MESOSCALE POWER GENERATION BY A CATALYTIC COMBUSTOR USING 
ELEcroSPRAYED LiQUID HYDROCARBONS

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The development of a mesoscale catalytic combustor to be coupled with direct energy conversion modules for power production is presented. The combustor has a volume on the order of a few cubic centimeters and operates on JP8 jet fuel, which is electrosprayed at a flow rate on the order of 10 g/hr and equivalence ratios varying from 0.35 to 0.70. Temperatures in the range 900–1300 K are achieved with a ± 5% uniformity over the top circular surface of the burner. Using gas chromatographic analysis of the exhaust gases, a combustion efficiency on the order of 97% is estimated. Remarkably, no fouling, nor soot, nor NOx were detected in the exhaust gases. The system resulted in clean and efficient combustion of even environmentally problematic liquid hydrocarbons.

Introduction

The development of microelectromechanical systems technology in the past 10 years has spurred interest in the realization of combustion on scales much smaller than previously explored. The primary incentive for using combustion stems from the large power density of liquid hydrocarbon fuels, on the order of 42 MJ/kg, that is, two orders of magnitude larger than that of the best (e.g., lithium ion) batteries available on the market today. A successful program in this area would lead to the development of the ultimate distributed power source that may replace batteries in all kinds of applications, ranging from portable electronics to powering micro-air vehicles and other military applications.

Combustion on small scales presents numerous challenges. As size decreases, the combustor surface-to-volume ratio increases. This, in turn, is a rough indicator of the ratio between heat loss and heat release rates. On a sufficiently small scale, losses will be too high for combustion to be sustained, and extinction will occur. Control of quenching problems requires a careful design and a judicious selection of operating conditions and wall materials and ultimately sets a lower limit to the combustor dimensions. The short residence times in a small-scale combustor may prevent complete fuel conversion to CO2 and H2O, reduce heat release, and create emission of pollutants such as CO and unburned hydrocarbons, which suggests the use of catalytic techniques for pollutant abatement and maximization of the combustor efficiency.

Most of the recent and often non-archival literature in this nascent field is reviewed in Ref. [1]. Efforts in this area can be classified into two main categories. A first category involves micro- or mesoscale devices operating on conventional power cycles, for example, gas turbines [2] and internal combustion engines [3–6]. In this approach, problems arise concerning balancing rotary components at high speed and sealing as well as microfabrication difficulties stemming from the intricacy of the geometrical configuration and the necessary tight tolerances. The second category involves a first stage of combustion under steady-state conditions, which could then be coupled to a direct energy conversion module such as a thermoelectric (TEG) [7,8] or thermophotovoltaic (TPV) generator. The advantage of fabrication simplicity of the second approach is often offset by the low efficiency of direct energy conversion (currently not higher than 15% [9–12]). However, if the approach is applicable to liquid hydrocarbons, the advantage of liquid fuels in power density should provide some latitude with respect to the acceptable efficiency of the subsequent energy conversion scheme, which makes this alternative still feasible.

For all these approaches, difficulties in heat management on the small scales arise, which may reduce significantly both the efficiency and the power density of such devices. Such difficulties are particularly severe in microscale applications (order of a few millimeters) but more manageable at the mesoscale (order of a few centimeters).

The present contribution falls in this second category. The emphasis is on the combustion design to
burn liquid fuels at steady state in a chamber with characteristic dimensions on the order of a few centimeters, thereby bypassing quenching issues, arising at submillimeter scales [13] and extracting some of the thermal energy conversion processes with a minimum of moving parts. Even at such a mesoscale, the adaptation of fabrication techniques that have been developed for the solid-state electronics industry may be necessary because design features (contouring, streamlining, coating, multiplexing, etc.) need to be resolved on the submicron scale.

**Burner Design**

**Liquid Dispersion**

Preliminary estimates of the target overall system efficiency (~20%) for the delivery of 20 W of electric power using state-of-the-art thermal-to-electric energy conversion schemes resulted in a combustion power output on the order of 100 W, which translates to a liquid mass flow rate of 7–8 g/hr for the typical heating value of a liquid hydrocarbon. For military applications, the fuel of choice is a jet fuel, such as JP8, because of its availability in the battlefield. Combustion can be implemented after the liquid fuel has been either prevaporized in microchannels or sprayed. A potential difficulty in vaporizing thin films of heavy hydrocarbons is that the fuel decomposes thermally in the presence of hot surfaces, in which case fouling and plugging of the miniaturized channels may ensue. The problem is particularly severe at the location where the phase change occurs and is affected by the residence time [14].

