Experimental Characterization 1-Butanol Electrospray Phenomenology

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Abstract
This study presents the visualization and characterization of electrosprays of 1-butanol that burn in air. Spray phenomenology was visualized and features such as shape, size, and stability of the spray flame were determined. Phase Doppler Anemometry (PDA) and Mie-scattering-based droplet size measurements were performed in both reacting and non-reacting electrosprays. In general, a strong peak in the distribution of droplet diameters centered between 40 and 50µm was observed in the non-reacting case, with a second smaller peak observed around 200µm. In the reactive case, a broader size distribution was observed, still with a peak between 40 and 50µm, but also with a larger percentage of droplet diameters between 20 to 50µm. The Mie scattering results were compared with PDA measurements taken at the center of the spray for several voltage and flow rate conditions. In both cases, Reynolds and Weber numbers were calculated and were related to observed spray and flame phenomenology. In addition, velocimetry measurements provided insight to the velocity of the droplets inside the burning spray. Some droplets were observed not to vaporize completely and passed through the flame. Photographic evidence was acquired of droplet fission in the burning spray. Charge transferred by the spray was also measured and compared with the Rayleigh limit criterion for droplet fission. An analysis of the evaporation time, based on droplet size and velocity, was used in order to rationalize the results. Indeed, large droplets traveling with the measured velocities could pass through the flame without completely evaporating. It was concluded that the electrosprays of 1-butanol could sustain flames stabilized with the assistance of electrostatics. Spray flames with electrostatically assisted atomization and stabilization were shown to have characteristics substantially different from the spray flames of non-charged fuels.

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1 Introduction

Electrospray is an injection system which causes liquid atomization using electrostatic forces, without the use of a large pressure differential. Previous electrospray combustion research has been primarily conducted with traditional hydrocarbon fuels, such as heptane. Gomez and his collaborators at Yale University performed studies with heptane in a counter flow burner, injecting through an electrospray and studying the effect on the flat flame produced in their configuration [1]. Specifically mentioned in that paper was the inability for direct electrospray combustion, citing the breakdown of the electric field because of the high concentration of chemi-ions in the flame. Additionally, larger droplets were observed to have a higher average velocity than smaller ones, and, therefore, had lower residence time in the flame [1]. In a subsequent study, the same group sought to detect and capture evidence of droplet fission in the flames of a counter-flow burner. Their theory sought to explain the rapid loss of larger sized droplets, given that evaporation effects were insufficient to account for this finding. In several images, convincing evidence was presented of abnormally close droplets in areas with otherwise sparse populations [2]. This would suggest that one larger droplet broke apart into several smaller droplets as a result of Rayleigh Limit induced fission. Shirpton and his collaborators [3] have performed numerous studies on electrospray and droplet characteristics, most recently evaluating Reynolds and Weber number effects of both laminar and turbulent regimes on spray atomization. This group introduced a modified Weber number to take into account the effective reduction of surface tension in the presence of the electric field [3].

Electrospraying is particularly appropriate for bio-alcohols that have an electric conductivity of at least four orders of magnitude higher than the ones of hydrocarbons. With recent advances in large scale bio-butanol production [4, 5], butanol may become a mainstream transportation fuel in the future. Butanol has many advantages over ethanol, which is currently the most popular liquid biofuel. In addition to having a higher energy density than ethanol (36.4 vs 24.8 MJ/kg), butanol requires less molecular breakdown during production, which translates into less energy needed in production. Furthermore, butanol can be easily transported in pipelines and it can be mixed with diesel fuel without the need for additives. Promising combustion benefits with the use of electrosprays have been demonstrated in enhancing fuel injection in internal combustion engines [6, 7], and power generation [8] with a particular emphasis on micro-combustion [9, 10]. Research into electrospray enhancement of fuel injectors for use with ethanol-gasoline and butanol-gasoline blends has been conducted [7, 11], showing benefits in emission reduction. More recently butanol electrosprays have been compared to those of ethanol and conductivity enhanced heptane by Agathou and Kyritsis [12, 13].

Published literature lacks fundamental studies of butanol electrospray combustion. With this study, we would like to offer some initial experimental data about this combustion phenomenon. Flame phenomenology was described, and the effect of the presence of the flame on defining parameters of the spray, such as droplet size and velocity, was examined and compared to previous studies on electrospray flames. Of particular interest was the dual effect of electrostatics on both atomization and flame stabilization.

