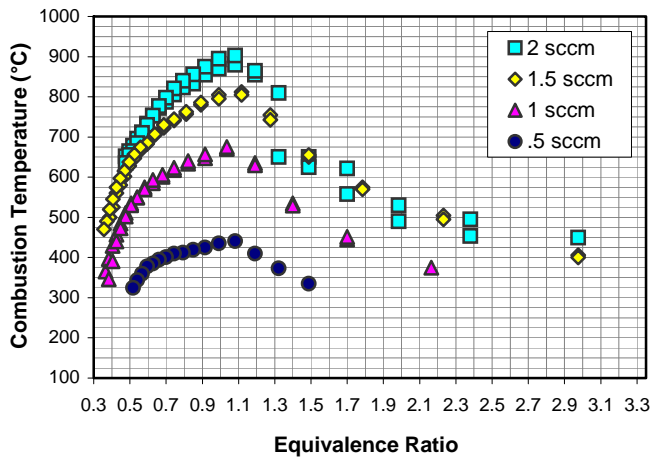


Figure 6: Results from the catalytic microcombustor experiment using propane.

### CONCLUDING REMARKS

In this set of experiments, a test system was developed to aid in the prediction of reaction behavior for microcombustor systems. This involved miniaturization of a catalytic combustor and measurements of the reaction zone temperature. Reaction temperature was plotted against equivalence ratio for a 0.25 mm<sup>3</sup> combustion region. Results showed that a self-sustaining combustion reaction could be attained in the microscale regime using propane and that reaction temperatures ranged from 400 to 900 °C depending on the fuel flow rate.

There are a number of factors that appear to affect combustion at the microscale level. Of interest in this research

was heat loss from the reaction zone of the combustor. More research is needed to quantify these effects and to determine the ultimate constraints on microcombustor size.

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