



Stabilization of monodisperse electrosprays in the multi-jet mode via electric field enhancement

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Received 6 October 2004; received in revised form 4 May 2005; accepted 30 May 2005

This paper was dedicated to the 70th birthday of Professor Daniel Rosner.

Abstract

The electrospray of conducting liquids operated in the cone-jet mode is well known to have the unique ability of generating droplets uniform in size over a phenomenal range of sizes depending primarily on the liquid flow rate and physical properties. Since there is a monotonic dependence of size on flow rate, the liquid flow rates that can be dispersed are modest if the goal is to produce very small (below a few micrometers in diameter) droplets. Yet, this is precisely the application niche for which few, if any, atomization alternatives are available. Multiplexing the spray source is indispensable for the electrospray capabilities to have an impact in high-value-added applications. We report here on a novel approach to multiplexing based on a well-known, but hitherto unexploited, regime of operation, the multi-jet mode. Ordinarily, such a mode is rather unsteady and the range of flow rates at which appreciable multiplexing is achieved is small. However, if the multi-jet mode is anchored by some sharp features (e.g., grooves, ridges, etc.) machined at the outlet of the atomizer, to intensify the electric field at discrete points around its perimeter, then the cone-jets are simultaneously anchored at these features and a stable mode of operation is identified over several hundreds of volts and a broad range of flow rates. Most importantly, so long as the machining is accurately reproduced from point to point, droplets generated do not vary significantly in size from spray to spray. As a result, a compact, inexpensive and versatile multiplexing system is realized without sacrificing droplet monodispersity.

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Keywords: Electrospray; Multiplexing; Multi-jet; Monodisperse; Droplets

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1. Introduction

Electrostatic means for liquid dispersion in minute droplets are used in a variety of technological applications (Bailey, 1988). The class of atomizers in which the dispersion of the liquid is driven exclusively by electric forces is referred to heretofore as electrospray (ES). Within this class we will further restrict our attention to a particular type characterized by the additional feature of a tight control of the size distribution of the resulting aerosol. Such a system can be implemented by feeding a liquid with sufficient electric conductivity through a small opening, such as the tip of a capillary tube or a suitably treated “hole”, maintained at several kilovolts relative to a ground electrode positioned a few centimeters away. The liquid meniscus at the outlet of the capillary takes a conical shape under the action of the electric field, with a thin jet emerging from the cone tip. This jet breaks up farther downstream into a spray of fine, charged droplets. In view of the morphology of the liquid meniscus, this regime is labeled as the cone-jet mode (Cloupeau & Prunet-Foch, 1989).

Among the key features distinguishing the cone-jet electrospray from other atomization techniques are quasi-monodispersity of the droplets; Coulombic repulsion of the charged droplets, which induces spray self-dispersion, prevents droplet coalescence and enhances mixing with the oxidizer; and the use of a spray “nozzle” with a relatively large bore with respect to the size of the generated droplets, which implies that liquid line obstruction risks are minimized. The cone-jet mode can produce droplets/particles over a wide size range, from submicrometer to hundreds of micrometers, depending on liquid flow rate, applied voltage and liquid electric conductivity. Especially in the submicrometer range, the capability of producing monodisperse particles with relative ease is unique among other aerosol generation schemes. The reader is referred to previous special issues in this journal for a series of articles on this technology (Volume 25 (6), 1994; Volume 30 (7), 1999).

Electrospray ionization mass spectrometry (ESI-MS), spearheaded by the pioneering work of John B. Fenn at Yale in the 1980s (Fenn, Mann, Meng, Wong, & Whitehouse, 1989), leading to his 2002 Chemistry Nobel Prize, is the only practical application of the electrospray in widespread use. Key drawbacks that have hampered applications to other areas are the low flow rates at which the cone-jet mode can be established and the restrictions on the liquid physical properties of the liquids that can be dispersed with this technique. The difficulty is particularly severe in applications requiring that the initial droplet size be small, as for example in drug inhalation or nanoparticle synthesis. Since the electrospray exhibits a monotonic dependence of droplet size on flow rate (e.g., Chen, Pui, & Kaufman, 1995; Ganan-Calvo, Davila, & Barrero, 1997; Tang & Gomez, 1996), if one needs small droplets to generate nanoparticles, production rate may be minuscule (Gomez, Bingham, de Juan, & Tang, 1997). The smaller the desired particle size, the smaller the production rate. To provide quantities suitable for biological tests, pharmaceutical treatment or other high-value-added applications, multiplexing the spray source, is indispensable.

Multiplexing by “brute force” duplication of several capillaries operating in parallel has been investigated to a limited degree (e.g., Kyritsis, Guerrero-Arias, Roychoudhury, & Gomez, 2002; Rulison & Flagan, 1993). It may be cumbersome and impractical by more than one order of magnitude. Furthermore, compacting the sources over a small atomizer “footprint” may prove challenging. The delivery of liquid flow from a single source through a distributor with serrations was reported by Almekinders and Jones (1999), but resulted in clearly polydisperse droplet size distributions. The multiplexing goal may become easier to tackle with recent progress in the field of MEMS (micro-electro mechanical systems) (Deng, Klemic, Li, Reed, & Gomez, 2005), namely, by adapting conventional silicon integrated circuit

fabrication technology (micromachining) to the manufacturing of electrospray sources, multiplexing devices at unparalleled scales and with micrometer precision may become feasible. Even if such an approach were to materialize, cost may remain prohibitive until the advent of mass production.

We report here on a particular approach to multiplexing based on a well-known, but hitherto seldom exploited, regime of operation of the electrospray, the multi-jet mode (Cloupeau & Prunet-Foch, 1990; Zeleny, 1915). At sufficiently high voltages a liquid that can be operated in the conventional cone-jet mode may disperse into a multitude of cone-jets emanating from a single bore and typically spreading out at an angle with respect to the axis of the nozzle through which the liquid is pumped. We will describe conditions under which stable operation in this mode may lead to a compact, inexpensive multiplexing system, without sacrificing the crucial feature of monodispersity of the generated droplets.