2 Experimental Apparatus

The experimental apparatus is shown schematically in Figure 1. The electrospray is formed by connecting a steel capillary connected to a syringe pump and high voltage source. The capillary had an inner diameter of 127 µm and a conical tip that was machined using an Electrical Discharge Machine to enhance the electric field at the nozzle.

The flow of liquid butanol ran opposite to gravity, and it was metered with a KD-Scientific syringe pump. n-Butanol was used in all of the tests because this is the dominant isomer in bio-butanol mixtures. The distance between the capillary tip and grounded mesh was fixed at 12.5 cm for all tests. Voltages of 6.5-7.5 kV and flow rates in the range of 10-20 ml/hr were found to produce self-sustaining air-stabilized flames. An air-stabilized flame is defined as a flame that burns at a substantial distance above the capillary tip, anchored at a location that is essentially determined by the fuel flow rate at a given voltage. At each flow rate, the potential field was varied at 6.5, 7.0 and 7.5 kV. However, the important parameter is the potential divided by the distance between the cathode and the anode. With the fixed gap of 12.5 cm, this translates to 520, 560, and 600 V/cm respectively. At each voltage setting, the butanol flow rate was varied at 10, 15, and 20 ml/hr.

Droplet size measurements were obtained using a Malvern Spraytec 97 droplet sizer. This device utilizes a 670 nm laser and detector system. Also, a Dantec PDA system with the capability of two-dimensional velocity measurements was used to capture both droplet diameter and velocity. The
Figure 1. Laser sheet through the air-stabilized flame

The system was equipped with a BSA P60 processor connected to 58N70 detector. The laser used in the PDA system was a Spectra Physics Stabilite 2017 Argon-ion.

3 Electrospay Phenomenology

The factor that determined the mode in which the electrosprays combusted was the ratio between droplet spacing and droplet size. As the droplets became bigger and spread farther apart, the single droplet combustion mode was preferred (Fig. 2). Large droplet spacing inhibited the flames of individual droplets from merging together and forming a larger flame fueled by droplets burning in the group combustion mode.

If the droplets are small and close to each other, the droplets burn as a group (left image in Fig. 3), with an intermediate state being internal group combustion (right image in Fig. 3). Clearly, electrostatic control can drastically affect flame morphology. Furthermore, in all of the test conditions, some of the droplets either deviated away from the flame before reaching it or passed at a distance from the flame that prevented complete evaporation and subsequent ignition.

Figure 4 shows a non-burning spray at 600 V/cm. Under this applied potential, an almost dual spray structure appears, that is much more pronounced than in lower voltage cases. Specifically, a narrow central core spray (red arrows) and a much broader outer spray (blue arrows) are clearly visible. When the flame burns in the mode shown on the left panel in Fig. 3, it burns on the fuel from the inner spray cone, whereas the much broader flame on the right panel of Fig. 3 received fuel from both the inner and outer spray cones. The existence of two cones is consistent with the findings of Tang and Gomez [14], who observed what they termed “satellite droplets” in their flames.

Figure 2. Single droplet combustion mode with laser sheet present
4 Droplet Size Analysis

Droplet size measurements were performed as close to the base of the flame as possible without having the measurement volume inside of the flame, so that beam-steering effects were avoided. This was 18 mm from the tip of the capillary. Many combinations of electric field and flow rate were tested; however, only results acquired with a field intensity 560 V/cm and mass flow rate of 15 ml/hr case are presented for the purpose of brevity. Probability density functions of droplet size are shown in Fig. 5 on the following page with and without a flame present. The probability density of a particular droplet volume is presented as a function of the diameter corresponding to this volume. Significant differences can be seen between the two cases. The size distribution when the flame is not present is notably different from the monodisperse sprays observed in previous studies in the cone-jet mode [2, 1].

In the reacting case, the distribution becomes much more peaked around a central value - in this case about 42 µm. Both the number of droplets larger and smaller than this central size have decreased, most likely a result of evaporation of the droplets as they approach the high temperature region of the flame. This would reduce the size of the largest droplets and nearly eliminate the smallest droplets.

The velocity component along the spray axis versus droplet diameter is presented in Fig. 6.

A wide range of velocities for each droplet size is observed; however, larger droplets on average have a lower velocity. During combustion, the droplets with the median size were measured to have a larger range of velocities than during the non-combusting case. This could be due to the large droplets evaporating but remaining at the same velocity as they approach the high temperature region of the flame. Furthermore, these droplets have the largest surface area and would vaporize at a faster rate than smaller droplets, as described in equation 4.

