Plastic mesocombustors

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Abstract
Recent experimental and theoretical studies of heat-recirculating combustors have demonstrated the importance of thermal conduction through the structure of the combustor on its performance. In particular, this solid-phase heat conduction inevitably degrades performance via transfer of heat out of the reaction zone to the surrounding structure, which is then lost to ambient. This in turn leads to a reduction of reaction temperature and thus sustainable reaction rates. By use of platinum-based catalysts in spiral counterflow "Swiss roll" heat-recirculating combustors, we have been able to sustain nearly complete combustion of propane-air mixtures at temperatures less than 150°C using combustors built with titanium (thermal conductivity (k) of 7 W/m°C). Such low temperatures suggest that high-temperature polymers (e.g. polyimides, k = 0.3 W/m°C) may be employed as a combustor material. With this motivation, a polyimide Swiss roll combustor was built using CNC milling and tested over a range of Reynolds numbers with propane fuel and Pt catalyst. The combustor survived prolonged testing at temperatures up to 450°C. Reynolds numbers as low as 2 supported combustion, with thermal power as low as 3 watts and temperatures as low as 72°C. These initial results suggest that polymer combustors may prove more practical for meso- or microscale thermochemical devices due to their lower thermal conductivity and ease of manufacturing. Applications to electric power generation via single-chamber solid oxide fuel cells are discussed.

Introduction
It is well known that hydrocarbon fuels contain 100 times more energy per unit mass than lithium-ion batteries, thus devices converting of fuel to electricity at better than 1% efficiency represent improvements in portable electronic devices and other battery-powered equipment [1]. At small scales, however, heat and friction losses become more significant, thus devices based on existing macro-scale designs such as internal combustion engines may be impractical. Consequently, many groups have considered heat-recirculating burners or “excess enthalpy” burners first studied 30 years ago [2,3] for thermal management and thermolectric, piezoelectric or pyroelectric devices, having no moving parts, for power generation. In heat-recirculating burners, by transferring thermal energy from the combustion products to the reactants without mass transfer (thus dilution of reactants), the total reactant enthalpy (sum of thermal and chemical enthalpy) is higher than in the incoming cold reactants and therefore can sustain combustion under conditions (lean mixtures, small heating value fuels, large heat losses) that would extinguish without recirculation.

At smaller scales heat losses become more important due to increased surface area to volume ratios. A potential means of reducing the impact of these losses is to employ catalytic combustion, which may allow lower temperatures and thus allow reaction to occur at higher heat losses than can possibly be sustained with non-catalytic combustion. The higher surface area to volume ratio at small scales makes area-limited catalytic combustion even more attractive as compared to volume-limited gas phase combustion. There are additional potential advantages of catalytic combustion as well. Since chemical reactions only occur on the catalyst surface, the location of the heat source is fixed. This makes heat transfer design simpler than gas phase combustion in which the location of reaction zone may change in undesirable ways. Also, the lower temperature of catalytic combustion makes thermal stresses and materials limitations less problematic.

In particular, our experiments and modeling [4,5] have shown that counter-current heat-recirculating reactors require thin walls with low thermal conductivity for maximum performance at small scales. Normally metallic or ceramic reactors are required for high temperatures, but metals have high thermal conductivity (thus relatively poor performance in small-scale devices) and thin-walled ceramics are too fragile and still have marginally acceptable thermal conductivity. Instead, we construct our reactors from polyimide plastics, which have far lower thermal conductivities than metals (typically 100x lower) or even ceramics (typically 10x lower). Polyimides have excellent high temperature resistance compared to practically all other plastics. Plastics have the additional advantages of low cost, ease and variety of fabrication techniques, durability and electrical insulation properties. The ease of fabrication moreover enables us to pursue more complex Swiss roll configurations as may be necessary for maximum fuel utilization.

Specific Objectives
Our experimental and theoretical studies [4,5] of heat-recirculating combustors clearly confirmed that thin walls with low thermal conductivity materials are needed for maximum performance at small scales. We have been able to sustain reaction of propane-air mixtures with Pt foil catalyst at temperatures less than 150°C using combustors

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built with titanium. Such low temperatures suggest that polyimides may be employed as a combustor material.

Consequently, the objectives of this work are to build and test polyimide Swiss roll combustors over a range of Reynolds numbers and assess the feasibility of polyimide Swiss roll combustors for self-sustained thermal management at the low Reynolds numbers that would be encountered in a microscale device.

**Experimental Apparatus**

The concept of polymer reactors were tested using reactors made from commercially available DuPont™ Vespel™ or Kapton™ polyimides. These polymers are completely non-flammable in air; the minimum oxygen concentration that sustains combustion is 53% / 45% for Vespel™ / Kapton™. Vespel™ is a hard plastic, typically sold in block form, that is ideally suited for CNC milling; the reactors (Fig. 1) were milled automatically from SolidWorks™ CAD files; with this scheme we were able to design, build and test new design configurations in 1–2 working days. The outside dimensions of Vespel™ burner are 46 mm wide by 46 mm deep by 25 mm tall with 0.5 mm wall thickness. The gap-width for each inlet and exhaust channel is 3 mm. Due to the limitation of CNC milling technique, very thin-walled structures are difficult to mill, another simple fabrication technique that is to fold 0.25 mm thick sheets of Kapton™ into spirals and glue them to base made by same material (Fig. 2) was developed. The outside dimensions of Kapton™ burner are 15 mm wide by 15 mm deep by 15 mm tall. The gap-width for each inlet and exhaust channel is 1 mm.