2. Experimental system

The experimental system consisted of a syringe pump to feed and meter precise liquid flow rates through a metal tube acting as the electrospray source. A Teflon tube connected the syringe needle to the metal tube at the outlet of which the electrospray was anchored. For the reference experiments in the single cone-jet mode, two tubes were used: a larger one (1.6 mm O.D., 1.2 mm I.D.) and a capillary (1.6 mm O.D., 0.11 mm I.D.). For the multi-jet mode of operation, two large tubes were used with approximately the same dimensions (3.2 mm O.D., 1.8 mm I.D.). One of them was in brass and had its outlet machined flat and polished. The other in stainless steel was machined at one end with 12 grooves using wire electrodischarge machining (EDM) to ensure good reproducibility of the geometric features of each groove (Fig. 1). The metal tube was charged at an electrical potential of several kilovolts. At a distance of a few centimeters from the tip of the metal tube was located the flat ground electrode, consisting of a metal ring covered with a metal mesh to prevent liquid accumulation on the electrode.

The current was measured by connecting the virtual ground plate to a voltmeter of known input impedance. Visual observation of the mode of operation was made through a telescope focused on the liquid meniscus. To count the jets accurately, a He–Ne laser beam was focused into a sheet by two lenses and shone perpendicularly to the axis of the metal tube, a few millimeters downstream of the cone-jets. Scattering of the charged droplets in each spray resulted in the visualization of individual spray cross-sections appearing as small discs.

Heptane was electrosprayed with an electrical conductivity enhancer, Stadis 450 (Du Pont), at a concentration of 0.3% by weight, similarly to our earlier studies (Tang & Gomez, 1996). The selection of a liquids fuel for the testing was motivated by the need to apply the technique to the dispersion of liquid fuels in the development of a mesoscale combustor (Kyrstis, Coriton, Faure, Roychoudhury, & Gomez, 2004). This fuel had been extensively used in our laboratory in fundamental studies on the combustion of electrosprays (e.g., Chen & Gomez, 1997). Before loading the liquid in the syringe, it was sonicated for a few minutes and care was taken in the loading procedure to minimize the formation of microbubbles.

The liquid conductivity was measured by feeding the liquid through a small Teflon tube with a metal capillary at each outlet, as originally suggested by J.B. Fenn and documented by Tang (1994). By applying different potentials across the capillaries and measuring the current in the circuit, the resistance of the liquid could be inferred. From $R = l/(k \times A)$, where k is the electrical conductivity, A the cross-section and l the length of the Teflon tube, one can calculate the conductivity. The underlying assumption is that the circuit consists of two resistances in parallel, with the Teflon resistance orders of magnitudes larger



Fig. 1. Close-up picture of the grooved tube.

than the liquid. The electric conductivity of the solution was measured at 1.4×10^{-6} S/m. The other physical properties of the fluid were not affected by the additive.

Different syringe sizes were used to ensure that the plunger would be displaced at a reproducible and accurate speed. For the low flow rates, small (0.25 or 1 ml) syringes were used. Indeed, the smaller the diameter of the syringe, the more accurate and steady the motion of the syringe plunger. As a result, the measurement of the current was more accurate and the range of stable flow rates for a given mode was broadened.

Droplet sizes were measured by a phase doppler anemometer (DANTEC, Elektronik) capable of measuring simultaneously droplet size and two velocity components from the scattering of a frequency-modulated argon ion laser beam (Spectra Physics). The electrospray set-up was mounted on a multi-direction translational stage allowing for the systematic scanning of the spray by the laser probe. This probe volume was imaged on the receiver optics, which was coupled to photomultipliers for the recording of the signal and subsequent processing. A dedicated electronic processor sampled and analyzed the signal using Dantek BSA Flow Software. For each measurement, 5000 counts per sample were taken for the statistics to be representative. Measurements were performed at a given flow rate by selecting the applied voltage so that the size distribution histogram would be as monodisperse as possible. In the cone-jet mode, although the bulk of the flow rate is dispersed in uniform size droplets, a small percentage is generally dispersed as much smaller satellites, unless special care is taken to identify conditions/methods avoiding the satellite formation (e.g., Cloupeau, 1994; Hartman, Brunner, Camelot, Marijnissen, & Scarlett, 2000). These satellites are electrostatically and inertially confined to the periphery of the spray (Tang & Gomez, 1994). To ensure that the primary droplets were sized up, the laser probe volume was positioned along the axis of each spray,

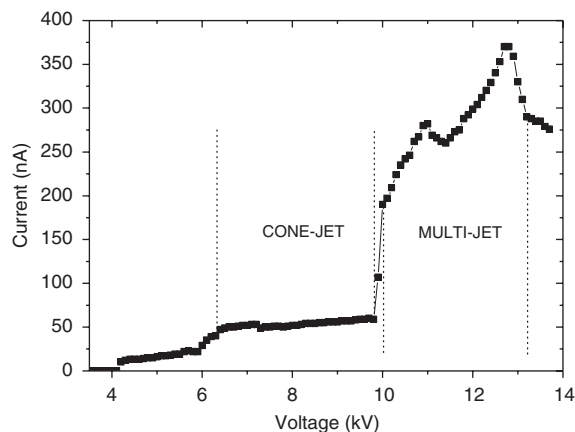


Fig. 2. Current vs. voltage for the smooth tube ($\dot{Q} = 30$ ml/h).

