

# DEMONSTRATION OF AN ELECTROSPRAY INJECTION SYSTEM

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**Abstract:** The use of liquid hydrocarbons as a chemical energy source is promising for the actualization of high energy density power sources in the near future. Electrospray injection method is a unique technique that provides equal droplet size distribution at very low flow rates. Therefore it is quite suited for the atomization of liquid hydrocarbon fuels in miniature energy conversion devices. This study reports the design and characterization of an electrospray system. First, the electrospray phenomenon is briefly discussed from an historical perspective. An experimental test rig is built and a proof-of-concept demonstration is provided. For the experiments methanol is used as the liquid fuel. Flow visualization is performed to identify the electrospray mode. Results suggest that a Taylor cone is cone formed when voltage is applied to the system. After a certain voltage threshold electrostatic forces overcome the surface tension forces and droplets begin to separate from the Taylor cone. For voltage values higher than 6 kV certain instabilities are observed. Starting voltage for the current configuration is measured to be 2.2 kV. This value is in close agreement the theoretical calculations which suggest that the starting voltage would be 2.5 kV. This 12% discrepancy can be attributed to experimental uncertainty. Flow rate from a single injector is found to be on the order of 2 ml/h. Therefore in order to utilize electrospray injection in practical power conversion devices manufacturing of high nozzle density multiplexed emitter arrays are needed.

**Key words:** Electrosprey, injection system

## Introduction

Liquid hydrocarbons have a much higher energy density than conventional batteries. For example; a typical liquid hydrocarbon has an energy density around 42 MJ/kg, on the other hand a lithium battery can only store about 0.6 MJ/kg (Deng et al. 2007). For this very reason, batteries that work with liquid hydrocarbons can make a technological breakthrough in this field. Even if, these systems have low energy conversion efficiency (i.e. thermal to electric), they can produce much more electrical energy per unit battery weight. Thus, these devices would be quite suitable for military and aerospace applications due to their light weight and size.

In micro scale systems, fuel must be in liquid phase, to get the desirable energy density. Gaseous fuel is simply not an option. Liquid fuel must be atomized, be injected into the combustion chamber and mixed with air. Electrospray injection is a suitable method for liquid injection within micro scale devices.

Electrospray injection is a unique technique suited for very low flow rates, which can assure a uniform spray and droplet size distribution. This technique relies on the ionization of conductive liquids under strong electrostatic fields. Meniscus occurs on the conductive liquid's surface whenever the liquid is affected by the presence of an electric field. This in turn causes electrostatic pressure on the free surface of the liquid. The surface tension of the liquid manages to neutralize the effect of electrostatic pressure. For weak electric fields, meniscus effect on the liquid surface is not much pronounced, however when stronger fields are applied, the liquid surface assumes a conical form. Finally, when a sufficiently strong electric field is applied to the system, electrostatic forces overcome surface tension forces, as a consequence separation from tip of the cone occurs. This phenomenon is termed as electrospray ionization (Krpoun, 2009).

Electrospray can be described with two different phenomena. The first one is a phenomenon where an electric field is used to charge pneumatically or mechanically sprayed droplets. The second one results from the use of the electric field in order to generate a spray from the fluid surface. This latter phenomenon has been given the name "electro-hydrodynamic spraying" (Cloupeau and Prunet-Forch, 1994). This paper deals with electro-hydrodynamic spraying.

First observations of this electro-hydrodynamic spraying phenomenon were conducted by John Zeleny in 1914. His experiments were done in air and he used a single glass capillary for spraying. He used diluted hydrochloric acid, ethanol and glycerin as the conductive liquid.

Later on in the 1930's, Macky conducted research about behavior of water droplets in strong electric fields. In 1965, Taylor found the semi vertical angle of cone at the tip of capillary to be about 49°.

Zeleny's phenomenon was first fully described by Dole in 1968. Furthermore, in 1992, Fernandez de la Mora's solution explains the observed departure of the liquid cone angles from the spray-free value of  $49^\circ$  and predicts the droplet density as a function of position.

Practical applications are quite new. For example, in 1989 Fenn et al. used electrospray technique in mass spectrometry. This invention won the Nobel Prize for J. B. Fenn in 2002. Besides, electrospray technique is also used in aerospace applications, such as thrust and orbital control (Valesquez-Garcia, BGLF et al., 2008).

