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# Electrostatic characterization of electrohydrodynamic atomization process for particle fabrication

Yanlin Zhao, Jun Yao *

*Beijing Key Laboratory of Process Fluid Filtration and Separation, College of Mechanical and Transportation Engineering, China University of Petroleum-Beijing, Beijing 102249, People's Republic of China*

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ABSTRACT

The electrostatics of electrohydrodynamic atomization (EHDA) process in a shuttle-chamber for polymeric particle fabrication was investigated. The electrical potential field between the spray nozzle and the ground plate inside the chamber was measured three-dimensionally and demonstrated that the electrical field strength increased with decreasing nozzle-plate distance and made more effect on liquid spraying. Four liquids, two inorganic liquids (water, KCl) and two organic liquids (PLGA + DCM ($w/v$ = 7%), PLGA + acetonitrile ($w/v$ = 8%)) were tested and the organic liquids were sprayed to fabricate micro-particles. For spraying inorganic liquids the current scaling was $I \sim Q^{1/2}$ but for spraying organic liquids the current scaling was $I \sim Q^{1/4}$. The accuracy of current scaling depended on the values of $\delta_\mu \delta^{1/3}$, which was independent of the nozzle-plate distances (electric field strength). In addition, the spray current increased with decreasing nozzle-plate distance (or increasing liquid flow rate or liquid conductivity). As a result of spraying polymer solution with volatile organic solvent, particles with size of 0.61– 36.10 μm were fabricated. Particle size decreased with decreasing nozzle-plate distance as well as increasing conductivity of polymer solution or decreasing the liquid flow rate. For all cases of particle fabrication, Rayleigh limit was never reached and no coulomb fission occurred in the experiments.



## 1. Introduction

Electrohydrodynamic atomization (EHDA), which is also known as electrospraying, has caught the attention of many aerosol scientists in the various fields of applications such as mass and size analysis, material synthesis, monodispersed nanoparticles production, drug inhalation, combustion etc. Ijsebaert et al. [1] employed EHDA to generate corticosteroid aerosols in a size range between 1 and 5 μm with a low geometric standard deviation and in quantities sufficiently high to make administration to patient feasible. Loscertales et al. [2] employed electrohydrodynamic (EHD) to generate coaxial electrified liquid jets within the ranges of micrometer to submicrometer. This method allowed a precise tailoring of both the outer radius and the outer-to-inner radius ratio by controlling the flow rates of both liquids and the applied voltage.

To understand the jet break-up mechanism in the electrohydrodynamic atomization, researchers have attempted to develop models and theories for predicting the electrospraying process. Gañán-Calvo et al. [3] performed measurements of the current and size of the primary droplets of sprays generated by electrohydrodynamic atomization of a variety of liquids with different electrical conductivities, permittivities, liquid-gas surface tensions, densities and viscosities. Scaling laws of the spray current as well as the charge and size of the droplets were obtained from a theoretical model of the charge transport. Hartman et al. [4] developed a physical model to calculate the shape of the liquid cone and jet, the electric fields in and outside the cone, and the surface charge density at the liquid surface. The results of this cone-shape model fit well with experimental values with respect to the cone shape and the current, which are both functions of the flow rate, conductivity, applied electrical potential on the electrode, and the electrode geometrical configuration. Later, Hartman et al. [6] developed a model to predict the droplet size, the velocity at jet break-up, and the dominant wavelength of jet break-up. At the end a new theoretical derivation of the droplet size scaling law was given. Recently, López-Herrera et al. [7] developed the electrified co-axial jets of two immiscible liquids emitted from the Taylor cone and investigated the effect of the flow rates of both liquids on the current transported by these coaxial jets and on the size of the compound droplets. It is found that the measurements of the current emitted through the coaxial jet when they are made dimensionless fit satisfactorily the current scaling law of regular electrosprays.

The physics of droplet instabilities due to electrostatic charging has intrigued researchers for over a century, beginning with Rayleigh [8] analyzing the instability resulting from evaporation of highly charged

* Corresponding author at: College of Mechanical and Transportation Engineering, China University of Petroleum-Beijing, Beijing 102249, People's Republic of China.
*E-mail address:* yaojun@xmu.edu.cn (J. Yao).