3. Results and discussion

3.1. Multiplexing from a single smooth tube

Fig. 2 shows a typical current versus voltage graph, obtained by electrospraying heptane in the multi-jet mode using the large brass metal tube with a polished end. The purpose of this figure is primarily to contrast the well-known behavior in the multi-jet mode to what will be found when the tip of the atomizer is suitably modified (see Fig. 4). In the lower voltage range 6.5–8.0 KV, a single cone-jet appeared, with blurred contour suggesting an inherently unstable mode of operation. As the voltage was raised in the multi-jet range of operation, few jets appeared and could be anchored stably at positions equidistant from each other. If the voltage was raised further, more cone-jets appeared. At the peak current, at about 12.5 KV, on the order of 20 cone-jets were present at the edge of the tube. The jets were not very stable. Examination of the droplets generated by phase doppler anemometry enabled us to measure the size in each jet. Much to our surprise, the average size from spray to spray was quite uniform, as shown in Fig. 3 in which the variability of the average droplet size in each spray is plotted versus the azimuthal coordinate, ϑ . Within each spray the size distribution was quasi-monodisperse, with typical size spread, defined as $(D_{0.9} - D_{0.1})/D_{0.5}$, less than 0.1. In the above relationship, D_i represents the value for which i -fraction of the liquid volume is in droplets of smaller diameters. This result suggests that the multi-jet mode consists of an array of monodisperse cone-jet electrosprays. The apparent instability of the regime is merely the result of some positional jitter of the individual cone-jets, rather than more serious instabilities that would affect the size distribution of the generated droplets. The uniformity in size would suggest that the total flow rate is equipartitioned evenly among the coexisting cone-jets, if, as in the present case, efforts are made to ensure symmetry in the geometry of the electrodes and the liquid injection. Repeating the same current/voltage experiment at 20 ml/h yielded similar results. At low flow rates, i.e. 6 ml/h, however, results were quite different. The cone-jet was never effectively stabilized, even with fewer jets. Notice also that the average flow rate per jet would still be well above the minimum flow rate that can be electrosprayed in an individual cone-jet for such a mixture (Tang & Gomez, 1996), although direct comparisons with our earlier work are difficult to make since this variable depends on the electrode configuration.

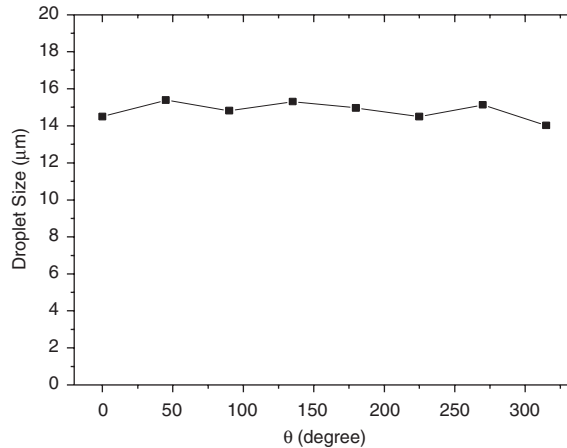


Fig. 3. Average droplet diameter in individual cone-jet electrosprays anchored in the multi-jet mode to the smooth atomizer ($\dot{Q} = 30$ ml/h; on the order of 25 jets).

The picture that emerges from these results is that the multi-jet mode is surprisingly effective at generating monodisperse droplets without the need of special manufacturing approaches. Yet, the range of flow rates over which this regime applies with an appreciable level of multiplexing of at least one order of magnitude is narrow and, even within this range, the number of jets is very sensitive to the applied voltage. This result, if generally applicable to liquids of different physical properties, poses two problems from an application viewpoint: first, the disappointing behavior at low flow rates suggest that the approach would fail precisely in the potentially most interesting regime in which the smallest droplets would be generated, which would be the one of interest in some of the high-value added applications; second, the variability of the number of jets with voltage and their positional instabilities suggest that in a “production mode” the electro spray would have to be constantly monitored and the applied voltage frequently tweaked to the design operation.

3.2. Grooved mode of operation

The observation that the cone-jet instability was positional, in the sense that conditions seem rather stable and promising from a monodispersity perspective, prompted us to attempt to anchor the multi-jet regime by designing identical geometric features at the atomizer outlet in a symmetric pattern, for the purpose of stabilizing the cone-jets at particular locations around the opening circumference. To that end, a number of grooves were machined on the tube face by wire electric discharge machining, as shown in Fig. 1. The grooves would anchor the multiple jets, once a sufficiently high voltage was applied with respect to a ground electrode, by virtue of the small radius of curvature and more intense electric field present at these locations. In that respect the shape of the indentation in the surface should not be critical.

Solutions of Laplace equation for the electric potential between the two electrodes were carried out with different spatial resolution using FEMlab (COMSOL Inc., 2005) for a simplified nozzle geometry with only four grooves. The field, computed at the outermost radial location, revealed the presence of spikes, corresponding the location of the grooves, where the field intensity was at least a factor of two larger than the background value along the rest of the circumference. Since no account was taken of