For many applications a single electrospray emitter cannot provide the required flow rate alone. Therefore it is often necessary to multiplex emitter arrays. Manufacturing of high density nozzle arrays is possible through state-of-the-art MEMS technology. For example, Kegi Tang et al. (2001) manufactured a multiplexed electrospray system with silicon micro fabrication and used it for mass spectrometry applications. Bocagnera et al. fabricated and tested an electrospray system with  $115 \text{ source/cm}^2$  density using a conventional CNC drilling method in 2005. This study proves that, there is no difference of flow rates between a single electrospray and a nozzle of multiplexed electrosprays. 1024 emitters within an area of  $0.64 \text{ cm}^2$  were manufactured and tested by Valesquez-Garcia for satellite thruster applications. Deng et al. (2007) manufactured a miniaturized ceramic combustor which has the same order of magnitude volumetric heat release rate with conventional gas turbines. This device is only  $0.22 \text{ cm}^3$  in volume. In 2009, Deng et al. fabricated an electrospray system with  $11,547 \text{ nozzles/cm}^2$  density. To date this value is highest nozzle density value reported in the open literature.

This paper is organized as follows; first the electrospray theory is briefly presented, and then a proof-of-concept demonstration is provided. Thereafter, the experimental results are compared against theoretical predictions. Finally, the paper concludes with a discussion section.

## Mathematical Model

Taylor explained the behavior of liquids issuing from the end of a thin tube due to electrostatic forces. He is the first to observe a conical form at the tip of the capillary needle when the charged liquid is attracted towards a ground electrode. This conical tip is named after him as the "Taylor cone". Taylor experimentally found that the cone half angle was  $49.29^\circ$  (Taylor, 1964). Later on he mathematically proves that, this angle is independent of fluid properties, applied voltage or the distance between the capillary and the ground electrode (Taylor, 1964).

Should a sufficiently strong electric field be applied to the system, electrostatic forces overcome the surface tension forces, as a consequence separation from tip of the Taylor cone occurs. The resulting flow is the so called cone jet. Starting voltage corresponding to this condition is provided in Eqn. (1).

$$V_{oc} = \sqrt{\frac{\gamma r_t}{\epsilon_0}} \ln \left( \frac{4d}{r_t} \right) \quad (1)$$

In the above expression (Eqn. 1),  $\gamma$  is the surface tension,  $r_t$  the radius at the tip of a hyperboloid needle and  $\epsilon_0$  is the permittivity of free space. Note that the free space permittivity constant is  $8,854 \times 10^{-12} \text{ C}^2 \text{N}^{-1} \text{m}^{-2}$ . Finally,  $d$  is the distance between tip of the capillary and the ground electrode. This mathematical relationship provides the minimum voltage needed to separate droplets from the tip of the Taylor Cone. When these droplets separate from the Taylor cone and fly towards the ground electrode the circuit is completed.

The minimum flow rate to start the electrospray operation is provided by Eqn. (2). Here  $\epsilon$  denotes the dielectric constant,  $K$  the conductivity and  $\rho$  the density of the liquid. This is the flow rate corresponding to the starting voltage.

$$Q_{\min} = \frac{\gamma \epsilon \epsilon_0}{\rho K} \quad (2)$$

The current of the system can be obtained with Eqn. (3). Inversely by measuring the electrospray current the flowrate can be obtained as well. Note that this relationship has been experimentally verified by Fernandez de la Mora (1994).

$$I = f(\epsilon) \sqrt{\frac{\gamma Q K}{\epsilon}} \quad (3)$$

The function  $f$  appearing in Eqn.3 is the derivative of the dimensionless spray current with respect to the dimensionless flow rate variable. It turns out that this derivative is only a function of the dielectric constant. Consequently, results from different liquids can all be collapsed into a single line. This result is quite important as it allows the indirect measurement of the electrospray flow rate via the current whose measurement is rather trivial. For further discussion reader is kindly referred to the extensive study of de la Mora (1994).

## Experimental Setup

A photographic view of the experimental setup is provided in Figure 1. A glass capillary tube of 0.1 mm inner diameter was used for electro spray injection. A syringe pump is utilized for bringing the liquid towards the tip of the needle. 1/8" O.D. tubing is used for connections between the capillary tube and the syringe pump. The liquid is positively charged by a high voltage DC power supply. The applied voltage is varied between 0-8 kV throughout the experiments. A ground electrode made of a conductive sheet metal is placed within a distance from the capillary needle.

