Electrostatic Spraying of Nonconductive Fluids into Conductive Fluids

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Electrostatic spraying is important in many applications where very fine droplets are desirable. Most electrostatic spraying systems developed to date, however, require that the electrical conductivity of the dispersed fluid is higher than that of the surrounding fluid. This work reports on an experimental investigation of the mechanism for successful electrostatic spraying of a nonconductive fluid into a conductive one. The key role played by the electric stress on the interface between the nonconductive and conductive fluids is evidenced by examining the variations of the emitted drop size, electrical current, and pressure inside the nozzle as functions of the applied voltage, nozzle geometry and distance between the high-voltage nozzle-electrode and the grounded electrode immersed in the surrounding fluid. A comparison of nonconductive-in-conductive and conductive-in-nonconductive spraying systems reveals a difference in behavior that is consistent with the theory of electrohydrodynamics.

The use of electrostatic fields to produce fine sprays of liquid droplets from nozzles dates back to as early as 1750 when Abbe Nollet, a professor of physics at Turin and Paris, conducted experiments with an electrified nozzle that was attached to the outlet of a water vessel (Felici, 1959). With the continuation of scientific studies over the years, electrostatic spraying has found many important applications, such as the production of ultrafine powders, electrostatic printing, paint spraying, and crop spraying (Bailey, 1984). Recently developed applications of electrostatic spraying in chemical processing have included the emulsion-phase contactor, which is an electrically-driven solvent extraction device (Scott and Wham, 1989), and the electric dispersion reactor, which is a multiphase reactor capable of efficient production of microreactor droplets (Harris et al., 1993). Based on the experience gained from numerous studies, most investigators to date have concluded that electrostatic spraying can be successfully applied only for dispersing relatively high-conductivity fluids into low-conductivity fluids (Bailey, 1986). Electrostatic spraying of nonconductive fluids into conductive fluids has been considered virtually impossible until recent experiments by Sato and coworkers (1979, 1980a,b, 1993). Despite its scientific interest and technological importance, the phenomenon reported by Sato and coworkers remains heretofore inadequately understood. Also, unlike spraying conductive fluids, the nozzle design used by Sato et al. (1993) for spraying nonconductive fluids is rather complicated in construction and, due to lack of complete understanding of the phenomenon, is not guided by fundamental physical principles. Accordingly, in this article we elucidate the physical mechanism of the phenomenon uncovered by Sato and coworkers by means of carefully designed experiments performed with two spraying systems of contrast: a nonconductive fluid dispersed into a conductive fluid (N-in-C) and vice versa (C-in-N). Here, whether a fluid is regarded as conductive or not is defined in terms of its time scale of electrical charge relaxation compared with the time scale of spraying.

Electric Stress on Fluid Interface

Based on the theory of electrohydrodynamics, the key to electrostatic spraying is the electric stress on the fluid-fluid interface. A strong enough electric stress can render the interface unstable and, thus, can result in the emission of fine droplets or bubbles (cf. Lord Rayleigh, 1882; Taylor, 1964). The electric stress tensor, $\underline{\underline{T}}_e$, in a linearly polarizable fluid can be written as (see, for example, Landau and Lifshitz, 1960; Melcher, 1981):

$$\underline{\underline{T}}_e = \epsilon \underline{E}\,\underline{E} - \frac{1}{2}\epsilon E^2 \underline{\underline{I}} \tag{1}$$

where $\underline{\underline{I}}$ is the unit tensor, ϵ is the permittivity, and $\underline{E}$ is the electric field whose strength is E. The electric stress on a fluid interface results from the discontinuity of the electrostatic field across the interface. For example, at an interface between a highly conductive (c) and a nonconductive (i) fluid, the net electric stress is given by:

$$\underline{\underline{n}} \cdot [\underline{\underline{T}}_e^{(i)} - \underline{\underline{T}}_e^{(c)}] = \frac{1}{2}\epsilon^{(i)} E^{(i)2} \underline{\underline{n}} \tag{2}$$

where $\underline{\underline{n}}$ is a unit vector normal to the interface pointing from the conductive fluid to the nonconductive fluid. Equation 2