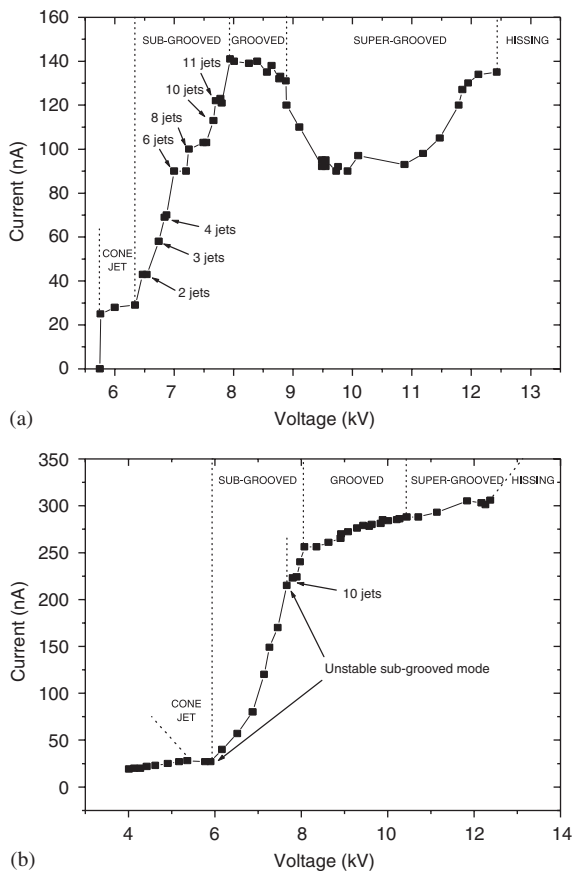


Fig. 4. Current vs. voltage for the grooved tube operated at $\dot{Q} = 6$ ml/h (a) and $\dot{Q} = 30$ ml/h (b).

the presence of the liquid that was wetting the surface nor of the charged droplets and attending space charge, the results are to be considered qualitative. Nevertheless, they are revealing since they show a periodic electric field along the circumference of the nozzle, with peaks in correspondence of the grooves that are dramatically larger than the background field at the nozzle, as intuitively anticipated from first principles. As shown below, under certain conditions, these spikes in field strength at the grooves may be sufficient to lock otherwise unstable cone-jets in place. Furthermore, the multi-jet regime can be reached at an operating voltage lower than in the case of smooth nozzles, thereby minimizing the risk of electric discharge.

Fig. 4 shows typical current versus voltage graphs, obtained for the same liquid at two flow rates, 6 ml/h (Fig. 4a) and 30 ml/h (Fig. 4b). Fig. 5 shows several pictures that were obtained under different conditions, as elaborated below. In Fig. 4a, one can notice the more or less monotonic increase of the current in a step-ladder pattern, as the spray transitioned from a well-pronounced initial plateau, corresponding to the single cone-jet (Fig. 5a), to a multi-jet regime with anywhere from 2 to 11 jets. Figs. 5b and c show the morphology in some of the initial phase of this transition, with the appearance of three and four equidistant jets along the tube circumference. This regime in which the number of jets is smaller than

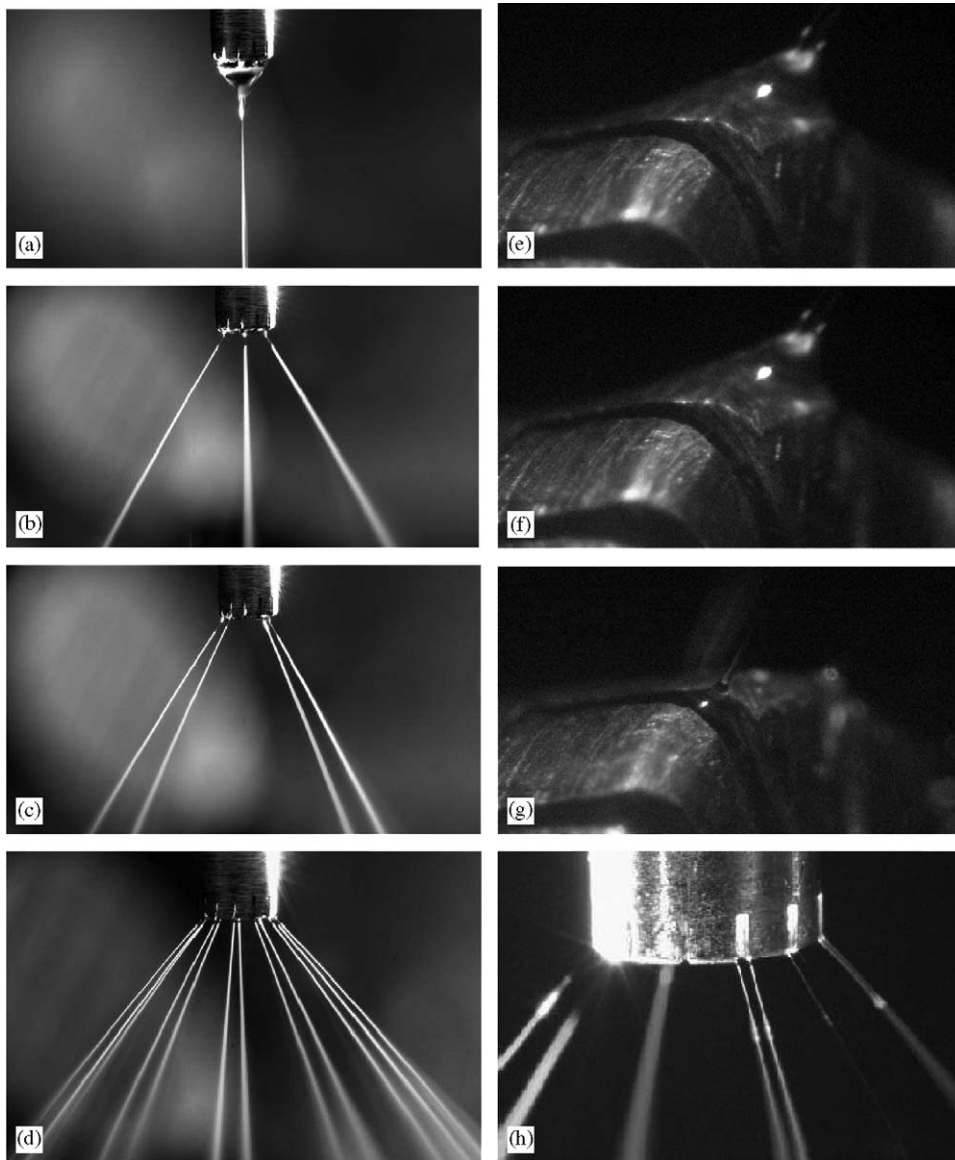


Fig. 5. Pictures of various modes of operation, with close-up views of the voltage effect in the grooved mode.