Images of electro spray injection are recorded by a CCD camera. A far field microscope with a focal length of approximately 20 mm is mounted in front of the camera in order to increase the spatial resolution of the images. In this study, 10x and 20x magnification lenses are mounted onto the far field microscope. Also note that, the CCD camera was mounted onto a two axis traverse. Another traverse can be used for vertical adjustments (i.e. the distance between the capillary tube and the ground electrode). Therefore, the camera can move in all three directions for capturing desired images.

A digital multimeter is used to measure the electro spray current. A 10 k $\Omega$  control resistance is connected between the negative electrode and the ground (see Figure 2). Voltmeter is shunted to this resistance (see Figure 2). The electro spray current is inferred from the voltage drop across this control resistance. The experiments were carried out with methanol as the dielectric liquid. Physical properties of methanol are summarized in Table 1.

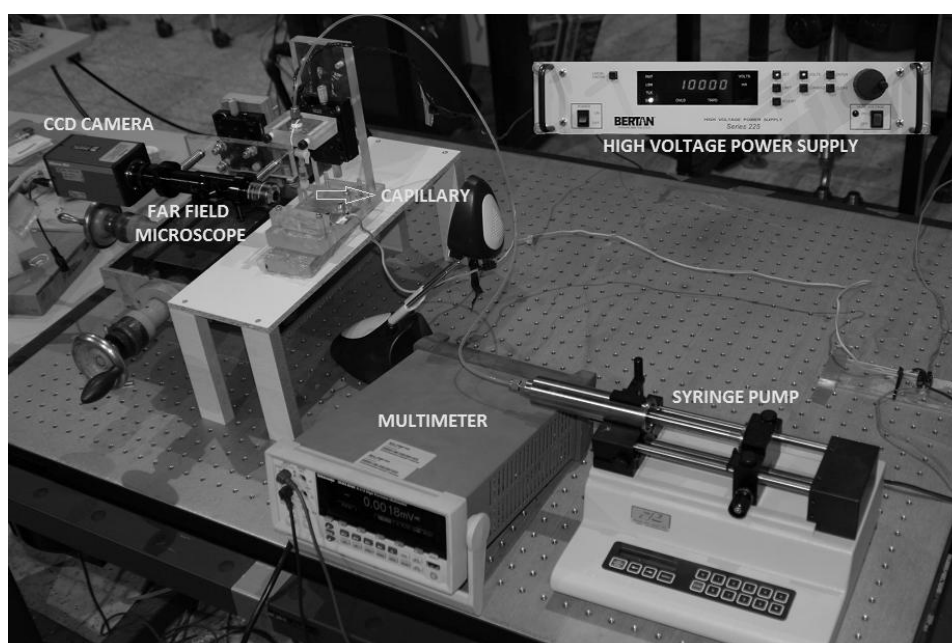


Figure 1 Experimental setup

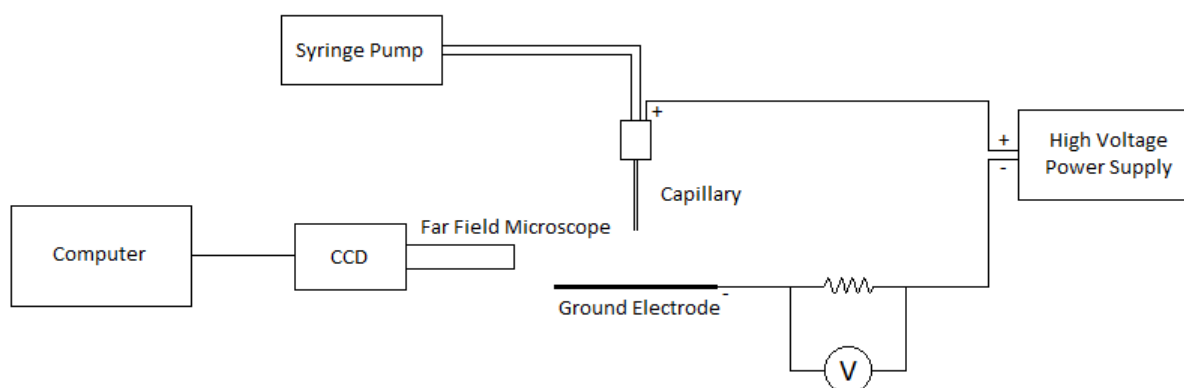


Figure 2 Schematic view of the experimental setup

Table 1. Physical properties of methanol

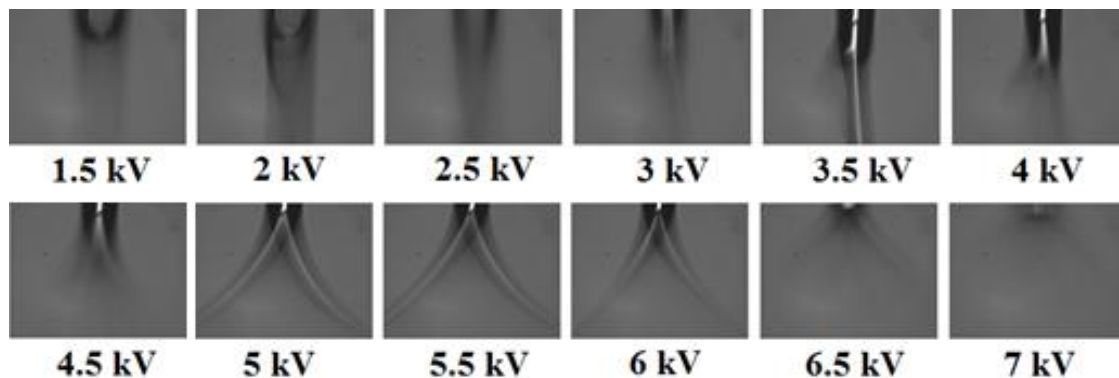
Density (kg/m <sup>3</sup> )	Surface Tension (N/m)	Relative Permittivity	Conductivity (S.m <sup>-1</sup> )
791.3	0.023	33.1	$2.1 \times 10^{-7}$

## Results

Note that as mentioned previously the syringe pump in the test rig is only utilized to bring the liquid towards the tip of the capillary tube. Consequently, electro spray occurs without infusion from the syringe pump, only with the applied potential difference.

Theoretical starting voltage value is calculated via Eqn. 1 for 15 mm distance between the tip of the capillary tube and ground electrode and methanol as the dielectric liquid. The theoretical value for the current configuration is 2.5 kV.