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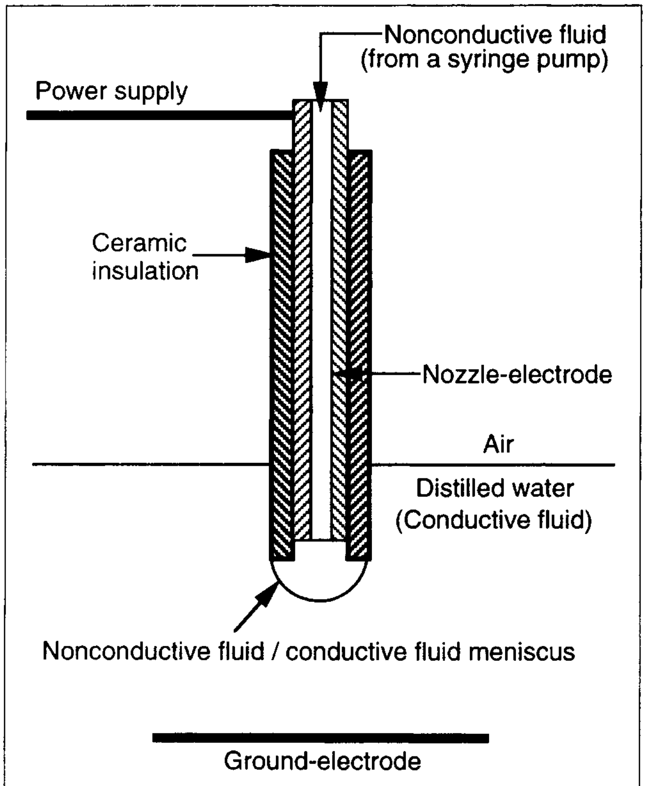

Figure 1. Nozzle assembly for electrostatic spraying of nonconductive fluids into conductive fluids.

During the spraying operation, nozzle tip is immersed in conductive fluid. Nonconductive fluid is delivered by syringe pump through metal capillary at constant flow rate, and forms a meniscus at nozzle tip, which undergoes periodic changes. The meniscus grows with time while attached to nozzle tip, then breaks into a droplet, and phenomenon is repeated with a new growing meniscus.

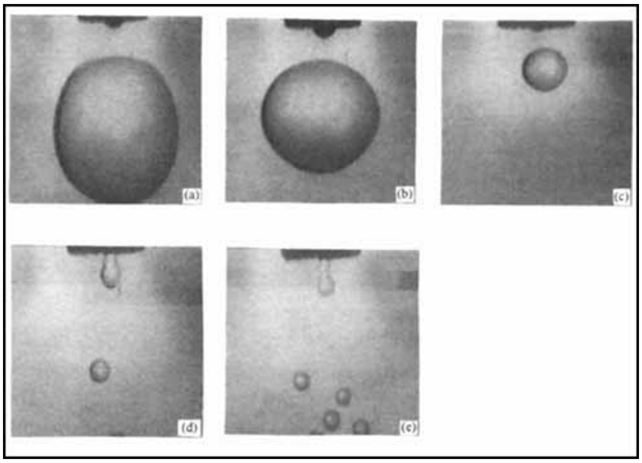

Figure 2. TCE drops formed in distilled water at tip of nozzle in Figure 1.