Droplet diameters were also measured using the Mie-scattering-based Malvern Spraytec. The Malvern takes a line average across the test region, unlike the essentially point measurement of the PDA. The Malvern also captured data at 18 mm from the capillary tip. It is further noted that, the Malvern results are volume-based PDFs, so a direct comparison between the output of the two instruments should be made carefully.

In the non-combusting e-sprays, the majority of the spray volume is contained in droplets between 20 and 60 µm using both measurement devices. Interesting differences between the two measurement devices do exist. Both droplets that are smaller and larger than the smallest and largest sizes measured in the PDA experiments are present. This could be due to droplets that have escaped the electric field and fall back under gravity could be measured again using the Malvern, increasing their volume percentage. This is not a issue encountered with the PDA because the measurement location was chosen to be at the center on the spray.

Furthermore, when measuring near the e-spray
Probability density function without combustion (top) and with combustion (bottom) flame, the droplet size distribution broadens across droplets sizes less than 100µm when using the Malvern but becomes narrower using the PDA. Previous work by Yule et al. [15] has suggested that the droplets on the periphery of electrospays are usually smaller and highly charged. Photographic data indicated that the droplet spacing in a combusting e-spray was lower than in the non-combusting e-sprays. In these more compact sprays, the larger droplets exist near the spray axis, whereas smaller ones are near the periphery. Since the measurement location of the PDA is fixed, it recorded a much higher percentage of larger-sized droplets. However, the line-integrated measurement of the Malvern continued to capture all of the droplets sizes present. Furthermore, a reduction in droplet size should be expected as the droplets approach the high temperature region near the flame. Because of the reduction of droplet spacing, the PDA should only record a reduction in the maximum size of the droplets, because the largest droplets are located at the core of the spray. Comparing both the non-combusting and combusting e-spray data taken with the PDA, the largest droplet size is reduced from 65µm to about 50µm during combustion. Data captured with the Malvern reveals a reduction of all droplet sizes and a broadening of the entire distribution, possibility due to the different rates of evaporation of droplets at different locations in the spray and with varying initial diameters. Another explanation of the reduction of the droplet diameters could be droplet fission, which will be discussed in detail in section 5.

In the Malvern data, droplets diameters around 300µm were measured with and without a flame. More detailed probing of both combusting and non-combusting electrospays with the PDA is underway at this time to help provide a more conclusive theory as to the differences between the measurement systems. In particular, the size and velocity of the droplets on the periphery of the e-spray.

The captured flame images and droplet velocity distributions reveal two important characteristics of these burning electrospays. First, droplets are not all completely vaporized by the time they reach the flame. Second, droplets moving at high speed (>10 m/s) are observed inside the flame envelope that

Figure 5. Probability density function without combustion (top) and with combustion (bottom)

Figure 6. Comparison between Droplet Diameter and Vertical Velocity (non-combusting top, combusting bottom)
surrounds the spray. Following the analysis of Turns \cite{16}, we provide a plausible explanation for droplets passing through the flame. In particular, estimating the thermal conductivity of the gaseous butanol and air mixture $k_g$ as

$$k_g = 0.4 \times k_{C_4H_{10}O} + 0.6 \times k_{Air}$$

(1)

Also, $B_q$ is the ratio of the sensible enthalpy in the vaporized butanol to the enthalpy of vaporization

$$B_q = \frac{C_{P_g} \times (T_\infty - T_{boil})}{H_{f_g}}$$

(2)

$T_\infty$ is the adiabatic flame temperature for a butanol-air flame, $T_{boil}$ is the boiling temperature for butanol at 1 atm, $C_{P_g}$ is the specific heat of the butanol vapor, and $H_{f_g}$ is heat of vaporization of butanol.

The evaporation constant “K” can be then estimated as:

$$K = \frac{8k_g}{\rho_{C_4H_{10}O} \times C_{P_g} \times ln(B_g + 1)}$$

(3)

In such a case, the evaporation time $t_D$ is given by, where D is the initial droplet diameter:

$$t_D = \frac{D_0^2}{K}$$

(4)

The results of this calculation are presented in Table 1, based on an observed flame size of 6.5 cm. The critical speed, as defined here, is the maximum speed at which a droplet can traveling to ensure complete vaporization before leaving the flame region. Therefore, droplets traveling faster than this velocity would pass through the flame before vaporizing completely.