Experiments were carried out with voltage values are applied between 0-6 kV at 0.5 kV intervals. Electro spray views in different voltages are provided in Figure 3. In this experiment syringe pump was adjusted to 1 ml/h flow rate and distance between tip of the capillary and ground electrode is 15 mm. Besides, methanol was used as a fuel. When 1.5 kV was applied to the system, droplets were separated from the Taylor Cone. If the applied voltage is increased, length of the tip of the Taylor Cone approaches to the ground electrode. In our system, Taylor Cone combines at 3.5 kV and flow of electro spray becomes stable. This stabilization can be seen between 3.5 kV and 6 kV in Figure 3. Two sided flow can be seen in 5, 5.5 and 6 kV. The reason is that the ground electrode is rather wider from the diameter of capillary and consequently the shape of the consisted magnetic field is affected. In the frames corresponding to 6.5 and 7 kV test cases the image resolution turns out to be somewhat poor, since the capillary tube which is secured to the test rig like a cantilever beam cannot stand the electrical forces and starts to vibrate.



**Figure 3** Electro spray views for different electric field intensities

In Figure 4, the electro spray current is plotted as a function of potential difference between the charged liquid and the ground electrode. Results indicate a linear trend between the applied voltage and the electro spray current. A trend line is also plotted in Figure 4. The correlation coefficient of the line fit is  $R^2=0.99$ . The intersection point of curve fit and x-axis corresponds to the starting voltage of the system. As seen in the figure this value is about 2.2 kV. Note that when the electro spray initializes there is a sudden jump in the voltage drop across the shunt resistance. It is also possible to infer the starting voltage from this sudden jump.