the number of grooves in the atomizer tip is labeled sub-grooved mode. Next, as the voltage was further raised, a new plateau was reached, spanning several hundred volts, within which the grooved regime was established, with as many jets as there were grooves (Fig. 5d). The current was nearly constant in this mode and, as the voltage was raised, each cone-jet shrunk. The cone-jet appearance is documented in the sequence of Figs. 5e–g and is consistent with observations made in the single cone-jet mode, reporting shrinkage of the cone as the voltage rises (Hayati, Bailey, & Tadros, 1987). The anchoring of

the cone-jet may occur either at the top of the groove or directly inside the groove. Beyond 9 KV, the current surprisingly dipped in correspondence of a collapse of the grooved mode: some jets were, in fact, disappearing and leaving their groove empty, while others appeared in grooves that had already been occupied. As the current began to rise again, more and more jets formed and several but not all of the grooves anchored two jets. Fig. 5h shows one such an example. Finally, at the highest voltage, a hissing sound became audible, which was a prelude to corona discharge and unstable spray disruption. Beyond this limit, the cone-jets were not stable but appeared to vibrate in the grooves.

Similar graphs were obtained at higher flow rates, as in Fig. 4b corresponding to 30 ml/h. The trends are similar to the lower flow rate case with some noteworthy differences:

- (a) In the transition from single cone-jet to grooved mode, the current increase showed less of the staircase pattern that had been observed at 6 ml/h and becomes strictly monotonic.
- (b) In this transition, it was difficult to establish a subgrooved regime with few jets.
- (c) The current in the grooved mode increased, rather than being constant as the flow rate increased.
- (d) The dip in current, past the grooved regime, became less and less noticeable as the flow rate increased and the flow rate transitioned to a condition in which the flow rate effluxing through a single groove was split into two stable jets, resulting in a total of 24 jets. While this regime is potentially the most desirable in terms of multiplexing, it did not appear to be as stable as the grooved mode, although its stability improved as the flow rate increased.
- (e) Eventually, also at larger flow rates, the hissing regime was reached.

A comparison of the system behavior of the grooved mode versus the results obtained in Fig. 2 for a smooth nozzle suggests that the stability of the system improved significantly, which allowed us to explore features of this electric field-anchored multi-jet mode in greater detail, as elaborated in the remainder of the article.

3.3. Current and stability domain

To relate the behavior of the multi-jet regime to the better known single cone-jet mode, the average current per jet was compared to that measured in a separate experiment with the smaller tubes operated in the single cone-jet regime. In other words, with the electrospray in either the sub-grooved mode or grooved mode, that is, with up to 12 jets, the total current was measured and the number of jets were counted, with the ratio of the values yielding the average current per jet. Results are plotted in Fig. 6. It appears that the average current per jet obeys, within the experimental scatter, the same power law as in the single cone-jet mode. As a result, the current power law for the multi-jet must be close to that of the single jet. Although, considerable work has been made in deriving scaling laws for different liquids that are largely based on dimensional analysis (Fernandez de la Mora & Loscertales, 1994; Ganán-Calvo et al., 1997), in the present case we will refrain from applying such laws since they are not well established for the low conductivity liquid used in these experiments and no comparisons with other fluids will be made in this study. Thus, it suffices to compare the relative behavior of the multi-jet mode versus the single cone-jet counterpart. Part of the scatter may be due to small variations in current that depends on the number of jets, with the grooved mode resulting in the largest current per jet. The average current per jet scales with the average flow rate per jet as $I/n \sim (Q/n)^{0.36}$, which is essentially the same, within the experimental scatter, as $I \sim Q^{0.35}$ obtained for the single cone-jet mode. This finding can also help us

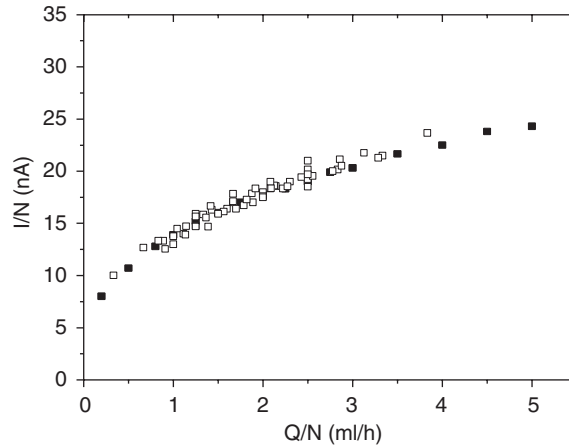


Fig. 6. Current versus flow rate for a multi-jet of heptane + 0.3% of Stadis (open symbols: average values for multi-jet; full symbols: single cone-jet).

explain the results of Fig. 4 on the current increase as more and more jets appear. In fact,

$$I = \sum_1^n I_i \propto \sum_1^n \dot{Q}_i^\alpha = \sum_1^n \left(\frac{\dot{Q}}{n} \right)^\alpha = n^{(1-\alpha)} \dot{Q}^\alpha,$$

where α is the exponent of the power law, n is the number of jets, \dot{Q} is the total liquid flow rate and \dot{Q}_i is the flow rate through the i -cone-jet. The second equality relies implicitly on the assumption that the flow rate is uniformly distributed among the n cone-jets, as will be confirmed below. At constant total flow rate, \dot{Q} , the current increases as n increases, consistently with the findings in Fig. 4. The dip at low flow rates and high voltage in Fig. 4a suggests that the system must be operating in a rather unstable mode, with probable loss of monodispersity. Within the grooved mode, the reasonably constant current in Fig. 4a is also consistent with the equation above. At higher flow rates, on the other hand as in Fig. 4b, as the voltage is increased in the grooved mode, the current increases by 10% or so.