Inside diameter of the metal capillary is 0.5 mm, outside diameter is 1.6 mm, and outside diameter of ceramic insulation tube is 3.2 mm. Flow rate of TCE in this case is 0.5 mL/min. Photograph sequence corresponds to (a) 0 V, (b) 200 V, (c) 400 V, (d) 600 V, and (e) 800 V. (The conductivity of nonconductive chemicals used in this study ranges from 8×10^{-9} (TCE) to 10^{-16} (hexane) mho/m. The conductivity of distilled water is 2.5×10^{-4} mho/m.)

follows from the fact that the electric field strength becomes zero inside the highly conductive fluid. Thus, at the interface, the electric force acts perpendicularly and in the direction from the conductive fluid to the nonconductive fluid. A strong electrostatic field on the interface is ensured by maintaining a large electrical potential difference between the interface and a nearby electrode. The presence of a strong electrostatic field usually implies that there are no effective conductivity paths that allow substantial flow of electrical current. This requirement is easily satisfied when a conductive fluid is sprayed into a nonconductive fluid. In this case, the conductive fluid inside the nozzle is in direct contact with one of the two electrodes. The other electrode is placed at an appropriate distance from the nozzle tip and is well insulated by the surrounding continuous phase. When a potential difference is imposed between the two electrodes, the electrostatic field that is present in the nonconductive phase exerts an outward force on the surface of the conductive drop hanging from the nozzle tip, according to Eq. 2. This force is responsible for breaking up the interface to produce fine sprays (Bailey, 1984, 1986).

Spraying of Nonconductive Fluids into Conductive Fluids

When a nonconductive fluid is ejected from a capillary nozzle into a conductive fluid, it would be very difficult to create electrostatic fields of significant strength in the continuous phase which exhibits a tendency to establish an equipotential state due to its high conductivity. However, it is possible to create an electrostatic field in the nonconductive fluid that is located between the conductive fluid and an adequately insulated electrode of different potential. This is achieved here with a nozzle assembly that consists of a metal capillary sheathed in a ceramic insulation tube, as shown in Figure 1. This kind of nozzle design allows easy adjustment of the distance between the end of the metal capillary and that of the ceramic tube, in contrast to the nozzle used by Sato et al. (1993). The metal capillary serves as one electrode, called the nozzle-electrode, and is connected to a high-voltage power supply. The other electrode, designated as ground-electrode, is immersed inside the surrounding continuous fluid and is connected to electrical ground. When emerging from the nozzle tip, the nonconductive fluid forms a meniscus that insulates the metal capillary from the surrounding conductive fluid. Thus, a strong enough electric stress for electrostatic spraying can be generated at the meniscus, if the end of the metal capillary is not located too far away from the meniscus. Experiments with various flow rates of the nonconductive fluids, dimensions of the nozzle-electrode and insulation tubes provided qualitatively comparable results. The general performance of the system is shown in Figure 2 in a series of photographs of trichloroethylene sprayed into distilled water (TCE-in-water system) at various values of the applied voltage. Clearly, the size of the droplets issuing from the nozzle decreases as the applied voltage increases which is consistent with the results reported by Sato et al. (1993).

Experimental Results and Discussion

The foregoing experiment verifies that not only is electrostatic spraying possible with C-in-N systems, as widely recognized, but also with N-in-C systems as well. There are,

Table 1. Effect of Ground-Nozzle Distance on Drop Size and Current

App. Voltage (V)	Drop. Dia. (mm) Ground-Nozzle Distance		Current (mA) Ground-Nozzle Distance	
	50 mm	25 mm	50 mm	25 mm
0	2.74	2.80	0	0
200	2.35	2.50	0.062	0.068
400	1.81	1.59	0.133	0.146
600	0.77	0.65	0.217	0.233
800	0.49	0.53	0.298	0.299

Table 2. Effect of Nozzle Insulation on Electrostatic Spraying

App. Voltage	Drop Diameter (mm)			Current (mA)		
(V)	Case 1	Case 2	Case 3	Case 1	Case 2	Case 3
0	2.74	2.74	2.74	0	0	0
200	1.59	2.35	2.69	0.029	0.062	0.202
400	0.93	1.81	2.44	0.092	0.133	0.294

Case 1: Electrode tip is 1 mm inside the ceramic tip.
Case 2: Electrode tip is even with the ceramic tip.
Case 3: Electrode tip extends 1 mm outside the ceramic tip.

