Dielectric Materials with Deposited Electrode Layers for Electrospray Arrays

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A two-dimensional electrostatic analysis is used to model the effects of geometric and material variations of the extractor of a single electrospray emitter. In particular, a geometry is modeled where the emitting capillary is embedded in a countersunk hole in a layer of Teflon and the extractor is comprised of a 20 nanometer thin-film of directly deposited gold on the dielectric. COMSOL software is used to solve Maxwell’s equations to determine the effect of the dielectric walls and emitter geometry on the electric field in the vicinity of the emitter. It is found that the dielectric only marginally changes the magnitude of the field but not the shape, which suggests that conventional scaling laws for onset voltage, applied potential, and flow rate still apply. These scaling laws then are used to design and construct a prototype of this configuration, baselining the ionic liquid 1-ethyl-3-methylimidazolium tetrafluoroborate (EMI-BF4) as a propellant. The design process, results of numerical model verification, as well as the initial prototype fabrication are presented.

I. Introduction

In the last few decades, growing interest in electric propulsion (EP) systems has been spurred by their relatively high specific impulse compared to chemical propulsion systems. This characteristic makes EP an enabling technology for a new class of deep space missions. The higher specific impulse (by an order of magnitude) inherently reduces the propellant mass of the spacecraft and therefore reduces the launch cost.¹ Of all modern EP devices, electrospray thrusters are particularly attractive as they have the highest power efficiency, a widely throttleable specific impulse (> 1000s), and the potential for having the highest thrust density (and therefore the smallest footprint).² These thrusters are based on electrospray technology where nano-sized conductive droplets, solvated ions or both, are emitted from a capillary through a fluid structure formed by a balance between surface tension and electrostatic forces, known as a Taylor cone singularity, and accelerated to high velocities through an electrostatic field that is created between an extracting electrode and an ionic liquid emitter.³ Individual electrospray thrusters or low-density arrays are a mature technology.⁴ These serve the purpose of station-keeping and precise orbital maneuvering, and have had marked success. With that said, these are goals of nano- or micropropulsion, and efforts to scale up electrosprays have been limited to date. However, the ability to reliably and optimally scale electrospray arrays in size could fundamentally advance space flight technology. Large-scale electrospray systems could operate at the theoretical limits of 90% efficiency with 1800-2000s of specific impulse and thrust levels only limited by the available power on orbit.

With that in mind, there have been a number of recent attempts to leverage advances in micro- and nanofabrication technology to try explore the production of large scale arrays of densely populated electrospray emitters. These attempts to date have reached certain upper bounds in array size, around 480 sites per square cm.⁵ Problems stem from issues related to alignment, manufacturing tolerance, chemical degradation, and arcing between the electrode and conductive liquid.⁶ This has limited arrays with a large number of electrospray thrusters to a low thrust range of 1-100μN with relatively short material lifetimes. While these performance metrics are still useful for a variety of micropropulsion applications, such as Cubesats, given the potential of these systems for future space exploration, the

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need is apparent to attempt to find first-principles and engineering solutions to these outstanding problems to allow the technology to scale to higher power.

While work continues to resolve these issues in the context of existing, established manufacturing techniques, we examine here a new architecture to attempt to scale these devices in size. This includes a careful consideration of geometry, fabrication, and material. Most notably, we propose to explore two innovations: first, etching a single homogeneous dielectric material for the emitter and the space between the capillary and the electrode; second, integrating recent advancements in 'self-healing' ablative conductive materials for the electrode. These ablative materials allow conductive surfaces to 'renew' in an arcing event by ablating or shedding singular layers of material and have the potential to extend lifetime performance of electrospray arrays. These new approaches are additionally poised to benefit future large-scale space systems by incorporating flexible design constraints based on simplistic fabrication methods.