In view of such difficulties, liquid atomization was deemed the alternative of choice, for which virtually the only candidate suitable at the small flow rate of interest is electrospray atomization [15,16]. The electrospray is defined here as a system in which the spray spreading in the radial direction may be affected by space charge effects, namely, the interaction of homopolarly charged sprays that would repel one another.

**The Need for Catalytic Combustion**

TEG and TPV techniques and alkali metal thermal to electric conversion are some of the potential candidates for subsequent combustion interfacing. The specific technique affects combustion only with respect to the temperature that is desirable at the interface with the energy conversion module, which is anticipated to be in the range of 900–1500 K, and its uniformity. In view of the relatively low combustion temperatures, catalytic combustion offers the necessary prerequisite of stability. The feasibility of catalytic oxidation was demonstrated in micromachined reactors in low-temperature silicon-based systems [17].

Particularly well suited for small-scale applications is a recently developed catalyst substrate design (Microlith) [18]. It consists of a number of catalytically coated grids or screens, each with short channel lengths, high cell density, and low thermal mass, stacked serially. The resulting reactor is very compact, has rapid transient response and high energy density, and requires small loadings of precious metal catalysts. Catalyst formulations have been selected from platinum group metals and an alumina washcoat with appropriate additives. The advantages of this design stem from the reduction in mass transfer limitations, compared to conventional monoliths having relatively thick boundary layers. Also, under conditions of kinetic control, the particular design can pack more active area into a small volume, which means that insertion of such catalysts in a flow can provide more effective fuel conversion for a given pressure drop.

**A Case for Multiplexing Liquid Dispersion**

The main drawbacks in burning the entire flow rate from a single source is that it is difficult to achieve the required degree of uniformity of surface temperature for subsequent energy conversion and to maintain small combustor dimensions. Thus, we partitioned the total liquid flow rate into a multitude of small electrosprays. The main advantages of multiplexing are twofold. First, by diminishing the flow rate through each capillary, the generated droplet size is also reduced, since the latter scales with flow rate approximately as \( \frac{1}{2} \) [19]. For example, if the total flow rate dictated by energy requirements is partitioned into \( n \) electrosprays, the droplet size at the injection is reduced by \( (1/n)^{2/3} \) and since, in the first approximation, the droplet evaporation time varies with the square of the droplet diameter, the evaporation time should be reduced by \( (1/n)^{4/3} \). Second, establishing a shroud flow of oxidizer around each spray ensures that the mixing occurs rapidly and homogeneously. A potential disadvantage of multiplexing is that the spray spreading in the radial direction may be affected by space charge effects, namely, the interaction of homopolarly charged sprays that would repel one another.
likely to result in larger combustor volumes. Conversely, one could ignite farther downstream after substantial mixing with the oxidizer has occurred, using a gauze or honeycomb to separate the combustion region from the electrospray and to prevent flashback. This premixed combustion alternative was explored decades ago in some pioneering work by Thong and Weinberg [20]. Here, we consider the catalytic alternative, in which the Microlith acts both as catalyst and as stabilizing grid for the flames.