however, significant differences between the two systems. In the case of C-in-N systems, both the location and shape of the electrode immersed in the surrounding phase are important, because they are the main factors that determine the electrostatic field strength and distribution, as evidenced by previous calculations (see, for example, Basaran and Scriven, 1990; Harris et al., 1993; Harris and Basaran, 1993). For a given applied voltage, the closer the ground-electrode is located to the nozzle-electrode, the stronger the electrostatic field becomes, and, therefore, finer sprays are produced. In the N-in-C system, however, neither the location nor the shape of the electrode in the surrounding phase should significantly affect the spraying behavior, because the high conductivity of the surrounding fluid maintains every part at virtually the same potential or voltage as that imposed at the electrode connected to it. This fact is made evident in Table 1 by the virtually identical drop sizes and currents for different distances between the nozzle-electrode and the ground-electrode in the TCE-in-water system.

Whereas the geometric configuration of the nozzle-electrode has virtually no effect on the C-in-N system, it plays a central role in the spraying behavior of the N-in-C system. For example, as shown in Table 2, both the size of the droplets that issue from the nozzle and the amount of electrical current in the TCE-in-water system are significantly influenced by the distance between the end of the metal capillary and that of the ceramic tube. Furthermore, data in Table 2 also show that droplets of comparable size can be obtained at drastically different values of electrical current. Specifically, the drop size decreases and the electrical current increases as applied voltage increases in each case of fixed geometric configuration (columns), but a comparison of all three geometric configurations at a given voltage shows that the drop size increases as electrical current increases (rows). Thus, the amount of electrical current is not the determining factor of the size of the drops produced by electrostatic spraying. This observation accords with physics, because it is the electrostatic field strength rather than the amount of current that determines the electric stress (see, for example, Eqs. 1 and 2). Hence, from the consideration of energy efficiency, the optimal nozzle design should minimize the electrical current flow while being able to maintain a strong electrostatic field at the nozzle meniscus. Our results in Table 2 indicate that the case in which the end of the metal capillary is located one mm inside the ceramic tube gives the best performance. This stands in contrast to the nozzle used by Sato et al. (1993), which has a metal capillary protruding one mm outside the end of the glass insulation and is thus expected to produce a significant amount of electrical current.

When examining the effects of electric force from Eq. 2, one recognizes that, for the N-in-C system, the force arising from the electrostatic field acts inward at the nozzle meniscus, in contrast to the C-in-N system that has an outward force. This inward force is expected to give rise to a general increase of pressure inside the nozzle, as is verified by pressure measurements for the TCE-in-water system in comparison with that for the C-in-N systems as shown in Figure 3.

The photographs in Figure 4 show interesting details of drop breakup and spraying at the nozzle for both the N-in-C and the inverse system. For the N-in-C case (a), the breakup is due to the pinch-off action near the location where the meniscus meets the outer wall of the capillary, because the inward electric

