

# “Hissing” Electrospray and Combustion at the Mesoscale

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## Abstract

An application of the electrospray to mesoscale combustion is studied, with the ultimate goal of coupling the combustor with direct energy conversion modules for power production. The combustor design relies on fuel dispersion by multiplexed electrosprays coupled with a series of catalytic meshes acting as ground electrode and initiators of the fuel oxidation. The combustor has a volume on the order of few cm<sup>3</sup> and operates on JP-8 jet fuel, which is electrosprayed at a flow rate on the order of 10 g/hr. The behavior of the electrospray in high temperature environments is examined and the phenomenology of the electrospray in the combustion chamber is visualized using planar laser-induced fluorescence from a fluorescent tag doped into the fuel. The electrospray is operated in an unsteady mode accompanied by a “hissing” sound, characteristic of the onset of corona discharge, which lead to optimal mixing of the fuel and the oxidizer upstream of the catalyst. Under such conditions, combustion efficiencies on the order of 99% are achieved with uniform (within  $\pm 5\%$ ) temperatures at the catalyst in the range of 900K-1300K. Remarkably, no fouling, soot, or NO<sub>x</sub> were detected in the exhaust gases, which resulted in clean and efficient combustion of even environmentally problematic liquid hydrocarbons.

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## **1. Introduction**

An electrostatic spray (ES) offers several advantages compared to alternative atomization techniques: monodispersity of the generated droplets, spray self-dispersion, absence of droplet coalescence, and minimal clogging risks of the atomizer, the inner dimension of which is orders of magnitude larger than the generated droplets. It can be established by charging a conducting liquid emerging from the tip of a capillary tube to a sufficiently high potential with respect to a ground electrode a short distance away, so that the liquid meniscus takes the shape of a cone from the tip of which a thin liquid thread emerges (cone-jet mode). This microjet breaks into a stream of charged droplets, which eventually spread to form a spray, more properly, an electrospray. Cloupeau and Prunet-Foch [1] give a comprehensive account of the phenomenology of various modes of operation, including the cone-jet mode.

The electrospray has initiated a veritable revolution in the area of mass spectrometry, spearheaded by the pioneering work of John B. Fenn at Yale in the 80's [2], culminating in his 2002 Nobel Prize in Chemistry. Applications to other areas have been hampered by the low flow rates at which the spray can be established. Although microfabrication techniques may circumvent this problem in the future, enabling large scale multiplexing, presently commercial applications are pursued in areas well suited for low flow rates, such as chemical analysis or drug inhalation.

## **2. Mesoscale combustion application**

Specific to combustion, the feasibility of ES burning in a counterflow premixed flame was demonstrated in the pioneering work in [3]. The behavior of electrospray in diffusion flames was extensively studied in our group in both counterflow and coflow configurations [4-6]. In this experimental work the electrospray was operated at flow rates on the order of 0.5 ml/min. Such a flow rate has been traditionally regarded as large enough for laminar laboratory-scale experiments, but too small for any technological application, relegating the electrospray to the status of intellectual curiosity, useful only for fundamental work. This state of affairs changed recently, when combustion became attractive for small-scale either electric or propulsive power generation, because of the large power density (power/volume) offered by liquid fuels, up to two orders of magnitude larger than the best batteries available on the market today. The power requirements of all these applications are on the order of tens or hundreds of Watts, with the overall dimension of the device of, at most, a few cm.

In [7, 8] we discussed in detail the unique advantages an approach based on the combination of electrospray liquid fuel atomization and catalytic combustion provides for power generation in the meso- and, potentially, microscale. Liquid fuel combustion can be implemented after the liquid fuel has been either pre-vaporized or sprayed. However, one potential difficulty in pre-vaporizing some of the heavy fuels of interest is that the fuel may decompose thermally in the presence of hot surfaces, in which case carbon-based deposition, fouling and obstruction of the microchannels may ensue. The alternative to pre-evaporation is dispersing the fuel using an electrospray technique, probably the only viable approach to burn directly minute amounts of liquid hydrocarbons. Since the size of the generated droplets and, consequently, the droplet evaporation time are monotonically decreasing functions of the liquid flow rate, it is best to multiplex the electrospray and use a system of several capillaries mounted in parallel. This approach would also help in achieving a

reasonably uniform temperature in the combustor cross-section, which is important for subsequent thermal-to-electrical energy conversion.