### Experimental Apparatus

A photograph of the experimental burner is shown in Fig. 1. The fuel was metered with a syringe pump. The burner was operated with three fuels, all of them with 0.3% per mass of an antistatic additive (Stadis 450, Du Pont). The fuels were JP8 jet fuel; n-dodecane, which is the single-component fuel, the physical properties of which are closest to JP8 [18]; and 1,3-di-isopropylbenzene, which has a structure and properties similar to the most common aromatics in JP8 [21,22]. For the results to be reported here, the flow was distributed through a polyether ether ketone (PEEK) manifold into seven stainless-steel capillaries (1.59 mm o.d., 127 µm i.d., 10 cm length) arranged in a hexagonal pattern. The capillaries were mounted through a Teflon flange, and their tips were sharpened and polished to eliminate burs that would affect the electric field pattern. In the future, more extensive multiplexing, on the order of 100 electrosprays in parallel, is planned by microfabrication techniques that have already been applied to the electrospray for chemical analytical applications [23]. The flange supported a cylindrical pyrex chamber 38.1 mm i.d. and 38.1 mm in height that provided optical access. Air was provided through the flange into the chamber at a temperature of 200 °C to simulate recuperation in a real device. The chamber was capped at the top with a metal holder in which the Microlith catalyst screens were housed. Each screen was an approximately orthogonal grid of 125 µm wires with 1 mm pitch. Once the capillaries were charged to a voltage on the order of several kilovolts relative to the catalyst holder that was grounded, seven electrosprays were simultaneously established at the capillary tips. The pressure loss across the capillaries is minimal, that is, approximately 20 cm of water (~2 kPa), and was measured by supplying the liquid gravimetrically. The pressure drop across the catalyst grid is so small that it cannot be measured accurately (~< 1 kPa). The power consumption of the electrosprays is minimal. The current carried by the sprays was measured to be on the order of 100 nA, which yields a parasitic loss of less than 1 mW for approximately 100 W of thermal power.

The temperature on the catalyst surface was measured using a PV-320 Electrophysics infrared video camera and a germanium objective lens. To avoid signal saturation, a glass filter of 0.3 OD and a 2.5 µm cutoff was used, so that the recorded signal on the camera chip was from the 1.5–2.5 µm part of the spectrum. The temperature measurements were calibrated using a K-type thermocouple coated with the catalyst material. Exhaust gas sampling downstream of the catalyst was used to determine exhaust gas composition. Major combustion products and light species (N₂, O₂, H₂, CO₂, CO, CH₄, C₂H₆, and in general hydrocarbons with three or less atoms of carbon) were measured using a two-channel microgas chromatograph from Agilent Technologies. The device employs two channels, one with a molsieve and one with a PoraPLOT backflush injector. Acquisition of an exhaust gas sample lasted 60 s, and argon was used as a carrier gas for both injectors.

### Results and Discussion

The objective of the fuel delivery system is to produce a lean, uniform mixture of fuel and air upstream of the catalyst. The mixture should burn completely as it passes through the screens and provides a uniform surface temperature in the range 900–1500 K, for subsequent coupling with direct energy
conversion modules. The mechanism of electrospray atomization, which determines the quality of air/fuel mixing, was studied in detail for isolated electrosprays in Ref. [19]. Because of space charge effects, the results of such studies are only of qualitative validity when multiplexed sprays are considered. The appearance of these sprays for JP8 injection is shown in Fig. 2, which was taken without the Pyrex chimney and in the absence of combustion. Similar photographs during actual operation are less informative due to reflections on the Pyrex tube but show a similar structure. A cone of liquid fuel was formed on each capillary tip from which a short fluid ligament protruded. Atomization was achieved through the breakup of these ligaments similar to Ref. [19], and the Coulombic repulsion among droplets of different sprays facilitated fuel dispersion. The length of the plumes in Fig. 2 was on the order of 5 mm and is expected to be significantly smaller during actual operation because of heat diffusion from the reaction region.

The degree to which uniformity of equivalence ratio is achieved by the time the mixture reaches the catalyst determines the quality of the performance of the device. To characterize such a performance, the surface temperature of the screens and the exhaust gas composition were measured and their uniformity was investigated. From the exhaust gas composition, the combustion efficiency can be determined.

A two-dimensional map of temperature on the catalytic screen acquired with infrared photography is presented in Fig. 3 for JP8 fuel, at an equivalence ratio $\phi = 0.48$ and a fuel flow rate of 9.8 g/hr. The dark holes in the pattern are void regions through which the exhaust gases flow. Radial temperature scans on the catalytic screen with data acquired from the thermography pictures are presented in Fig. 4a for $\phi = 0.48$ and a fuel flow rate of 12.6 ml/hr for three different fuels: JP8, $n$-dodecane, and 1,3-diisopropylbenzene. There is a $\pm 5\%$ variation around the mean value and a mild temperature gradient across the screen for the two single-component fuels. Better uniformity of the equivalence ratio is achieved with JP8, possibly because of the presence of lighter components which vaporize faster and mix more effectively than the heavier fuels. A maximum temperature of approximately 1100 K is achieved which is within the range of target temperatures for direct energy conversion. This temperature is significantly lower than the adiabatic flame temperature of, for example, dodecane, for the same equivalence ratio, that is, 1550 K. An energy balance in a control volume surrounding the screens yields