More generally, the implication of the above equation is that if the application for which the electrospray is used requires the highest possible charging of the liquid, as for example in ESI-MS, the multi-jet mode will yield a current gain by a factor $n^{1-\alpha}$ in the current passed through the spray, where α is invariably less than unity (even in the case of polar liquid for which the value of 0.5 is well established (Fernandez de la Mora & Loscertales, 1994)).

In order to minimize the scatter in the comparison of the single cone-jet with the multi-jet mode, it was decided to focus subsequent measurements only on the grooved regime, yielding the highest level of stable multiplexing. Indeed, this mode appears to be better defined, as the curves current–voltage for various flow rates showed in Fig. 4. A voltage–flow rate stability domain, which shows the operational domain of this regime, is shown in Fig. 7. The lowest curve is the onset value of voltage for which the 12 jets are well anchored in the grooves, the intermediate line corresponds to the onset voltage of the supergrooved regime, with more than 12 jets appearing, the upper curve corresponds to the hissing mode, a regime beyond which corona discharge will occur. This stability domain confirms that the range of operating voltages for the grooved mode is relatively large on the order of 1–2 KV, which is sufficiently

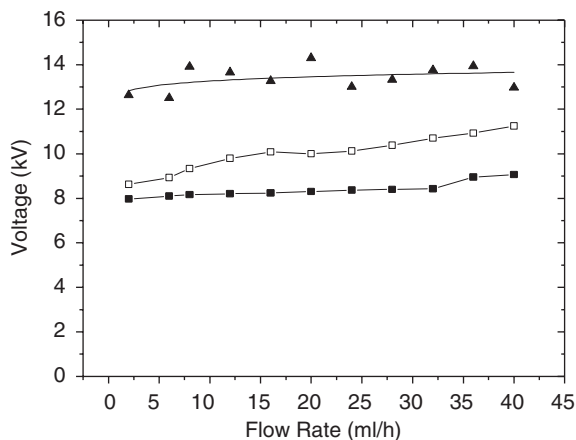


Fig. 7. Stability domain in the voltage/flow-rate plane for the grooved multi-jet mode (full squares: voltage onset; open squares: upper voltage of grooved mode; full triangles: hissing mode).

broad to ensure easy establishment of this regime in an experiment. Similar behavior was observed with other hydrocarbons such as JP-8 of interest in the development of mesoscale combustors (Kyritsis et al., 2004). This result should be contrasted with the far more unstable behavior of the system with a smooth nozzle, as discussed in connection with Fig. 2.

3.4. Droplet size measurements

The comparison of current power laws in the single cone-jet mode and multi-jet modes suggests that each cone-jet in the multi-jet regime behaves in the same way as an isolated cone-jet. Confirmation of this observation is now sought with respect to the monodispersity of the generated droplets and their uniformity from jet to jet. Since the grooves are symmetrically and uniformly disposed, except for machining imperfection or asymmetries in the liquid pumping system, there is no reason to expect that the flow rate be not equipartitioned among the multiple cone-jets, which should lead to uniformity throughout the spray cloud. Confirmation of this supposition with respect to the uniformity of the jets, and hence that of the flow rate, can be offered by the measurements of the size of the droplet emerging from the jet, thanks to the monotonic relation between the flow rate and the droplet size as shown for the single cone-jet (Tang & Gomez, 1996). Fig. 8 presents the average droplet size for three flow rates, 6 ml/h, 12 ml/h and 18 ml/h, respectively. To quantify the size scatter from jet to jet, a relative standard deviation (RSD) is defined as

$$\text{RSD} = \frac{1}{\bar{D}} \sqrt{\frac{\sum_{i=1}^N (\bar{D}_i - \bar{D})^2}{n-1}},$$

where \bar{D}_i is the average droplet diameter for jet i , and $\bar{D} = \sum \bar{D}_i / n$ is the droplet size averaged over all jets. The uniformity of the droplet size is good, with the RSD < 10%, which is comparable to the degree of non-uniformity in size within a single jet. The relative standard deviation was found to be significantly

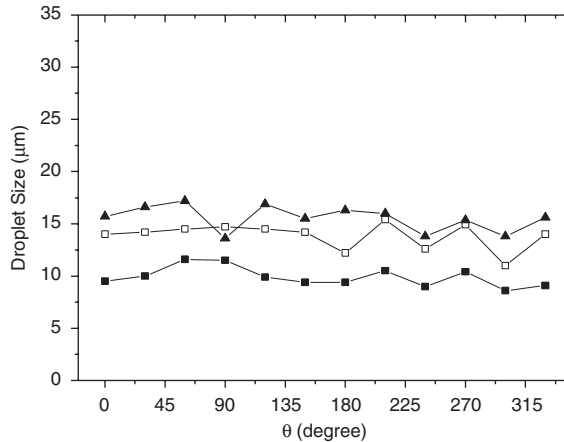


Fig. 8. Average droplet diameter at different flow rates in individual cone-jet electrospays anchored in the grooved mode to the grooved atomizer (full squares: $\dot{Q} = 6$ ml/h; open squares: $\dot{Q} = 12$ ml/h; full triangles: $\dot{Q} = 18$ ml/h).

larger in the sub-grooved mode, which is another indirect confirmation that flow equipartition is improved in the grooved mode.