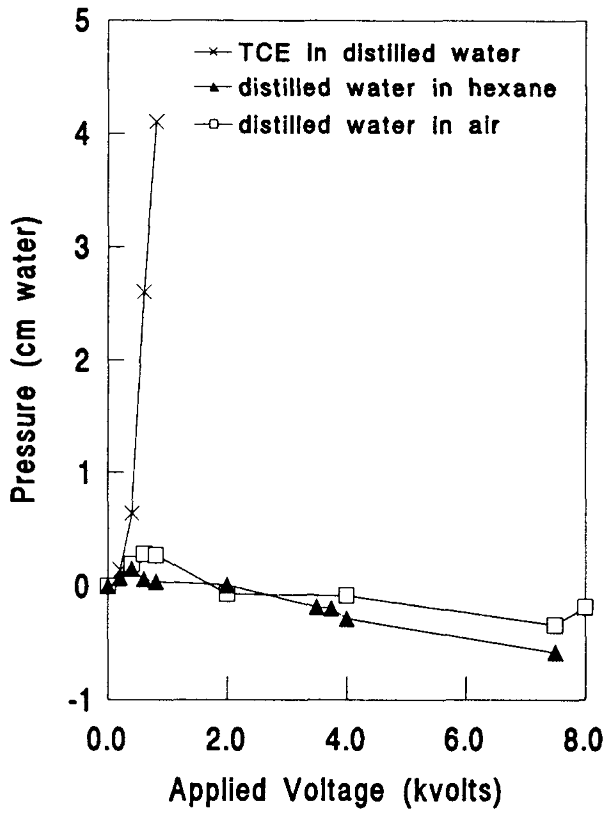

Figure 3. Variation of fluid pressure inside nozzle measured 20 cm upstream of tip of ceramic tube as function of the applied voltage.

Results for TCE in distilled water (nonconductive-in-conductive) and distilled water in hexane and air (conductive-in-nonconductive). Pressure measurements are obtained by a transducer connected to a data acquisition system, which consists of an analog to digital converter and a personal computer. The data shown in this figure are values averaged over a period of 100 s.

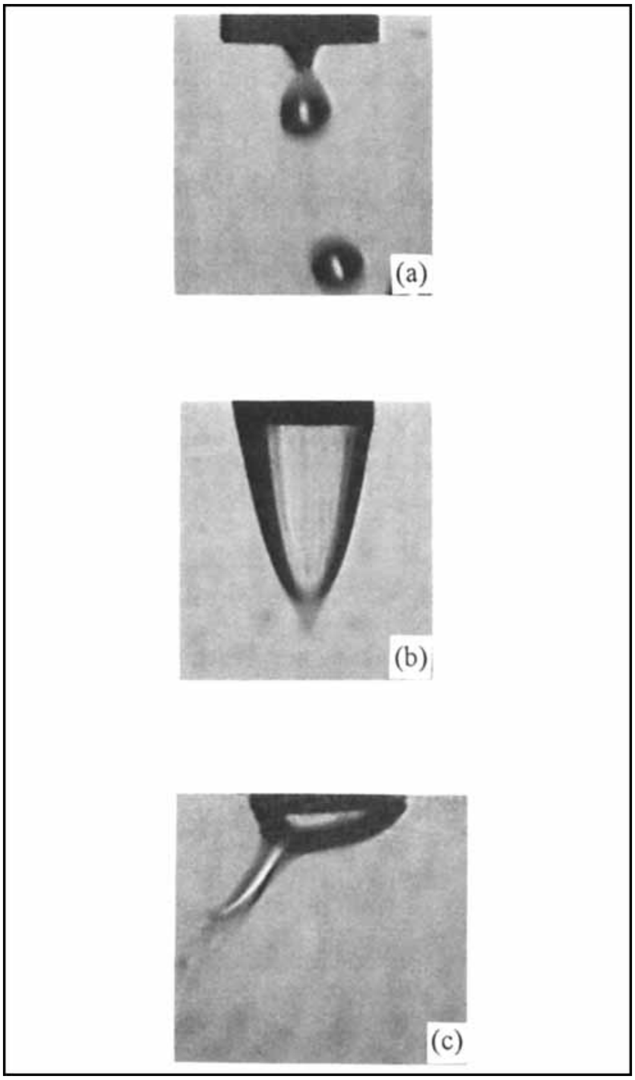

Figure 4. Drop breakup in the system of (a) TCE in distilled water (nonconductive-in-conductive), (b) distilled water in air (conductive-in-nonconductive) at relatively low electric field, and (c) distilled water in air (conductive-in-nonconductive) at relatively high electric field.

Nozzle outside diameter is 1.6 mm.

force is strongest in the region where the interface is closest to the end of the metal capillary. For the C-in-N case (b), however, the droplets are produced at the elongated conical tip of the meniscus as a result of the strongest outward electrical force in that region. When the electric field strength is high enough, the elongated meniscus, instead of pointing toward the other electrode, loses its axial symmetry (c). In this case, tiny drops are emitted from the highly pointed part of the meniscus that jumps from side to side, in accordance with earlier observations (Zeleny, 1915; Cloupeau and Prunet-Foch, 1989).