The likelihood of short residence times in the combustor may create significant emissions of CO, and probably, unburned hydrocarbons, in which case catalytic combustion strategies would be particularly promising. A recently developed catalyst substrate design (Microlith<sup>®</sup>) [9] is particularly well suited for small-scale applications. It consists of a number of catalytically coated grids or screens stacked serially, each with short channel lengths, high cell density, and low thermal mass. The resulting reactor is very compact, has rapid transient response and high energy density, and requires small loadings of precious metal catalysts.

With these considerations in mind, we designed a JP-8 mesoscale combustor consisting of a system of multiplexed electrosprays for the liquid fuel dispersion and a series of catalytic Microliths a few centimeters downstream of the spray sources acting as the ground electrode. The prototypical combustor had a volume on the order of few cm<sup>3</sup> and JP-8 was electrosprayed at a flow rate on the order of 10 g/hr, corresponding to approximately 100 W of thermal power, and equivalence ratios varying from 0.35-0.70. Temperatures in the 900K-1300K range were achieved with a  $\pm 5\%$  uniformity over the top circular surface of the burner, which is compatible with the input requirements of current direct energy conversion technologies (e.g., thermo-electric generation, TEG, or thermo-photovoltaics, TPV). Combustion efficiency in excess of 99% was estimated and, remarkably, no fouling, nor soot, nor NO<sub>x</sub> were detected in the exhaust gases, which resulted in clean and efficient combustion of even the notoriously polluting JP8 fuel. Durability tests demonstrated the robustness of the approach over 500 hrs of continuous operation [8]. In the remainder of this article, we focus on the behavior of the electrospray in these high temperature systems, as revealed by laser diagnostics.

### 3. Experimental system

A schematic of the experimental burner is shown in Fig.1. The fuel, JP-8 doped with 0.05% per mass of an antistatic additive (DuPont, Stadis 450) to enhance its electric conductivity, was metered with a syringe pump. The flow was distributed through a poly-ether-ether-ketone (PEEK) manifold into 19 stainless steel capillaries (1.59 mm O.D., 127  $\mu$ m I.D., 10 cm length) arranged in a hexagonal pattern with two concentric hexagons surrounding the central capillary. The capillaries were mounted through a ceramic flange and their tips were

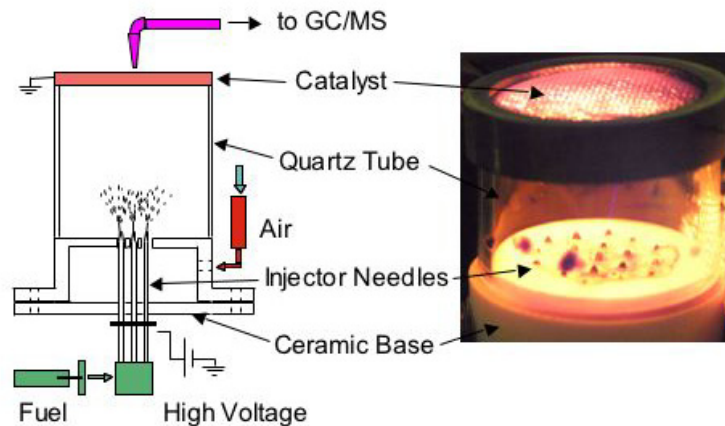


Fig. 1: Schematic and picture of the prototype burner operating with electrosprayed JP-8 fuel

sharpened and polished to eliminate burs that would affect the electric field pattern. The flange supported a cylindrical Pyrex chamber 38.1 mm in I.D. and 38.1 mm in height that provided optical access. Air was provided through the flange into the chamber at a flange temperature of 473 K to simulate recuperation in a real device. The chamber was capped at the top with a metal holder in which three Microlith<sup>®</sup> catalyst screens were housed. Each screen was an approximately orthogonal grid of 125  $\mu\text{m}$  wires with 1 mm pitch. Once the capillaries were charged to a voltage on the order of several kV relative to the grounded catalyst, several electrosprays were simultaneously established at the capillary tips. The pressure loss across the capillaries was approximately 20 cm of water ( $\sim 2$  kPa) whereas the corresponding pressure drop across the catalyst grid was so small that it could not be measured accurately ( $< 1$  kPa). The current carried by the sprays was measured to be on the order of 100 nA, which yielded a parasitic loss of less than 1 mW for approximately 100 W of thermal power. Fuel atomization, dispersion and vaporization as well as mixing with the coflowing air stream occurred in the cylindrical chamber while combustion was established at or near the catalytic grids. The burner used for diagnostics was slightly modified in that a square geometry, consisting of four fused silica windows pushed together by a Teflon base, is used. This modification allowed for easier optical access without the reflections and distortion that the cylindrical geometry would introduce, and for easier cleaning of the windows.

