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# Continuous droplets' charge method for the Lagrangian simulation of electrostatic sprays

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ABSTRACT

A main drawback of classical simulation of electrostatic sprays based on the Lagrangian description of droplet trajectories is the large number of droplet-to-droplet electrical interactions that must be computed. We present and assess a new methodology in which some of these interactions are computed using a mean electrical field due to the droplets space charge considered as a continuum. This method has been applied to two systems, comprising 26000 droplets and 3500 droplets, resulting in 112 and 9 times faster computation, without loosing accuracy, as demonstrated in the predictions of impinging flux, droplet number density, and local droplet diameter.



## 1. Introduction

Electrostatic spraying of liquids from electrified capillary tubes can result in quasi-monodispersed droplets with diameters ranging from nanometers to micrometers [15]. The electrical charge carried by the droplets prevents their coalescence, and enables the control of the droplets' trajectories by means of electrodes. These properties are critical in the technique of electrospray ionization mass spectrometry (ESI-MS) [8], in the electrospray production of pharmaceutical particles [6,7,20,24], and other electrospray applications such as micro- or nanoparticles and thin coatings [3,5,16–18,21].

The numerical simulation of electrospray systems, which are characterized by a large number of design variables, helps to reduce the number of experimental tests needed when developing designs for specific applications. In addition, numerically simulated systems can be interrogated in much more detail than experimental systems.

The most detailed mathematical descriptions available for simulating droplets plume dynamics are based on Lagrangian models, which track the forces acting on each droplet, and predict the resulting individualized droplets' motions [10–12,23]. Such individualized tracking of the droplets is straightforward to model; however, the numerical simulation of practical systems is hampered by long computation times (CPU times).

At the root of this problem is the requirement to compute $N(N-1)/2$ droplet–droplet electrostatic interactions at each time step of the computation, for $N$ droplets in the plume. For instance, as reported by Grifoll and Rosell-Llompart [11]; a simulation of 0.2 s of an electrospray comprising 26000 droplets took 1658 h of CPU time for an integration time step of 1 μs. This 0.2 s period covered the transient and a portion of the steady state needed to generate a statistically significant sample of independent snapshots of the charged spray.

The challenge of describing electrospray plumes with reasonable CPU times has motivated several studies. Higuera [13,14] has proposed an Eulerian (continuum) model for a dilute spray, which gives realistic results at a fraction of the computational cost of a Lagrangian simulation. The Eulerian model can be applied in the regions where the fluctuations of the droplet velocity are negligible over its local average; for example, away from the droplet formation region.

Within the Lagrangian framework, Yang et al. [25] have used graphics processing units (GPUs) to simulate millions of droplets trajectories with a (theoretical) computational power of 10 Tera FLOPS. Currently, this promising alternative to CPU-based

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calculations must be coded using a low-level assembly language that runs on both GPUs and CPUs.

Another Lagrangian methodology, implemented by Grifoll and Rosell-Llompart [11] on a standard computer (single CPU), is based on a coarse-grained description of the electrical forces due to faraway droplets. This approach reduced the CPU time by a factor of 40 for the aforementioned example of 26000 droplets. Yet, these gains in CPU time could still be insufficient for practical scenarios, in which droplets can easily number in the hundreds of thousands in a single electrospray.

The present study aims to further reduce the CPU time in the Lagrangian framework, by implementing a continuous description of the electrical field created by the droplets' charge (*space charge*). In previous studies, and for computational purposes, the *total electrical field* sensed by each droplet is split into an *external field*, generated by the static parts of the system (electrodes), and a *space charge field* due to the droplets charge. In our proposal the *total electrical field* is computed from a continuous charge distribution which represents the droplets' charge, and is updated periodically.

## 2. Model and governing equations

### 2.1. Lagrangian model

Electrostatically charged droplets are injected into the system near the end of an electrified capillary tube, which holds a conical liquid meniscus called a Taylor cone [9]. The droplets travel towards a collection plate which is Earth-grounded. The Lagrangian model describes the droplets' spray plume as a $N$-body system governed by Newton's laws of motion

$$\frac{d\mathbf{R}_i}{dt} = \mathbf{V}_i \tag{1a}$$

$$m_i \frac{d\mathbf{V}_i}{dt} = \mathbf{F}_i \tag{1b}$$

where $\mathbf{R}_i = (x_i, y_i, z_i)$ is the position vector of droplet $i$ (m), $m_i$ is its mass (kg), $\mathbf{V}_i$ its velocity (m/s), and $\mathbf{F}_i$ is the sum of all forces acting on it (N). In this study, the $z$ axis coincides with the capillary tube axis, and $x$ and $y$ are transversal axes. The resultant force can be decomposed as

$$\mathbf{F}_i = \mathbf{F}_{\mathrm{D}\,i} + \mathbf{F}_{\mathrm{elec}\,i} \tag{2}$$

where $\mathbf{F}_{\mathrm{D}\,i}$ is the drag force arising from the droplet's motion relative to its gas surroundings (N), and $\mathbf{F}_{\mathrm{elec}\,i}$ is the electrical force (N) experienced by droplet $i$. Considering the droplets as spherical particles surrounded by a still gas, the drag force is calculated as