The present approach to multiplexing has some similarity with the dispersion through a serrated opening by [Almekinders and Jones \(1999\)](#). No details are given in that article of the mechanism at work and whether the serration was used for a electric field-based stabilization effect or as a means to distributing the flow rate at the outlet. Furthermore, the reported size distributions are significantly polydisperse, which implies that our primary objective of preserving the uniformity of droplets size from jet to jet was not fulfilled in their study. We conclude that the similarity is not substantive and the approach described here must be significantly different from theirs. Similarities may exist with approaches involving a variety of electrodes with peculiar geometries that are discussed in the patent literature but were never documented quantitatively (e.g., [Coffee, Noakes, Bancroft, & Bals, 1987](#); [Colclough & Noakes, 1987](#); [Escallon & Tyner, 1987](#)).

In [Fig. 9](#) the average droplet diameter in the grooved mode has been plotted versus the average flow rate \dot{Q}/n and compared with data obtained in the single cone-jet mode for which $n=1$. The results for the multi-jet regime appear to be consistent with the size versus flow rate curve of the single jet. However, the scatter of the points plotted for the grooved mode is non-negligible, e.g. 15% at 18 ml/h. The discrepancy seems to increase with the flow rate, which may be attributed to the fact that the range of voltage within which the grooved mode is operated becomes larger for large flow rates. As a consequence of the measurement technique, the droplet size might have been measured for a voltage somewhat higher than the onset value. For a given flow rate, the droplet size decreases when the voltage increases, particularly at large flow rate ([Tang & Gomez, 1996](#)).

3.5. Groove geometry, size uniformity and machining accuracy

We assessed the influence of geometric parameters on the electrospay behavior, including number of grooves, groove width and depth. The conclusion from these tests was that the first two parameters are the most important. Depending on how wide is the groove either one or two cone-jets per groove can

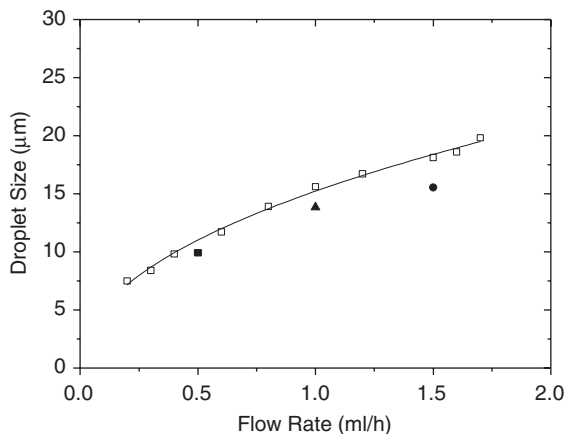


Fig. 9. Comparison of droplet size dependence on flow rate. Open squares: droplet diameter versus flow rate for single cone-jet. Full triangles: average droplet diameter versus average flow rate per cone-jet in the multi-jet regime.

be stabilized. Clearly, there is a tradeoff between widening the groove and fitting more of them on the annular surface at the outlet of a tube.

A detailed examination of the size variation from jet to jet in similarly grooved atomizers that had been machined with a small blade, less reproducibly than with wire EDM, showed some pattern of the average size with jet position which was repeated for almost all the measurements and seemed to correlate with fabrication defects. This observation raised the concern that non-uniformities may stem from machining defect at the edge of the grooves. Whether the uniformity can be further improved with more precise machining remains to be seen. Certainly, conventional machining with spatial resolution better than wire EDM is difficult to achieve. Probably, the only approach is via microfabrication. We can test this hypothesis by checking if a deliberately introduced defect significantly worsens the non-uniformity from jet to jet. To that end, a grooved atomizer was damaged in two positions, as highlighted by the circled areas in Fig. 10.

Size measurements obtained with such a tube are shown in Fig. 11. The grooved mode was difficult to establish. A 12-jet pattern was actually obtained in a super-grooved mode, with one empty groove and another exhibiting a double jet. A 10-jet subgrooved mode looked more stable, with no time dependent behavior of the jets. However, the non-uniformity in average size is dramatically worsened, as compared to Fig. 8. For each flow rate, the curves present a peak, which corresponds to two neighboring jets of relatively large droplet size and hence high flow rate. These two jets were easy to localize on the edge of the tube because their ligaments were much longer than the others, which is consistent with a high flow rate through each jet. This double peak shifted in azimuthal position depending on the flow rate, as highlighted by the dashed ellipse in the figure. This can be explained by the fact that the spray was not operated in the grooved mode. For example, one extra jet had appeared in a slot where a jet was already anchored, at the beginning of super grooved mode, but the physical position around the edge of the tube remained the same regardless of flow rate. Moreover, as we can see in Fig. 12, it appears that those two jets with larger flow rates were not anchored inside the slots, unlike the regular jets in grooved mode, but on the tip, whereas the neighbor slots were empty. The labeled jets in the figure correspond to the peak sizes within the elliptic region in Fig. 11.

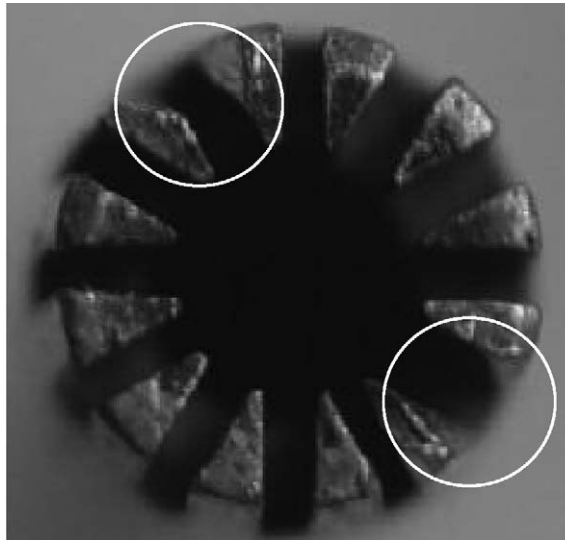


Fig. 10. Grooved atomizer with deliberately induced fabrication damages in two circled areas.