This paper is organized as follows: in section A, we review electrospray scaling laws for single emitters and develop conclusions about how to build a single cone for a given voltage and propellant. In section B, we propose a design with novel materials utilizing the parameters quantified by the previous scaling laws and then identify key concerns with this new geometry. Section C uses modeling to address the concerns with both electromagnetic software and computational fluid dynamics. In section D, we outline the fabrication of the experimental prototype and introduce questions for experimental verification. Finally, we discuss the experimental setup along with planned diagnostics and propose a test plan for the prototype.

II. Methods

A. Scaling laws for electrosprays

In order to optimize the performance parameters and provide a basis for the initial prototype that incorporates novel materials and geometry, we aim to design a single emitter based upon a foundation of theoretical scaling laws and prior experimental results. These scaling laws will focus primarily on the relationship between the dimensions of the emitter capillary, as well as the geometry of the region between the emitter tip and the extracting electrode, and their subsequent effect on electrospray onset voltage, droplet formation, and flow rate.

1. Onset Voltage

Electrosprays are driven by the electrophoretic mechanism to produce charged droplets at the tip of the emitter capillary (Figure 1), where the radius of the capillary is very small (10-100 μm), leading to a high electric field (on the order of 10^6 V/m) in air at the tip. This relationship can be expressed as

\[ E_c = \frac{2V_c}{r_c \ln\left(\frac{4d}{r_c}\right)} \]  

where \( E_c \) is the electric field, \( V_c \) is the applied potential, \( r_c \) is the outer capillary radius, and \( d \) is the distance from the capillary tip to the extracting electrode. This expression originated directly from the solution of an electric field at a point, approximating the tip as a revolution of a hyperboloid, and furthermore, assuming the capillary radius was approximately equal to the tip size. We can see here that the electric field is then inversely proportional to \( r_c \) and the field decreases slowly due to the logarithmic dependence of emitter distance, thus the capillary radius has a larger influence on the field. Moving forward, we will set \( d \) based on previous experimental prototypes and work to solve for the capillary radius, \( r_c \).
To find the range of applied potential for nominal operation, the minimum potential required to overcome the surface tension within the conductive fluid must be determined. This is referred to as the onset voltage $V_o$, and has the following relationship not only with the dimensions of the device, but to the surface tension of the fluid $\gamma$ as well, where $\varepsilon_0$ is the permittivity of free space.$^{15}$

$$V_o = \frac{\sqrt{\gamma r_c \ln\frac{2d}{r_c}}}{\varepsilon_0}$$

(2)

The physical origin of this equation can be derived from the condition of the interface of the fluid film deforming as the outward peak electric stress, $\sigma_E$, defined as

$$\sigma_E = \frac{1}{2} \varepsilon_0 E_c$$

(3)

becomes greater than the surface tension stress

$$\sigma_\gamma = \frac{2\gamma}{r_c}$$

(4)

By combining Eq. (1), (3) and (4), we arrive at the conclusion that the electric field determines the onset of emission from an electrospray capillary, and thus, once the onset voltage is chosen for a particular fluid, we can derive the necessary geometric parameters for the capillary. In general, the applied voltage of the electrospray should be at least several hundred volts higher than the onset voltage required, in order to support a stable spray.$^{16}$ From here, the applied potential then brings us into the operational emission regimes of the electrospray thruster: droplet mode, ion mode, and a mixed ion-droplet mode. For the purposes of this simple prototype model, we will consider the parameters within the droplet regime only.

2. Droplet Formation

The atomization process of the electrospray plays a large role in the performance of the system as a propulsive device, primarily due to the charge-to-mass ratio, $q/m$, which is directly related to the measure of energy available in the fuel and its respective thrust conversion efficiency. This quantity, known as specific impulse, $I_{sp}$, is defined as

$$I_{sp} \equiv \frac{\tau}{m \cdot g_0} = \frac{v_e}{g_0}$$

(5)
where $T$ is the thrust, $\dot{m}$ is the mass flow, $g_0$ is the gravitational acceleration of the earth, and $v_e$ is the exhaust velocity of the thruster. For an electric propulsion device, the exhaust velocity can be calculated from the kinetic energy of a charged particle