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droplets. Due to the charged nature of the droplets, there is a maximum limit of surface charge density, termed Rayleigh limit. Once the electrostatic force overcomes the surface tension at the surface of a droplet, it causes the Coulomb fission of the liquid droplets into smaller droplets. The rupture phenomenon was captured by Gomez and Tang [9] in microphotographs that typically showed a droplet with one or two, diametrically opposed, conical protrusions terminating in a fine jet ejecting a stream of much smaller, apparently equisized offsprings when the ruptured droplet had charging level between 70% and 80% of the Rayleigh limit. The experiments yielded disruption thresholds which were lower than the Rayleigh limit, a discrepancy which had been attributed to external perturbing electric fields in the experimental setups, to contaminants in the droplets or to aerodynamic effects [10]. Loscertales and Mora [11] produced a monodisperse cloud of charged droplets and measured the charge and diameter of the residue particles after complete evaporation of the solvent. They suggested that when the droplets contain small monovalent dissolved ions, the electric field on the surface of their solid residues is independent of the diameter of the residue particles and when the ion-solvent pair is well defined, the electric field is also independent of the liquid conductivity and the nature of the counterion. Taflin et al. [10] studied the stability of charged evaporating droplets and the characteristics of their rupture by trapping a single microdroplet in the superposed ac and dc electrical fields of an electrodynamic balance. They found that surface contamination or thermal fluctuations in charge may cause the droplet to explode before the Rayleigh limit. Recently, Smith et al. [12] presented an experimental technique for measuring the droplet size and charge of individual electrosprayed droplets using phase Doppler measurement as they reside in a uniform electric field. This work confirmed that the Rayleigh's analysis [8] of charged droplet instabilities resulting from solvent evaporation is remarkably accurate in predicting the observed discharge events for all employed solvents. More recently, Nakajima [13] employed a simple LDV system with its sensing volume located within a quadruple electrode assembly to measure the temporal variation of the size and charge of a single evaporating droplet. In this work, Rayleigh instability of charged droplets was studied and three types of Rayleigh fissions were observed.

Present work aims to employ the electrospray technology to fabricate particles and to quantitatively investigate the electrostatic effect of working elements, i.e. nozzle-plate distance (electrical field strength), liquid conductivity, liquid flow rate on liquid spraying and particle formation. Three nozzle-plate distances were applied and the corresponding electrical potential fields were measured. Based on liquid spray currents measurements, inorganic and organic solutions are characterized using the current scaling laws. With the EHDA product obtained, the morphology of particles was investigated for the electrostatic effect as well as be verified by a physical model. At the end, the particle formation mechanism was analyzed.

## 2. Experimental design

The experimental setup is shown in Fig. 1. It was modified from the one used by Ding et al. [14] and Xie et al. [15,16]. The setup consists of a glass chamber enclosing a nozzle and a ring electrode, which were both applied with high electrical potential (12 kV and 9 kV) relative to a grounded plate. In the chamber the spray section contained the nozzle and ring, which were independently connected to two high-voltage DC power supplies (Glassman High Voltage Inc., NJ, USA). The electrical potential could be directly shown by the DC power supplies. The copper plate (90 mm × 103 mm) grounded and placed underneath the EHDA spray, worked as the counter electrode to the nozzle-ring and then an electric field was formed between them. The electrical current was measured by an electrometer (Advantest R8252 digital electrometer, Advantest Co. Ltd., Tokyo, Japan) connected to the plate. EHDA spray could be well controlled due to the ring. In this work, three distances of nozzle-plate were used as 55, 70 and 110 mm. A programmable

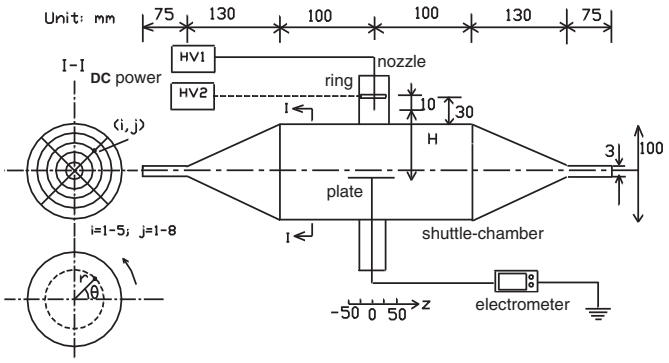


**Fig. 1.** Schematic of the geometry of the experimental setup and the coordinates for electrical potential field measurements.