$$\frac{dm}{dt} Q_{LHV} = \frac{dm}{dt} c_p (T - T_o) + \varepsilon \sigma A (T^4 - T_o^4) + Q_{\text{cond}}$$

where $m$ and $m_f$ are total and fuel mass, respectively; $Q_{LHV}$ is the lower heating value of the fuel; $\varepsilon$ is the emissivity of the catalyst material which was approximated as the one of the steel material of the wires; $A$ is the radiating surface which can be estimated from the dimensions of the Microlith; and $Q_{\text{cond}}$ is the heat loss due to conduction through the catalyst material and the rest of the burner hardware. $Q_{\text{cond}}$ is difficult to measure directly but substitution into equation 1 yields that it must be on the order of 15% of the total energy input, which is entirely conceivable for the current experimental setup. Selection of appropriate insulating materials or heat management schemes should reduce this loss and make the same temperatures of operation possible for even leaner mixtures.

The maximum temperature as a function of equivalence ratio for a given fuel mass flow rate is shown in Fig. 4b for JP8 combustion. The equivalence ratio was varied by changing the air flow. Since the overall stoichiometry is always lean, the temperature should
increase with equivalence ratio. Yet, an increase of the equivalence ratio from 0.40 to 0.53 results in an increase in the temperature on the catalyst by only 100 K, whereas the calculated adiabatic flame temperature increases by approximately 500 K. The temperature on the catalyst appears to decrease for $\phi > 0.53$. This is likely due to the change in residence times within the catalyst.

The fact that the operation of this experimental burner is heat loss limited is shown more clearly by the dependence of maximum temperature on fuel flow for fixed $\phi = 0.48$, as shown in Fig. 4c. If the equivalence ratio is kept constant, the adiabatic flame temperature is expected to be constant. This is not the case for non-adiabatic operation. The energy input to the system (left-hand side of equation 1) is proportional to the fuel flow rate, which in turn is proportional to total flow rate, since $\phi$ is fixed. The heat loss (two last terms on the right-hand side) is mainly determined by the geometric and heat transfer properties of the system, which do not correlate with the flow rate equally strongly. As a result, a decrease in flow rate results in a lower temperature on the catalyst. There is actually a threshold fuel flow rate ($\approx 6 \text{ g/hr}$) below which operation can not be sustained. On the other hand, if the flow rate is increased above a limit ($\approx 25 \text{ g/hr}$), the residence time in the burner is not sufficient for the fuel to react and the mode of operation changes completely with the establishment of a partially premixed flame in the gaseous phase downstream of the catalyst screens.

Even though operation of the burner with a commercially available fuel blend has been established, it is preferable to investigate the composition of the exhaust gases for a single-component fuel, for which the stoichiometry can be calculated precisely and the combustion efficiency can be calculated more accurately. For a base case of operation with n-dodecane with $\phi = 0.48$ and a fuel flow rate of 9.7 g/hr, gas chromatographic measurements of mole fractions of the main components, N₂, O₂, CO₂, and CO, in a dry sample from the exhaust gas yielded 81.5, 11.5, 6.4, and 0.11%, respectively. Light hydrocarbons like CH₄ and C₂H₆ as well as H₂ were below the detection threshold of the particular gas chromatography apparatus (50 ppm). The corresponding composition of the exhaust gas mixture for complete combustion can be easily calculated as 81.9% N₂, 11.3% O₂, and 6.8% CO₂. The proximity of the exhaust gas composition to the one for complete combustion and the fact that no light hydrocarbons were detected indicate that combustion is very efficient. We can estimate this efficiency by assuming that all of the 0.4% deficit in the measured molar balance consists of combustible material and must be counted as a loss. To this loss we add the percentage of carbon which turns to CO which is estimated by the CO/CO₂ ratio. This way, an efficiency of 97% is estimated.