$$v_e = \sqrt{\frac{2\Phi_B m_0 q}{m_0}},$$  \hspace{0.5cm} (6)$$

with $\Phi_B$ as the potential difference that accelerates the particle. The mass flow rate, assuming charge conservation (although droplet size varies and thus charge-to-mass ratio is not constant, we can assume an average), is

$$\dot{m} = I \frac{m_0}{q},$$  \hspace{0.5cm} (7)$$

where $I$ is current. From here, we can see the fundamental difference between thrust and specific impulse in relation to charge-to-mass ratio is given respectively as

$$T = I \sqrt{\frac{2\Phi_B m_0}{q}},$$  \hspace{0.5cm} (8)$$

$$I_{sp} = \frac{1}{g_0} \sqrt{\frac{2\Phi_B q}{m_0}}. $$  \hspace{0.5cm} (9)$$

For electrospray devices, it is the charge-to-mass ratio, in addition to the current, that determines which regime the thruster operates within. The droplet mode has a lower charge-to-mass ratio, and therefore operates at a higher thrust and inversely, the higher the charge-to-mass ratio, the higher the specific impulse. In addition to current, applied potential, and charge-to-mass ratio, the emission mode and operation of the electrospray is also a function of flow rate.

3. Flow Rate

The evolution of the surface of the fluid into a cone can be treated as a simple steady state equilibrium balance of surface tension and electrostatic force. At the apex of this perfect Taylor cone, the normal electric field would be infinite, but this does not actually exist, as the singularity is broken by emission of charge through droplets or ions from the tip region. For this to occur, current must be conducted through the liquid to the apex of the cone through an ohmic radial electric field, driving current against the conductivity of the fluid, $K$, in which the radial field becomes tangential to the cone surface through convection of current, creating a shear stress on the liquid. The liquid is then accelerated towards the apex, along with the ions and is released as a thin jet, or a stream of ions, or both. This combination of effects was considered and the current transported in the cone-jet formation of an electrospray device, operating within the droplet regime, was experimentally verified by de la Mora as

$$I = f(\epsilon_r) \sqrt{\frac{\rho K Q}{\epsilon_r}},$$  \hspace{0.5cm} (10)$$

where $\epsilon_r$ is the dielectric constant of the fluid, $K$ is the fluid conductivity, $Q$ is the flow rate, and $f(\epsilon_r)$ is an experimentally determined constant. There is a limitation to the dropwise emission, which is not well understood, corresponding to a minimum flow rate, and it has been correlated to a strictly experimental, dimensionless parameter

$$\eta = \left( \frac{\rho K Q}{\epsilon_r} \right)^{\frac{1}{2}},$$  \hspace{0.5cm} (11)$$

where $\eta$ reaches values of 1. This is a ratio of the minimum flow rate that supports a Taylor cone and thus atomization of the liquid can be approximated as $\lim \eta \rightarrow 1$, resulting in the following relation that is entirely a function of the fluid properties.
where $\rho$ is the fluid density. While there is no direct scaling law for the length of the capillary in regards to flow rate or current, experimental results show that flow impedance occurs with higher aspect ratio capillaries, and thus also drives the mode of operation. Therefore, deriving a minimum flow rate as a lower bound with the necessary pressure drop across the capillary length for operation will be an important quantity to verify for experimental testing within the emitter prototype.

From these relations between onset voltage, droplet formation and flow rate, we can choose a conductive liquid and derive a theoretically driven model for the geometry of an electrospray thruster to operate within the desired regime and performance requirements.