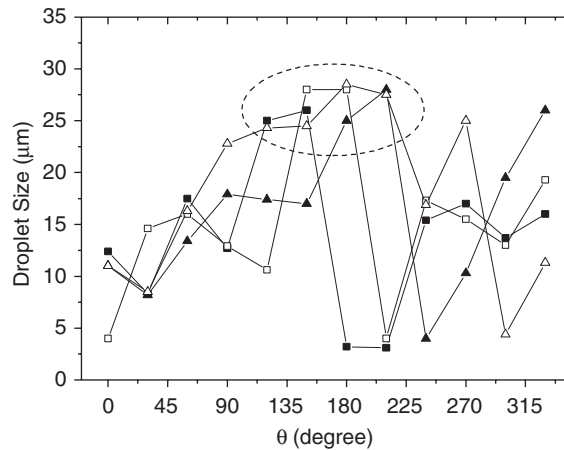


Fig. 11. Average droplet diameter at different total flow rates in cone-jet electrospays simultaneously anchored to the damaged grooved atomizer.

3.6. Further multiplexing

Conventional machining with feature size smaller than 0.1 mm is not achievable. Examining Fig. 1 one can see that in principle twice as many grooves, that is, a total of 24, can be fitted on this particular atomizer. If the goal is to achieve an even higher level of multiplexing, the only avenue, short of microfabricating atomizer “tips”, would be to multiplex grooved nozzles by brute force multiplication of the geometry in Fig. 1. Thus, we located three grooved nozzles at the vertices of an equilateral triangle and observed the

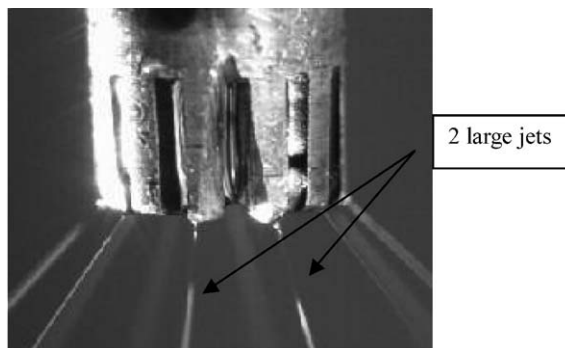


Fig. 12. Close-up views of asymmetric behavior in the damaged grooved atomizer.



Fig. 13. Picture of multiplexed grooved atomizers.

behavior of a fourth nozzle positioned in the center of the triangle. We machined a reservoir that would distribute the liquid to the four nozzles, through plastic tubing. The plastic tubing allowed us to test the nozzles at distances ranging from 5 to 9 mm from the central nozzle. Because the multiplexed nozzles required a stronger electric field, we moved the ground plate up so that the nozzles were approximately 1 cm away from the ground. For any configuration, Coulombic repulsion of positively charged jets resulted in strong repulsion of cone-jets as they form. As we increased the voltage, the first cone jets would form on the outer radii of the three outer nozzles. They would have several cone-jets working before the central nozzle would establish its first few jets. By the time the central nozzle had developed a few of its 12 cone-jets, the potential difference had reached a level at which sparks connect from the nozzles to the ground plate. The only geometric arrangement that allowed us to establish four stably working nozzles (see Fig. 13) was with the 9 mm spacing, since the larger distance reduced the effects of Coulombic repulsion. Notice that the footprint of such an atomizer would be on the order of 94 mm^2 and would yield the generation of 48 cone-jets. As a result the multiplexing density is a respectable 50 cone-jets per cm^2 , that is, a factor of three smaller than the value obtained for a single grooved atomizer and a factor of five smaller than what reported with a more sophisticated MEMS-based approach (Deng et al., 2005). Of course, since this behavior is dependent on space charge effects, results may differ with liquids of different physical properties. Further studies will be conducted on various liquids to generalize the behavior of the grooved atomizers.

4. Conclusions

An experimental investigation was performed on electrified menisci simultaneously anchored to a single atomizer in the so-called multi-jet mode. When the cone-jets were anchored to a smooth metal tubes, the

regime was found to be unstable in terms of the number of cone-jets that could be anchored and their positions around the tube circumference. Yet, the size distribution was surprisingly monodisperse, which suggests that the instability is inconvenient from an operational viewpoint but is not severe enough to disrupt the cone-jet behavior. Furthermore, the range of flow rates within which appreciable multiplexing was achieved, say, by one order of magnitude, was very narrow.

In contrast, when sharp features were machined on the face of the tube, the electric field was locally raised and dramatically enhanced the stability of the multi-jet mode. As a result, a comparison of the behavior of the multi-jet mode with the better known single cone-jet mode could be carried out systematically. Each cone-jet was found to obey, on average, the same power laws that had been observed for single cone-jet mode, both in terms of current and droplet size dependence on flow rate.

If care is taken to ensure a symmetric and precise machining of the tip of the atomizer, the flow rate is equipartitioned among the many jets, which results in uniform size from jet to jet. Furthermore, this mode of operation could be stabilized over a broad range of liquid flow rates. By deliberately damaging some of the grooves of the atomizer and monitoring the effect on droplet size, it was ascertained that machining defects at the edge of the grooves are responsible for non-uniformities in the spray.

The grooved atomizer was found to be a simple and easily controllable means of multiplexing an electrospray by one or two orders of magnitude, yielding a density on the order of 50 cone-jets per cm², at least for liquids with modest electric conductivity (1.4×10^{-6} S/m).

Acknowledgements

We would like to acknowledge the help of Mr. Nick Bernardo, from the engineering machine shop at Yale, who manufactured most essential pieces. The support of the DARPA Palm Power Program under Grant no. DAAD19-01-1-0664 (Dr. Richard J. Paur, ARO, Contract Monitor) is gratefully acknowledged.

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