The performance of these reactors was tested using propane fuel and Pt foil catalyst specially treated with NH$_3$, which we have found yields far superior performance at lower temperatures [4], were placed along the walls at the center of the burner. The top of the burner is sealed with fibrous ceramic blanket, backed by aluminum plates, secured with Heat Shield ceramic adhesive. The fresh fuel-air mixture is plumbed through a manifold attached to the inlet of the Swiss roll. Electrically heated Kanthal wire was used for ignition. The burner was instrumented with thermocouples located at the center and in each inlet and exhaust turn (six total). Hastings mass flow controllers were used to regulate the flow rate of fuel and air through the burner. Labview data acquisition software was used to record the response of each thermocouple and to control the mass flow controllers.

**Results and Discussion**

Recently, our experimental and theoretical studies [4,5] of heat-recirculating combustors indicate that optimal combustor performance could be achieved by reducing wall thermal conductivity. A Vespel™ (k ≈ 0.29 W/m°C) Swiss roll that was geometrically similar to the 3.5-turn titanium (k ≈ 7 W/m°C) combustor used previously [4] was constructed to test this hypothesis.

Figure 3 shows a comparison of the extinction limits obtained between with the Vespel™ and titanium Swiss rolls. These limits are shown in Figure 3 over the range of Reynolds numbers tested. The Reynolds number is defined based on the area-averaged gas velocity at the inlet, the gas kinematic viscosity at the inlet, and the gap width. Lean and rich extinction limits were determined by starting from a steady burning state and decreasing or increasing the fuel concentration with the igniter off.

The figure demonstrates that reducing wall thermal conductivity extends extinction limit to lower Reynolds numbers and allows weaker mixtures to be burned, particularly at low Reynolds numbers when heat loss becomes predominant. For both the Vespel™ and titanium
burners, the lean flammability limit is actually rich of stoichiometric at low Reynolds numbers.

Figure 4 shows the maximum temperatures at the lean and rich extinction limits obtained over the range of Reynolds numbers tested for both the Vespel™ and titanium reactors. In other words, the minimum temperatures capable of sustaining reaction (at the center of the burner) at the Reynolds numbers indicated are shown. We have been able to sustain reaction of propane-air mixtures at temperature of 72 °C using Vespel™ Swiss roll reactor and even lower temperature of 60 °C using titanium Swiss roll reactor.

Note that the maximum temperatures obtained for both the lean and rich extinction limits are in good agreement with each other. Note too that the minimum temperature required to sustain combustion exceeds Vespel™ material limit at Reynolds number 20 and higher. Therefore, polymers are not suitable material of reactor for high Reynolds numbers, but for low Reynolds numbers that meso- or microscale devices should be more concerned.

CNC milling is not suitable fabrication technique for small-scale reactor. The primary limitation of this technique is that very thin-walled structures preferred for maximum performance are difficult to mill. A second, even simpler fabrication strategy developed was to fold very thin sheets of Kapton™ into free-standing spirals and glue them to a polymer base. Moreover, the thermal conductivity of Kapton™ (k ≈ 0.12 W/m°C) is less than half of thermal conductivity of Vespel™ (k ≈ 0.29 W/m°C).

The maximum temperature recorded by thermocouple in the center of both Vespel™ and Kapton™ reactor is shown for a range of fuel concentrations in Figures 5. The results shown in Figure 5a were obtained from Vespel™ reactor and those in Figure 5b were obtained from Kapton™ reactor. For both the CNC-milled Vespel™ and the much smaller, folded Kapton™ reactors, continuous, sustained operation of these reactors at temperatures up to 450°C were demonstrated. These temperatures are high enough to provide excellent single-chamber solid oxide fuel cells (SOFC) performance (not shown) when using our ruthenium and rhodium anode catalysts [6,7]. Moreover, the external temperatures of reactor were below 50°C in practically all cases, which leads to minimal thermal signature and touch-temperature hazards. Consequently, we can conservatively design devices for 400°C continuous operation, thus conservatively 200 mW/cm² power density (not shown), with the likely possibility of 500°C / 600 mW/cm² operation [6].

Conclusions

Experiments mimicking combustion in microscale combustion devices at low Reynolds numbers are possible, but require heat-recirculation via the Swiss roll or similar heat exchanger geometries, as well as catalytic combustion. Experimental results reveal that optimal performance can be obtained by constructing burners using a thin material with low thermal conductivity, as suggested [4,5]. Reducing wall thermal conductivity leads to lower heat losses and therefore increases operating temperatures and extends flammability limits.
Polymer combustors are feasible for meso- or microscale thermochemical devices and may prove more practical due to their lower thermal conductivity, low cost, durability, ease and variety of fabrication techniques, and electrical insulation properties.

Furthermore, by developing and incorporating advanced fuel cell electrocatalysts [6,7] that enable reduced temperature operation (350 - 500°C), utilization of plastics for reactor components becomes practical.

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References