To visualize the fuel distribution in the operating device, planar laser-induced fluorescence (PLIF) of a fuel tracer was used. Fuel PLIF has been used in sprays and combustion devices both qualitatively and quantitatively [10]. In LIF experiments, a molecular transition is excited by the laser. The molecule relaxes to the ground state via fluorescence or quenching by collision with other molecules. In PLIF a laser sheet from a pulsed laser at a suitable wavelength provides excitation, and a camera images the fluorescence. Because the laser pulse and fluorescence decay times are typically shorter than the flow time scales and the signal is relatively strong, instantaneous measurements with a single shot are possible.

As a tracer, 5% of 1-methylnaphthalene is added to the fuel surrogate (n-dodecane). The boiling points of tracer and fuel ( $240^\circ\text{C}$  and  $216^\circ\text{C}$ , respectively) are sufficiently close for the tracer to follow the fuel in its evaporation behavior. In addition, naphthalenes typically make up about 5% of the target fuel JP-8. The fluorescence decay time for this tracer is on the order of 100 ns. For a chemically similar tracer, toluene, it has been shown that, since oxygen is practically the only effective quencher, the fluorescence signal is essentially proportional to the local equivalence ratio (or fuel/air ratio) [11]. Although we have not verified that this is also the case for 1-methylnaphthalene under the conditions in the evaporation chamber, for qualitative imaging we will assume it to be true. A quantitative investigation of this and the temperature dependence of the fluorescence, which is also not addressed here, is currently under way. The experimental arrangement is shown in Fig. 2. Excitation is provided by the fourth harmonic of an Nd:YAG laser (Continuum Powerlite) with a wavelength of 266 nm, a pulse duration of  $\sim 10$  ns, and a repetition rate of 10 Hz. A combination of spherical and cylindrical lenses forms a vertical sheet of suitable dimensions ( $\sim 18.5$  mm high and 0.5 mm thick) in the evaporation chamber. The burner is mounted on a motorized computer-controlled translation stage such that the entire volume of the chamber can be scanned. Fluorescence occurred broadband in the 330-380 nm range and was detected by an image-intensified CCD camera (Photometrics 512 x 512 pixels lens-coupled to 19 mm DEP MCP intensifier). Scattered laser radiation and longer-wavelength fluorescence of material accumulating on the windows was suppressed by a band pass

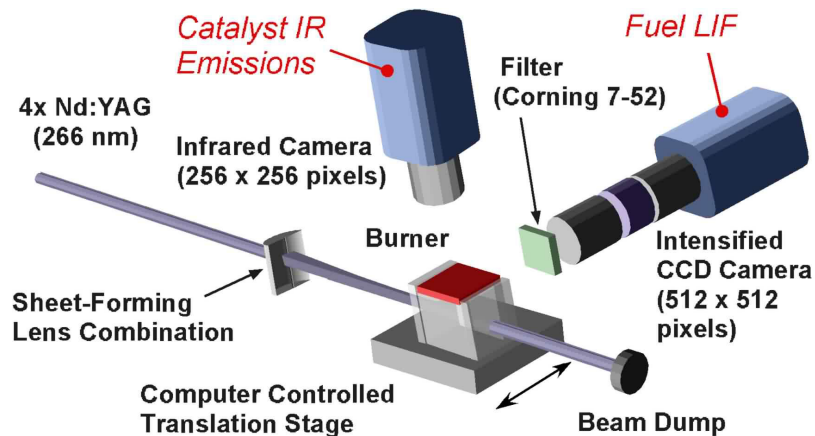


Fig. 2: Experimental configuration for fuel-tracer PLIF

colored glass filter (Corning 7-52); the short gating time of the intensifier (200 ns) avoided interference from the catalyst luminescence. Also shown in the schematic is an infrared-sensitive camera that was used to image the catalyst temperature.

#### 4. Results and discussion

The mechanism of electrospray atomization of liquid hydrocarbons was studied in detail for isolated electrosprays [12]. Multiplexing, depending on the compactness of the configuration, may lead to a somewhat different behavior. At a minimum, one would expect that because of space-charge effects each individual spray angle would be smaller as compared to the isolated case. The appearance of these sprays for JP-8 injection is shown in the photograph of Fig. 3, which was taken in room-temperature air so that the relevant structure would be clearer. Also in the multiplexed case, the characteristic cone-jet structure is clearly visible.