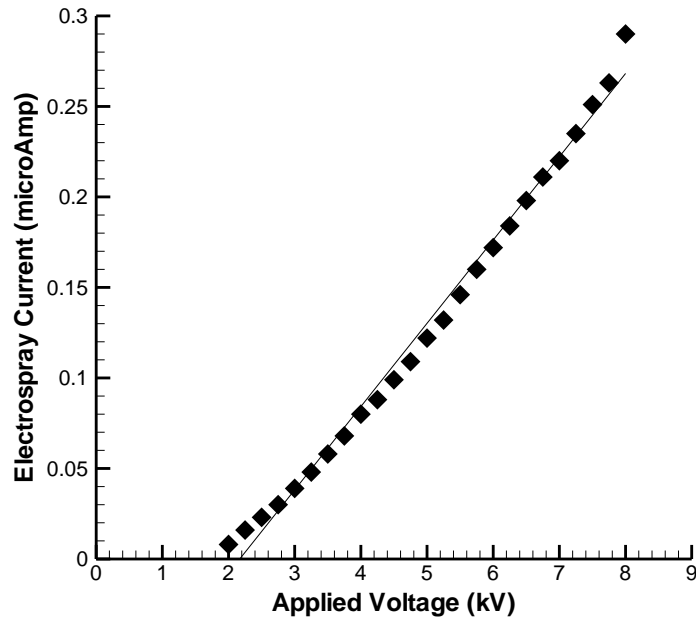


Figure 4 Electro spray Current versus Applied Voltage

Electrospray flow rate was calculated with Eqn. (3). In this equation, all values, except  $f(\epsilon)$  is known from Table 1.  $f(\epsilon)$  values vs.  $\epsilon$  for pure solvents is plotted by Fernandez de la Mora in 1994. Relative static permittivity for methanol is 33.1, and this value corresponds to  $f(\epsilon)=15$ . Thus, electro spray current vs. electro spray flow rate is plotted in Figure 5.

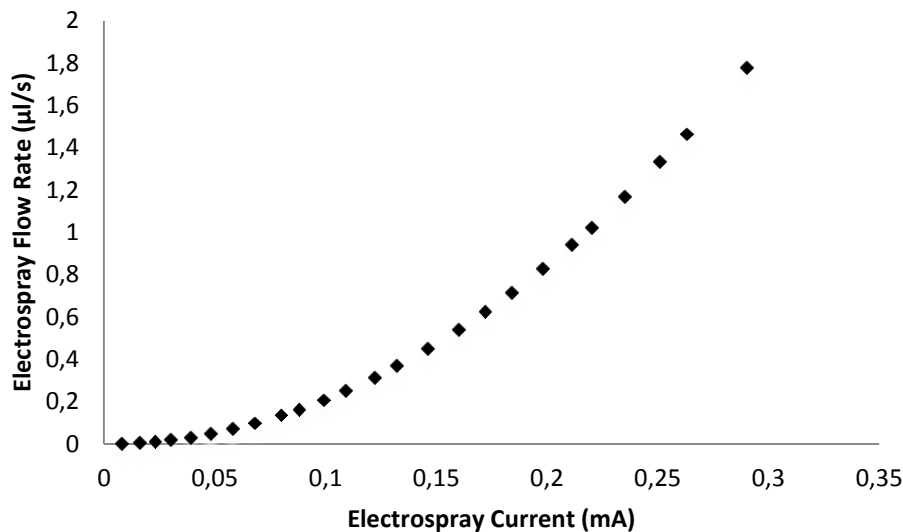


Figure 5 Electro spray Current vs. Electro spray Flow Rate

## Discussion

Starting voltage for methanol was found 2.5 kV theoretically and 2.2 kV experimentally. This 12% difference can be attributed to experimental uncertainty. Electro spray views in Figure 3 proves that the experimental system works properly and concludes the proof-of-concept demonstration of an electro spray injection system. Prospective studies shall focus on the manufacturing of dense electro spray emitter arrays. This experimental setup will be used and a similar set of experiments shall be performed for a compact multiplexed electro spray array manufactured with MEMS technology.

Flow rate from a single emitter is on the order of 2 ml/h. This barely corresponds to 17 W of thermal power for a typical liquid hydrocarbon fuel. Typical energy conversion efficiencies of thermoelectric generators are about 4% with the current state of the art. This would correspond to 0.67 W of electric power from a single emitter. For a 100 W electric power output, one would need about 150 emitters. Therefore it is imperative to use a multiplexed array of fuel emitters. Manufacture of high

density nozzle arrays is possible with the utilization of MEMS technology. Ongoing efforts at the ITU-MEMS laboratory aims at manufacturing electrospray emitter arrays at a density of 1000 nozzle/cm<sup>2</sup>. This suggests that for one centimeter square area of the emitter array 17 kW of thermal power can be generated. This thermal power can then be converted to electric power via a thermoelectric energy conversion system.

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## Nomenclature

d	Distance between tip of capillary and ground electrode [m]
Q	Volumetric flow rate [m <sup>3</sup> /s]
K	Conductivity [S/m]
r <sub>t</sub>	Radius at the tip of a hyperboloid needle [m]
V	Voltage [V]
V <sub>oc</sub>	Electrospray starting voltage [V]

## Greek Letters

$\gamma$	Surface tension [N/m]
$\epsilon$	Dielectric constant
$\epsilon_0$	Permittivity of the free space [C <sup>2</sup> /Nm <sup>2</sup> ]
$\rho$	Density [kg/m <sup>3</sup> ]

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