### B. Design Parameters

We begin with the choice of conductive liquid. Given the successful precedence in electrospray thruster research, 1-ethyl-3-methylimidazolium tetrafluoroborate, (EMI-BF4) will be used for the prototype design and experimental testing. Table 1 lists the fluid properties, in SI units.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\gamma$</td>
<td>0.0452 N/m</td>
</tr>
<tr>
<td>$K$</td>
<td>1.4 S/m</td>
</tr>
<tr>
<td>$\rho$</td>
<td>1240 kg/m$^3$</td>
</tr>
<tr>
<td>$\epsilon_r$</td>
<td>12.9</td>
</tr>
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</table>

From Eq. (2) we see the linear dependence of onset voltage and extractor distance. Working from experimental and simulated results for electrosprays that utilize EMI-BF4,$^{22-23}$ we can begin with a baseline onset voltage of 1400 V, and an extractor electrode distance $d$ of 1 mm. Plugging these values into Eq. (2), we solve for the capillary radius of 17 $\mu$m. By halving the extractor distance to 0.5 mm, the starting voltage can be dropped to closer to 1200 V. Using the above fluid parameters, we can use Eq. (12) and calculate the minimum flow rate required to sustain droplet formation to be $0.003$ nL/s. Further calculating theoretical performance parameters, if we choose a nominal operational flow rate of 100 nL/s for Eq. (10) and use de la Mora’s approximate value of $f(\epsilon_r)$~18, we calculate the current to be 12.6 $\mu$A. Based on experimental data from studies that employ EMI-BF4, the average mass-to-charge ratio at room temperature is found to be 1.5E-6 kg/C.$^{24}$ Assuming 2000 V as the applied potential, we calculate a theoretical thrust from Eq. (8) as 975 nN. Without efficiency losses, this number is somewhat higher than what is found experimentally from single emitters, but it gives us a basis for the dimensions of the prototype that should operate based on the above theoretical framework.

The first objective in our design architecture is to eliminate alignment issues by removing the traditionally separate electrode grid altogether, and instead deposit a thin-film conductor directly on the emitter substrate, as modeled in Figure 2. The emitter will be constructed from a dielectric substrate, with a low relative permittivity of 2 - 10 F/m. The second objective aims to reduce the occurrence of material degradation and destructive arcing incidents across an array of emitters by employing a ‘self-healing’ electrode film. These films can support 600 – 800 V/$\mu$m$^{25-27}$ and can be fabricated through vacuum evaporation directly onto the substrate array. If an arc occurs between a Taylor cone and electrode, the current conducted through the electrode ablates the material and the failure occurs only locally, so the other emitters in the array are not impacted.
Due to the presence of a dielectric material within the extraction region and the new electrode material itself, important questions arise from the design, including the effect of the dielectric walls on the electric field and how the geometry might alter the onset and applied voltage in order to maintain operation and atomization of the propellant. Indeed, it is possible that the dielectric may preclude the development of the required field shape to promote Taylor cone formation, as all known electrospray devices operate within air at atmospheric conditions or under vacuum between the extractor electrode and the emitter, which have a dielectric constant between zero and one. We explore this question numerically for a single emitter geometry in the next section.

C. Numerical Verification

The most used physical model for droplet formation from a steady field is the Taylor-Melcher Leaky Dielectric model, developed by J.R. Melcher and G.I. Taylor in the mid-1960s. This model directly couples electrostatic phenomena with hydrodynamics through the Maxwell stress tensor and allows free charge to exist at a liquid-gas interface, as shown in Figure 3. Developing a numerical simulation from electrohydrodynamic (EHD) atomization usually requires the input of an electric field, coupled with the EHD equations by modifying the Navier-Stokes equations to include electric body forces, as seen in the model used by O. Lastow and W. Balachandran.

Due to the inherent difficulties of coupling a conductive fluid model with an electrostatic field, in addition to the mathematical singularities found at the apex of a fluid Taylor cone and the boundary conditions along the surface of the cone, we opted to explore the effect of the dielectric on the electric field using a zeroth order electrostatic model substituting a conductive solid cone (aluminum) with the same dimensions as a perfect steady-state liquid Taylor cone, in place of the ionic propellant. Using COMSOL Multiphysics (Version 5.3) AC/DC module, we implemented a model of the dielectric emitter and the geometric parameters to determine their impact on the electric field in the vicinity of the emitter tip. This code is 2-D axisymmetric and solves Maxwell’s equations for steady-state electrostatic potential between two electrodes.
\[ \nabla \cdot \mathbf{D} = \rho_v \tag{11} \]
\[ \mathbf{E} = -\nabla V \tag{12} \]

By employing a potential voltage of 1kV on the extracting electrode while holding the Taylor cone as ground, we were able to confirm the electric field streamlines to be normal to the cone surface in Figure 4. Figures 5 and 6 respectively show the electric potential contours and a field intensity to be within the same order of magnitude that is seen in electrospray devices, with a peak threshold at the apex.