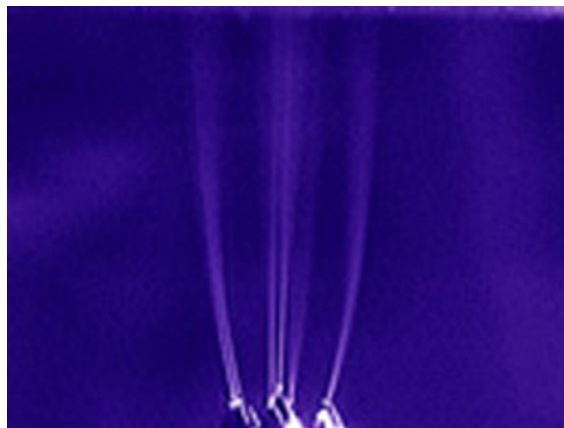


Fig. 3: Atomization pattern for electrospraying of JP-8 into ambient air conditions

No such photograph is available at the high operating temperatures of the combustor. However, we can postulate the effects of elevated temperatures on the electrospray behavior. If the fundamental mechanism of electrospray generation is envisioned as a balance between Coulombic force on the liquid and surface tension, the elevated temperatures associated with combustion applications are expected to have two major effects on the physical properties that are relevant to the ES. First, the surface tension will decrease, and so will the voltage necessary for the establishment of the cone-jet mode; second, the fluid electric conductivity will increase with temperature, as a result of a decrease in liquid viscosity and the consequent enhanced ion mobility, which may affect the stability domain of a given electrospray regime [12]. Furthermore, since the fluid patterns associated with the electrospray usually consist of high surface-to-volume ratio morphologies (cones, elongated cylindrical ligaments), one would expect enhanced evaporation of the fluid in hot environments, which may also affect the ES stability.

PLIF is ideally suited to probe the relevant phenomenology in situ, because in the confinement of a small combustor its signal can be discriminated from the large scattering interference at the same wavelength of the probing laser beam. Figure 4 shows a series of time-averaged (row i, top) and single-shot (row ii, bottom) images taken at a location in the evaporation chamber such that the laser sheet intersected the axis of two of the 19 sprays. In each series, images from operation at varying electric potential between needles and catalyst are shown. Without voltage, the capillaries operate as evaporators, which is sustainable because of the intake air heating and the back radiation from the catalyst grid. The corresponding single-shot image is essentially identical (apart from intensity and noise) to the time-averaged one, except for the small fluctuations caused by the stepper motor of the syringe pump (Fig. 4, column a). The partial recirculation of fuel vapor downwards along the left and right window, caused by the fact that not the entire chamber bottom has intake holes, can also be seen. Temperature measurements for these conditions with the infrared camera indicate that the temperature distribution on the catalyst is very inhomogeneous with a “spotty” pattern of hot reaction regions surrounded by cold, non-operating parts of the catalyst. Apparently, liquid evaporation and diffusion in the gaseous phase is not an effective mechanism for fuel dispersion and mixing. This mode of operation is also very unfavorable because coking develops quickly.

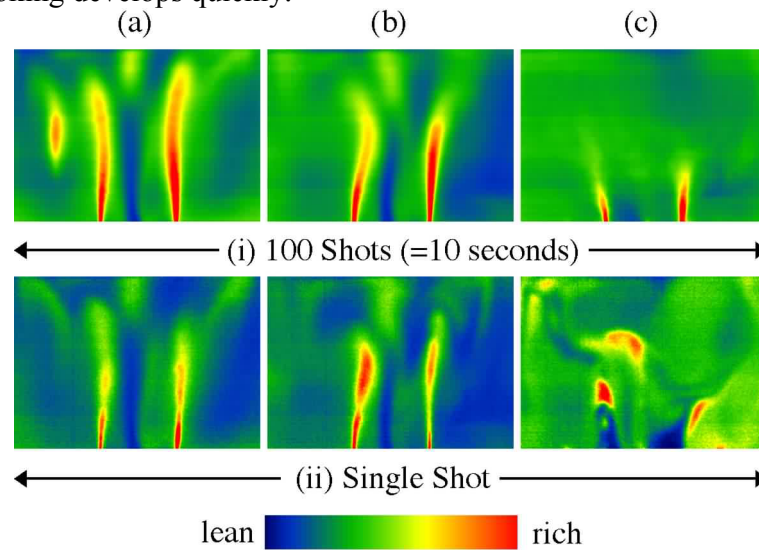


Fig. 4: PLIF images of the operating combustor. See text for details.