The basic mechanism of electrostatic spraying in the N-in-C system is made clear by the analysis of the electric stress at the fluid-fluid interface and supporting experimental measurements presented in this article. Future improvements are, however, needed to ensure that the process is applicable to systems consisting of conductive phases having a wide range of conductivity and other fluid properties. For example, the nozzles used so far cannot be successful for spraying when the conductivity of the surrounding fluid greatly exceeds that of distilled water, because the applied high voltage often results in significant current flow and sparks at the end of the metal capillary. The large amount of electrical current indicates either local breakdown of the nonconductive meniscus or inability of the nonconductive fluid to cover the end of the metal capillary due to certain wetting problems. The effect of electrostatic fields on liquids wetting solid surfaces is still an unresolved issue.

Acknowledgments

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Literature Cited

Bailey, A. G., "Electrostatic Spraying of Liquids," *Phys. Bull.*, **35**, 146 (1984).

Bailey, A. G., "The Theory and Practice of Electrostatic Spraying," *Atomisation Spray Technol.*, **2**, 95 (1986).

Basaran, O. A., and L. E. Scriven, "Axisymmetric Shapes and Stability of Pendant and Sessile Drops in an Electric Field," *J. Colloid Interf. Sci.*, **140**, 10 (1990).

Cloupeau, M., and B. Prunet-Foch, "Electrostatic Spraying of Liquids in Cone-Jet Mode," *J. Electrostat.*, **22**, 135 (1989).

Felici, N. J., "Recent Developments and Future Trends in Electrostatic Generation," *Direct Curr.*, **4**, 192 (1959).

Harris, M. T., W. G. Sisson, and O. A. Basaran, "Better Ceramics Through Chemistry," *Mat. Res. Soc. Symp. Proc.*, **271**, 945 (1993).

Harris, M. T., and O. A. Basaran, "Capillary Electrostatics of Conducting Drops Hanging from a Nozzle in an Electric Field," *J. Colloid Interf. Sci.*, **161**, 389 (1993).

Landau, L. D., and L. M. Lifshitz, *Electrodynamics of Continuous Media*, Pergamon Press, New York (1960).

Melcher, J. R., *Continuous Electromechanics*, MIT Press, Cambridge, MA (1981).

Rayleigh, L., "On the Equilibrium of Liquid Conducting Masses Charged with Electricity," *Phil. Mag.*, **14**, 184 (1882).

Sato, M., "Cloudy Bubble Formation in a Strong Nonuniform Electric Field," *J. Electrostat.*, **8**, 285 (1980a).

Sato, M., M. Kuroda, and T. Sakai, "Effect of Electrostatics on Bubble Formation," *Kagaku Kogaku Ronbunshu.*, **5**, 380 (1979).

Sato, M., M. Saito, and T. Hatori, "Emulsification and Size Control of Insulating and/or Viscous Liquids in Liquid-Liquid Systems by Electrostatic Dispersion," *J. Colloid Interf. Sci.*, **156**, 504 (1993).

Sato, M., Y. Takano, M. Kuroda, and T. Sakai, "A New Cloudy-Bubble Tracer Generated Under Electrostatic Field," *J. Chem. Eng. Jpn.*, **13**, 326 (1980b).

Scott, T. C., and R. M. Wham, "An Electrically Driven Multistage Countercurrent Solvent Extraction Device: The Emulsion-Phase Contractor," *Ind. Eng. Chem. Res.*, **28**, 94 (1989).

Taylor, G. I., "Disintegration of Water Drops in an Electric Field," *Proc. Roy. Soc.*, **A280**, 383 (1964).

Zeleny, J., "On the Conditions of Instability of Electrified Drops, with Applications to the Electrical Discharge from Liquid Points," *Proc. Cambridge Philos. Soc.*, **18**, 71 (1915).

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