These simulations were run with a material dielectric constant set to 2.0 F/m, but to further understand how the magnitude of the electric field would scale with the choice of dielectric material, we varied the dielectric constant and found a decreased field intensity with increased dielectric constant, shown in Figures 7 and 8. This led to the conclusion that a material with a lower dielectric constant (such as Teflon) would be a better choice for our prototype, in order to lower the required onset voltage and therefore the applied operational voltage.

Continuation of these numerical investigations coupled an electromagnetic solver with a volume-of-fluid (VOF) model using STAR-CCM+ (Version 12.06) to determine how more complex surface interactions between the dielectric and the ionic liquid interact, while not directly solving the EHD model equations, but with particular
attention as to whether or not the fluid will “creep” beyond the capillary opening or up the walls of the emitter. These 2-D axisymmetric simulations were run with the Eulerian Multiphase, VOF model using EMI-BF4 material properties and air under high vacuum conditions, with 1kV potential between the capillary and the extracting electrode. The inlet flow rate was varied from 1000-5000 nL/s. The contact angle at the capillary wall and outlet was also varied between 6 and 45 degrees, as the contact angle for ionic liquids on dielectric surfaces is difficult to quantify, due to surface texture and the amorphous nature of the material. Figure 9 shows the boundary conditions and dimensions of the final experimental prototype’s fluid region (detailed in the following section D), which was scaled up by three orders of magnitude for better meshing convergence. Figure 10 shows the scalar plot of the volume fraction of EMI-BF4 and Figure 11 shows a close-up of the same scalar plot at the interface. Using this model as initial verification, we have concluded that the ionic fluid will not flow beyond the capillary edge and indeed, the geometry and operational conditions should produce a Taylor cone formation.

D. Prototype Fabrication

Utilizing these design parameters, a single dielectric electrospray emitter was fabricated in-house from a Teflon substrate, using CNC boring with an imbedded fused silica capillary and vacuum evaporation for the thin-film electrode. Teflon test coupons 0.25-inch-thick were first sanded to even the surface for electrode deposition. The top side was then sputtered with gold for 4 minutes, to create a 20 nm thin-film electrode surface. Using a CNC and a micro drill bit of 0.0156-inch, we bored a hole flush through the coupon. A larger 0.125-inch bore was drilled from the bottom of the coupon up 0.15-inch towards the electrode surface to create a fluid reservoir for the capillary via a 0.125-inch outer diameter PEEK tubing fluid supply line. For the emitter capillary, a 2-inch long fiber optic fused silica capillary tube, with an inner diameter of 15± 2 μm, and an outer diameter of 363± 10 μm was placed inside the
smaller bore. The capillary tubing was pushed flush to the top electrode surface, and pulled out 0.02-inch from the electrode coated surface to create the extraction region and was secured into place with a cyanoacrylate adhesive. The PEEK tubing was inserted into the bottom of the coupon into the larger bore and secured in place with adhesive. Figure 12 outlines the final dimensions of the initial prototype. The fabricated prototype coupon is shown in Figures 13-15.