When the capillaries are charged at 6 kV, (Fig. 4, column b) most of the sprays operate in the stable cone-jet mode. Again, single-shot and time-averaged images look similar. The recirculation pattern cannot be seen as clearly anymore. Catalyst uniformity is improved because of the liquid dispersion due to the Coulombic force. When the voltage is increased further, the cone-jet mode becomes unstable for most sprays and is accompanied by a characteristic “hissing” sound. Measurements under cold conditions under the same “hissing” conditions showed a dramatic increase of the ES current, a prelude to the eventual corona discharge at even higher voltage and the complete disruption of atomization. It is now particularly interesting to compare single shot and average images (Fig. 4, column c). The time-averaged image shows a more homogenous mixture throughout the field and a shorter zone of nearly unmixed fuel, whereas the single-shot image reveals a completely unsteady structure comprised of fuel-rich pockets that are not necessarily where the richest mixture is found on average. PLIF does not provide any velocity information, but in view of mushroom-shaped fuel-rich eddies that can be observed on some of the single-shot images (not shown here) it is suggested that a highly unsteady flow field is established throughout the chamber, contrary to the intuitive image of a smooth laminar flow pattern. The source of unsteadiness is attributed to the irregular emission of droplets from the capillaries. Clearly, mixing is enhanced strongly in this unsteady mode of operation. The catalyst temperature and visual appearance are very uniform. This newly identified “hissing” mode seems to be preferable for the operation of the particular burner.

Since time-averaged images, even in the unsteady, “hissing sound” mode, are relatively stable, it is possible to reconstruct the information throughout the entire chamber by scanning in the third dimension, perpendicular to the catalyst grid, and combining the vertical slices obtained. This way a complete image of the time-averaged, three-dimensional mixing process can be formed. Images are taken in steps of 0.5 mm, to a distance of about 0.5 mm from each wall, giving a total of 58 slices. Figures 5a and 5b each show a series of horizontal slices through the 3D array for the case of 0 and 10 kV applied voltage, respectively. The lowest slice of both series is similar: the very fuel-rich jets (the false color scale has been saturated significantly here to be able to resolve finer shades in the higher

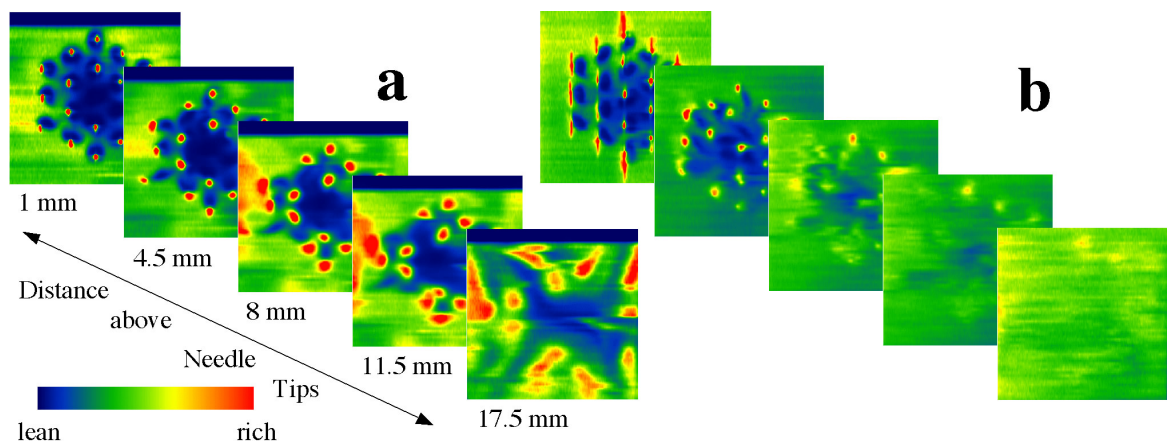


Fig. 5a and b: Mixing patterns in cross-sections parallel to the catalyst for 0 and 10kV, respectively

slices on the same scale) are surrounded by pure air from the intake holes. In this image series, it can be seen very clearly how much mixing is increased by the unsteady spray mode as compared to molecular diffusion in the laminar flow. The uniform mixing upstream of the catalyst suggested by Fig. 5b is in qualitative agreement with the uniformity of temperature of the catalyst, as evidenced by infrared thermography.

## 5. Conclusions

The electrospray application to mesoscale combustion was demonstrated to provide a unique means of dispersing the fuel and mixing it with the oxidizer in small volumes, for subsequent catalytic oxidation. High combustion efficiencies and uniform combustion temperatures were achieved when the electrospray was operated in an unstable mode, accompanied by a hissing sound characteristic of the onset of corona discharge. Laser-induced fluorescence proved to be a very useful tool to image the spray phenomenology in confined environments.

## 6. References

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