The ratio of CO₂/CO can thus be used as a measure of combustion efficiency and is presented in Fig. 5 as a function of equivalence ratio for a mass flow rate of fuel equal to 9.7 g/hr. In the same figure, the mole fraction of CO in a dry sample from the exhaust gases is presented. Maximum CO to CO₂ conversion is achieved for $\phi \approx 0.47$, close to $\phi \approx 0.53$ for which maximum temperature is achieved on the catalyst screen. The discrepancy is probably due to the fact that the CO₂/CO ratio correlates but is not exactly equal to the combustion efficiency. More detailed analysis of the exhaust gas composition is expected to
yield a value of $\phi$ for maximum efficiency even closer to the corresponding one for maximum temperature.

Figure 6 shows the CO$_2$/CO molar ratio and the mole fraction of CO in a dry sample from the exhaust gases as a function of the fuel mass flow rate for an equivalence ratio $\phi = 0.48$. Since the equivalence ratio is fixed, fuel flow rate is proportional to total flow rate and inversely proportional to residence time in the burner. As residence time decreases, it is clear that the CO to CO$_2$ conversion is kinetically more limited and less effective. The corresponding slight increase in temperature shown in Fig. 4c is apparently not sufficient to counterbalance the effect of reduced residence time. The relatively large ratios of CO$_2$/CO in Figs. 5 and 6 suggest that the majority of the fuel conversion occurs on the catalyst. More extensive analysis of the exhaust gas composition may reveal the presence and quantities of pollutants other than CO in the exhaust gases, such as unburned hydrocarbons. An accurate calculation of the combustion efficiency involves more detailed measurements of the unburned hydrocarbons in the exhaust so that a carbon balance can be determined. Preliminary results from microprobe gas sampling and online photoionization mass spectrometry seem to confirm the above estimate of combustion efficiency. A study focused on fuel conversion chemistry and on the pollutant emission is currently in progress.

Summary and Conclusions

A clean and efficient mesoscale combustor for power generation from liquid hydrocarbons has been developed. Difficulties associated with the combustion of even kerosene-type fuels, such as JP8, were circumvented with a novel combination of fuel electrospraying and catalytic combustion. Dispersion of the liquid fuel and subsequent mixing and vaporization were achieved in a small volume by electrospraying, which required minimal power and no elevated pressures. No significant amount of surface fouling, that is typically observed when the phase change of JP8 takes place near solid surfaces, was observed. Neither soot nor gaseous pollutants, except for a small amount of unburned hydrocarbons, were detected and almost complete conversion of CO to CO$_2$ was achieved, despite the low combustion temperatures and the relatively short residence times. The output of the burner is a uniformly hot surface at a temperature appropriate for coupling with direct energy conversion modules.

Specifically, testing was performed at a fuel flow rate on the order of 10 g/hr and an equivalence ratio ranging from 0.35 to 0.70. Depending on equivalence ratio and mass flow rate, the temperature on the catalyst surface varied in the range 950–1200 K, with a $\pm 5\%$ uniformity over the surface. Combustion efficiencies on the order of 97% were achieved for operation with n-dodecane, a hydrocarbon with physical properties similar to those of JP8.

Acknowledgments

We acknowledge Mr. R. W. Dean of PCI for his assistance with the micro-GC measurements and Mr. N. Bernardo of Yale University for machining the hardware. The support of DARPA, under grant DAAD19-01-1-0664 (Dr. Richard J. Paur, Contract Monitor) is gratefully acknowledged. Microlith is a trademark of Precision Combustion, Inc.

REFERENCES

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charged droplets. Further miniaturization is being pursued by multiplying the electrospray even further, using micro-

fabrication techniques. As a result, smaller droplets will be issued from each source, with more closely spaced sources. We anticipate smaller penetration in the streamwise direction, whereas penetration in the radial direction that is controlled by Coulombic repulsion will actually be favored. All these effects should favor more uniform mixing within shorter mesoscale combustors. However, modification to the air delivery system may be required to ensure a low momentum delivery of the necessary airflow rate.