$$\mathbf{F}_{\mathrm{D}\,i} = -C_{\mathrm{D}\,i}\frac{\pi}{8} d_i^2 \rho_g \mathbf{V}_i |\mathbf{V}_i| \tag{3}$$

where the drag coefficient, $C_{\mathrm{D}\,i}$, has been estimated from Ref. [1]

$$C_{\mathrm{D}i} = \frac{24}{\mathrm{Re}_i}\left(1 + 0.1104\sqrt{\mathrm{Re}_i}\right)^2 \tag{4}$$

valid for Reynolds number $\mathrm{Re}_i = |\mathbf{V}_i| d_i \rho_g / \mu_g < 5000$. In the above equations, $d_i$ denotes the particle diameter (m), $\mu_g$ (Pa s) is the dynamic viscosity of the gas and $\rho_g$ is the gas density (kg/m$^3$). While the restriction of still gas can be relaxed as shown by Arumugham-Achari et al. [2], in the present work we have kept it for simplicity.

The electrical force experienced by droplet $i$ is the product of the droplet charge $q_i$ and the electrical field at the droplet's position $\mathbf{E}(\mathbf{R}_i)$:

$$\mathbf{F}_{\mathrm{elec}\,i} = q_i \cdot \mathbf{E}(\mathbf{R}_i) \tag{5}$$

Depending on how the electrical field is calculated, one can distinguish different submodels, as described next.

### 2.2. Discrete Charge submodel

This is the classical approach adopted in most previous Lagrangian simulations of electrosprays. The electrical field sensed by a droplet $i$, $\mathbf{E}(\mathbf{R}_i)$ in Equation (5), is computed as the sum of the so-called *external* electrical field at the droplet's position ($\mathbf{E}_{\mathrm{ext}}(\mathbf{R}_i)$) due to the static structure (usually, electrodes), plus the field created by the *space charge* associated with all the other droplets in the spray plume and their images on the collection plate:

$$\mathbf{E}(\mathbf{R}_i) = \mathbf{E}_{\mathrm{ext}}(\mathbf{R}_i) + \mathbf{E}_{\mathrm{sc}\,i} \tag{6}$$

$\mathbf{E}_{\mathrm{ext}}(\mathbf{R}_i)$ is minus the gradient of the electrical potential which is found by solving Laplace's equation (excluding the spray charges), with appropriate conditions at the static boundaries. $\mathbf{E}_{\mathrm{sc}\,i}$, on the other hand, is calculated by adding all of the contributions from the other droplet charges [11]:

$$\mathbf{E}_{\mathrm{sc}\,i} = \frac{1}{4\pi\varepsilon}\left[\sum_{i\neq j}^{N} q_j\left(\frac{\mathbf{R}_{ij}}{R_{ij}^3} - \frac{\mathbf{R}_{ij}}{R_{ij}^3}\right) - q_i\frac{\mathbf{R}_{il}}{R_{il}^3}\right] \tag{7}$$

where $\mathbf{R}_{ij} = \mathbf{R}_i - \mathbf{R}_j$ is the displacement vector between the position vectors of droplets $j$ and $i$ (m), $\mathbf{R}_{ij} = \mathbf{R}_i - \mathbf{R}_J$ is the displacement vector between the position vectors of the image of droplet $j$, which is positioned inside the collection plate at $\mathbf{R}_J = (x_j, y_j, 2H - z_j)$, and droplet $i$ (m) (where $H$ is the separation between the capillary end and the collection plate), $\varepsilon$ is the gas permittivity (taken for air as $8.854 \times 10^{-12}$ A s/V m) and $N$ is the total number of droplets in the plume. As mentioned in the Introduction, it is the computation of the $\mathbf{E}_{\mathrm{sc}\,i}$ term through Equation (7) that significantly slows down the overall computation.

Fig. 1(a) shows the modulus of the transversal component of the space charge field $E_{\mathrm{sc}\,t} = \sqrt{E_{\mathrm{sc}\,x}^2 + E_{\mathrm{sc}\,y}^2}$ experienced by three droplets as they travel through a spray previously simulated by Grifoll and Rosell-Llompart [11] using the Discrete charge submodel (therein used in the so called “complete simulation”). The trajectories of the droplets projected on the $(r, z)$-plane are shown in Fig. 1(b) overlaid with a snapshot of the spray plume.

The transversal field modulus $E_{\mathrm{sc}\,t}$ is smooth far away from the centerline, but it is jagged close to the centerline, due to the influence of neighboring droplets. Similar trends have been found in analyses of other droplets' trajectories, not shown. The finding that the electrical field is very smooth within a large portion of the spray (away from the centerline) suggests that a continuous description of the electrical field can be adopted in this region. This possibility is explored in the next subsection.

### 2.3. Continuous Charge submodel

A local continuous density of charge, $\rho$ (C/m$^3$), and the electrical potential $\phi$ (V) satisfy Poisson's equation:

$$\nabla^2\phi = -\frac{\rho}{\varepsilon} \tag{8}$$

from whose solution the electrical field can be calculated with

$$\mathbf{E} = -\nabla\phi \tag{9}$$

However, upon injection, the droplets are organized nearly in a line [22]; therefore, the space charge density is ill defined there.