syringe pump (Stoelting Co., Illinois, USA) was used to pump liquids with nine flow rates 0.1, 0.2, 0.5, 1.0, 1.5, 2.0, 3.0, 4.0, and 6.0 ml/h. To improve the collection of particle generated by pneumatic conveying, compressed air (air pressure 60 psi, air flow rate 30 L/min) is well sealed in the chamber, which enters the chamber from the left side, exits from the bottom and the right side. The working conditions are listed in Table 1, where voltage used is defined as the voltage difference between nozzle and plate (ground). It is noted that, due to very low electrostatic charges carried by spraying particles, the effect on the measurements can be ignored. The morphology of the particle fabricated was observed by a scanning electron miscroscope (SEM) (Jeol JSM 5600 LV, Tokyo, Japan) and particle size was determined through the analysis of the SEM picture using the software Smile View (Ver 2).

Polylactide-co-glycolide (PLGA) is a common polymer and in present work was used to fabricate micro- and nano-particles. Dichloromethand (DCM) and acetonitrile (ACN) were used as the organic solvent and their physical properties are listed in Table 1. In this work, the measured conductivities of liquids were collected using the YSI3200 conductivity instrument (YSI Inc., Yellow Springs, OH, USA). The resolution of the YSI3200 on conductivity measurements is up to 0.0001 μS.

**Table 1**
Experimental conditions.

| | |
|---|---|
| Nozzle size | 0.72 mm |
| Compressed air flow rate | 30 l/min |
| Vacuum permittivity $\varepsilon_0$ | $8.85 \times 10^{-12}$ C$^2$/N·m$^2$ |
| Water surface tension γ | $7.28 \times 10^{-2}$ N/m (20 °C) |
| Water density $\rho_w$ | 1000 kg/m$^3$ |
| Water viscosity $\mu_w$ | $1.002 \times 10^{-3}$ N·s·m$^{-2}$ (20 °C) |
| Water electrical conductivity $K_w$ | $2.162 \times 10^{-4}$ s/m (20 °C) |
| Water to vacuum permittivity ratio $\beta_w$ | 81 (20 °C) |
| KCl solution ($w/v$ = 0.25%) surface tension | $7.29 \times 10^{-2}$ N/m (20 °C) |
| KCl solution ($w/v$ = 0.25%) density $\rho_{KCl}$ | 1002 kg/m$^3$ |
| KCl solution ($w/v$ = 0.25%) electrical conductivity $K_{KCl}$ | 0. 4135 s/m(20 °C) |
| DCM density | 1325 kg/m$^3$ (25 °C) |
| DCM surface tension | $28.1 \times 10^{-3}$ N/m |
| DCM viscosity | $4.4 \times 10^{-4}$ kg m$^{-1}$ s$^{-1}$ |
| Acetonitrile density | 786 kg/m$^3$ (25 °C) |
| Acetonitrile surface tension | $29.1 \times 10^{-3}$ N/m |
| Acetonitrile viscosity | $3.7 \times 10^{-4}$ kg m$^{-1}$ s$^{-1}$ |
| PLGA (P2191) density | 936.71 kg/m$^3$ |
| PLGA (W3066–603) density | 1101.84 kg/m$^3$ |
| PLGA(P2191) + DCM ($w/v$ = 7%) density | 1376 kg/m$^3$ |
| PLGA(P2191) + DCM ($w/v$ = 7%) electrical conductivity | $1.5 \times 10^{-8}$ s/m |
| PLGA (W3066–603) + acetonitrile ($w/v$ = 8%) density | 813.54 kg/m$^3$ |
| PLGA (W3066–603) + acetonitrile ($w/v$ = 8%) electrical conductivity | $1.05 \times 10^{-6}$ s/m |

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