Author’s Reply. Several phenomena observed in this mesoscale combustor differ significantly from the corresponding processes in the large scale and evidence intriguing challenges for further miniaturization. For example, the dependence of flame temperature on the fuel mass flow rate for a constant equivalence ratio and the appearance of a maximum temperature at a very lean equivalence ratio for a given fuel mass flow rate are surprising and a consequence of the large surface-to-volume ratio and short res-

idence time of this combustor, as compared to large-scale burners. The operation of Microlith catalysts in the mesoscale is also particular. Typically, such catalysts provide increased surface area for the same flow blockage and for strikingly shorter channel lengths compared to the classical monoliths to facilitate heterogeneous chemistry. For mesoscale operation, the Re and Pr numbers for the flow of the combustion products around the catalytic meshes are such that the boundary layers virtually block the flow. In this way the role of homogeneous chemistry is also boosted, which leads to the observed high-conversion efficiencies, despite the low blockage and few meshes used in these experiments. There is the behavior of multiplexed electro sprays, and their effect on mixing and vaporization in a small volume is quite distinct from that of conventional atomizers, as partially discussed in the reply to Professor Sirignano.

Gregory J. Fiechtner, Sandia National Laboratories, USA. Congratulations on your excellent results and presentation. There are three questions about your demonstration of electrospray: You emphasize a stream of monodisperse drops. For producing ions for a mass

COMMENTS

William A. Sirignano, University of California, Irvine, USA. You suggested that the technique for atomization could produce droplets of a few microns in diameter. However, have you quantified the required electrostatic force and energy to have the very small droplets penetrate the moving air stream?

Author’s Reply. For the burner described here, the electric field energy transferred to the droplets is of the order of 1 mJ, since the drops carry a current on the order of 100 nA across a potential difference on the order of 10 kV. This corresponds to less than 0.1% of the total thermal energy release and proves sufficient for homogeneous dispersion of the fuel even when the air was delivered through side ports. Under the present conditions, the momentum of the droplets is such that their motion is not disturbed by the surrounding air flow. Moreover, dispersion is facilitated by the Coulombic repulsion between homopolarly charged droplets. Further miniaturization is being pursued by multiplying the electrospray even further, using micro-

fabrication techniques. As a result, smaller droplets will be issued from each source, with more closely spaced sources. We anticipate smaller penetration in the streamwise direction, whereas penetration in the radial direction that is controlled by Coulombic repulsion will actually be favored. All these effects should favor more uniform mixing within shorter mesoscale combustors. However, modification to the air delivery system may be required to ensure a low momentum delivery of the necessary airflow rate.

J. P. Gore, Purdue University, USA. What new meso-

scale phenomena have you observed in your combustor to emphasize the adjective?
spectrometer, this is critical, but is it necessary for a combustor? That is, perhaps you could use other modes of electrospraying and not just the corr-jet mode.

There is evidence that oxygen dissolved in liquid fuel will react with the electrospray nozzle material under an applied electric field. Will the lifetime of the device be limited? For nozzle arrays micromachined with small aspect ratios, this could be particularly troublesome. Then, what is the minimum distance required for droplet formation and fission as related to the total size of the combustor?

Author’s Reply. Thank you for your remarks. In response to your insightful questions: Narrow droplet size distributions are important for the operation of miniaturized burners because they lead to a narrow range of vaporization times and therefore to a well-defined length over which full vaporization of the liquid fuel and mixing with the oxidizer are achieved. However, operation in the “cone-jet” mode is not a “must.” On the other hand, since this mode is well defined and a large literature exists on its characterization, it is an obligatory starting point to explore principles of operation of this novel combustor.

For the time scales of the experiments reported here (<4 hours of continuous operation), no obvious nozzle damage was observed. Possibly, because of the short residence times of the liquid in the conical meniscus at the relevant flow rates, dissolution is not significant, at least at atmospheric pressure. Lastly, even if the liquid fuel distributor lifetime were an issue, the use of microfabrication techniques, as discussed in the oral presentation, should reduce fabrication costs to the point of making disposable distributors economically feasible.

We have not yet implemented miniaturization much beyond the level discussed in the paper, for which the total combustor length is governed mostly by vaporization and mixing constraints of fuel and oxidizer, rather than by liquid break-up processes that typically occur over a millimeter length scale.