![Figure 12. Prototype dimensions](image)

Figure 12. Prototype dimensions

Figure 13. Prototype coupon electrode surface

Figure 14. Prototype underside view of capillary stem

Figure 15. Prototype side view of capillary stem without PEEK tubing reservoir

A Nikon AZ100 microscope was used to optically verify the bore and capillary radii, as well as examine the region surrounding the electrode bore. Figure 16 highlights several important observations, specifically the roughened surface of the electrode, which can be reduced in future iterations by polishing the Teflon surface first. Additionally, the bore hole is not symmetrical and has many jagged edges, which will likely affect the electric field due to increased charge density at the points, possibly compromising the performance of the emitter. Future prototypes might benefit from UV laser boring in place of a drilled hole for smoother electrode edges. Looking further down into the bore in Figure 17 we can see the reflected light through the smaller capillary radius, with some particulates along the bored walls of the dielectric, but the walls look to be relatively smooth, which ensures the electric field shape and magnitude will follow the modeled behavior.
Based on the theoretical and numerical framework presented here, we have started preparing an experimental facility to test this prototype. The Electrospray Thruster Array Chamber (ETAC), housed within the University of Michigan’s Plasmadynamics and Electric Propulsion Laboratory (PEPL) is a 3-foot-diameter by 4-foot-long steel chamber with nine ports and doubly-hinged caps on both ends which will be pumped to high vacuum with a turbomolecular pump, shown in Figure 18.

Planned diagnostics include high speed video to monitor the droplet emission beam, a single gate time-of-flight (TOF) mass spectrometer to provide the particle size distributions and charge-to-mass ratio, as well as a residual gas analyzer. An UltraVolt “HV Rack” 4-channel high voltage power supply will supply the voltage for the propellant feed line, the focusing lens, and the electrostatic gate for the TOF set up. The experimental configuration is shown graphically in Figure 19, where $\varphi_a$ is the applied voltage to the propellant line, $\varphi_f$ is the focusing lens voltage, and $\varphi_g$ is the gate voltage. A programmable pulse/delay generator will be used to produce the gate signal. The thin-film extracting electrode will be held to ground with a surge resistor between the extractor and an ammeter for measuring current. A second power supply will be used to bias the collector plate in place of an additional gate at the end of the TOF path to reduce secondary electron emission. Additionally, a variable pulse amplifier module will be used to improve the signal from the TOF apparatus to the oscilloscope.
Given the single emitter geometry and limited length of testing per each prototype coupon, we will not be using an alternating voltage circuit, in order to make energy distribution and current/voltage data collection easier in DC mode. The ionic propellant EMI-BF4 will be used, based on its successful precedence in current laboratory electrospray systems, which will be pumped by a Harvard Apparatus Model 11 single syringe pump. All systems and data collection will be run with LabVIEW software.

Figure 19. Proposed schematic of ETAC with diagnostics

IV. Planned Testing

In upcoming experimental tests, we plan to specifically define the minimum flow rate, onset voltage, and applied voltage for operation of the prototype. We also plan to produce a current vs voltage curve to quantify the emitter performance, in addition to finding the charge to mass ratio of the emission for a sweep of applied voltage and flow rates. Important questions we wish to address include what regime the single emitter prototype operates in (likely droplet or mixed ion-droplet), as well as whether the device geometry needs to be adapted due to beam interception, to improve flow impedance or other operational and performance metrics. We will also perform critical post-testing analysis to look for chemical degradation of the materials, especially along the walls of the dielectric and the surface of the electrode bore. As we have the capability for rapid-prototyping, we can iteratively refine models based on the results from these key experimental tests.

V. Conclusions

We propose to use this novel configuration of a dielectric substrate emitter with a directly deposited electrode for an electrospray thruster in order to remove the issues associated with optical alignment and address lifetime limitations by utilizing ‘self-healing’ conductive thin-films in the future. Outside of these potential benefits, we had several concerns about the operation of such a device. These included issues related to capillary action and shielding of the electric field due to the presence of a dielectric within the extractor region, preventing the onset of Taylor cone formation. We designed an emitter based on known scaling laws for a single emitter in vacuum, and then implemented geometry in two different models and showed these concerns numerically do not appear to be an issue. Following this, we have manufactured a prototype and have detailed an experimental setup to test it in the near future.
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References