<table>
<thead>
<tr>
<th>Droplet Diameter ($\mu$m)</th>
<th>Evaporation Time (ms)</th>
<th>Critical Speed (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>4.76</td>
<td>13.7</td>
</tr>
<tr>
<td>55</td>
<td>5.76</td>
<td>11.3</td>
</tr>
<tr>
<td>60</td>
<td>6.86</td>
<td>9.48</td>
</tr>
<tr>
<td>65</td>
<td>8.05</td>
<td>8.08</td>
</tr>
<tr>
<td>70</td>
<td>9.33</td>
<td>6.96</td>
</tr>
<tr>
<td>75</td>
<td>10.1</td>
<td>5.33</td>
</tr>
</tbody>
</table>

Table 1. Evaporation time and critical speed to completely vaporize

Indeed, 65-75$\mu$m droplets moving faster than the calculated critical speed were observed at the base of the spray flames. Thus, this simple model provided a plausible justification of the observations of droplets passing through the flame before completely vaporizing.

5 Droplet Fission

Strong evidence of droplet fission inside and above the electrospray flames was acquired through high-speed visualization, shown in Fig. 8. This particular image was taken at 520 V/cm electric field with 20 mL/hr butanol flow rate. Gomez et al. \cite{2} theorized droplet fission was a mechanism of reducing droplet size, but did not provide photographic evidence of the phenomenon. In our visualization data, it was occasionally observed that one droplet would be replaced by two or three droplets slightly downstream of the first in the immediately following frame. The parent droplet would first enlarge before separating into multiple smaller droplets. It is conceivable that droplets would have some out of plane velocity, i.e. moving in and out of the laser sheet, and would produce this same effect. However, several instances of fission occurred with few other droplets around. Furthermore, the newly formed droplets were very close together, within two pixels of each other (yellow box in Fig. 8 on the next page), this supporting the assertion that multiple droplets were produced from one larger droplet and that these were not droplets entering the imaging plane. Figure
8 provided strong photographic indication of droplet fission.

Figure 8. Apparent droplet fission in a combusting e-spray

In order to check our hypothesis, we compare the charge carried by the droplet with the theoretically predicted Rayleigh limit for droplet fission [17].

\[ q^2 = 8\pi \epsilon_0 \gamma_B D^3 \]  

(5)

Where \( q \) is the surface charge density, \( \epsilon_0 = 8.8542 \times 10^{-12} \, \text{C}^2\text{m}^{-2}\text{N}^{-1} \) [18] is the permittivity of free space, \( \gamma_B \) is the liquid surface tension of butanol (23.96 \times 10^{-3} \, \text{N/m}) [19], and \( D \) is the diameter of the droplet. As the droplet evaporates, the charge density on the surface of the droplet increases until the Rayleigh limit is surpassed and the droplet breaks apart into smaller droplets. The following table lists the Rayleigh limit for typical droplet diameters found in the butanol electrosprays of this study.

<table>
<thead>
<tr>
<th>Droplet Diameter (( \mu m ))</th>
<th>Rayleigh Limit (C)</th>
<th>(C/m^3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>7.301E-14</td>
<td>139.5</td>
</tr>
<tr>
<td>20</td>
<td>2.065E-13</td>
<td>49.31</td>
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<tr>
<td>30</td>
<td>3.794E-13</td>
<td>26.84</td>
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<tr>
<td>40</td>
<td>5.841E-13</td>
<td>17.43</td>
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<tr>
<td>60</td>
<td>1.073E-12</td>
<td>9.489</td>
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<td>70</td>
<td>1.352E-12</td>
<td>7.529</td>
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<tr>
<td>80</td>
<td>1.652E-12</td>
<td>6.163</td>
</tr>
<tr>
<td>90</td>
<td>1.971E-12</td>
<td>5.165</td>
</tr>
<tr>
<td>100</td>
<td>2.309E-12</td>
<td>4.410</td>
</tr>
</tbody>
</table>

Table 2. Rayleigh limit of butanol for several droplet diameters

6 Summary and Conclusions

Reactive bio-butanol electrosprays can operate both in the the individual droplet combustion mode and group-combustion. Changing the applied electric field and fuel flow rate resulted in changing the stable position of the spray flame as well as the droplet combustion mode of the spray. In the non-reacting electrospray, an almost bi-modal droplet size distribution was observed, but during combustion, the numbers of both the largest and smallest sized droplets were reduced.

Furthermore, the comparison of droplet velocity with and without a flame present revealed a narrowing of the droplet velocity with a flame present. The differences in the droplet size PDF’s between a point source and line-of-sight measurement devices were compared on the basis of decreasing droplet spacing in the combusting e-spray. Combining the measured droplet diameter and velocity information, a simple droplet evaporation model was used to suggest that some droplets could pass through the flame region without completely vaporizing. Furthermore, images of apparent droplet fission in a combusting e-spray were justified using a Rayleigh limit based analysis.

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