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Electrohydrodynamic atomization: A two-decade effort to produce and process micro-/nanoparticulate materials

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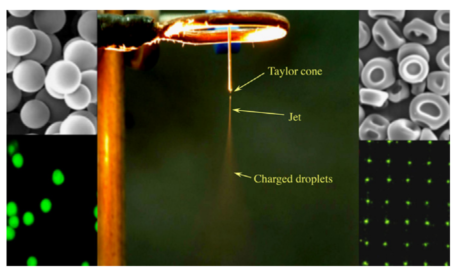
HIGHLIGHTS

- Extensive investigation of EHDA in the past two decades.
- Production of micro-/nanoparticulate materials with well-controlled properties.
- Generation of desired patterns/structures through jet writing or deposition.
- Biomedical applications including drug delivery and regenerative medicine.
- Understanding the mechanism of EHDA using modeling and simulation techniques.

GRAPHICAL ABSTRACT



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ABSTRACT

Electrohydrodynamic atomization (EHDA), also called electrospray technique, has been studied for more than one century. However, since 1990s it has begun to be used to produce and process micro-/nanostructured materials. Owing to the simplicity and flexibility in EHDA experimental setup, it has been successfully employed to generate particulate materials with controllable compositions, structures, sizes, morphologies, and shapes. EHDA has also been used to deposit micro- and nanoparticulate materials on surfaces in a well-controlled manner. All these attributes make EHDA a fascinating tool for preparing and assembling a wide range of micro- and nanostructured materials which have been exploited for use in pharmaceutics, food, and healthcare to name a few. Our goal is to review this field, which allows scientists and engineers to learn about the EHDA technique and how it might be used to create, process, and assemble micro-/nanoparticulate materials with unique and intriguing properties. We begin with a brief introduction to the mechanism and setup of EHDA technique. We then discuss issues critical to successful application of EHDA technique, including control of composition, size, shape, morphology, structure of particulate materials and their assembly. We also illustrate a few of the many potential applications of particulate materials, especially in the area of drug delivery and regenerative medicine. Next, we review the simulation and modeling of Taylor cone–jet formation for a single and co-axial nozzle. The mathematical modeling of particle transport and deposition is presented to provide a deeper understanding of the effective parameters in the preparation, collection and pattering processes. We conclude this article with a discussion on perspectives and future possibilities in this field.

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1. Introduction

The phenomenon of electrohydrodynamic atomization (EHDA), also simply called electrospray, was firstly observed and recorded by William Gilbert in 1600 (Gilbert et al., 1600). In 1750, Jean-Antoine (Abbé) Nollet, a French Clergyman and physicist, reported the earliest observation on electrospray, more than two centuries before the terminology was generated, demonstrating water flowing from a container would aerosolize if the container was electrified and put close to electrical ground (Dumont and Cole, 2013). Around one century later, Lord Kelvin invented a setup composed of two liquid nozzles that were bridged to opposite collection reservoirs and demonstrated small differences in charging between water dripping from the nozzles instantly caused differences in kilovolt scale and electrospraying from the nozzles (Smith, 2000). In 1882, Lord Rayleigh theoretically evaluated the charge that a liquid droplet could carry to the greatest extent, known as the "Rayleigh limit", which was confirmed experimentally 100 years later (Taylor, 1964; Gomez and Tang, 1994 and Duft et al., 2003). The first patent related to EHDA setup appeared in 1900 (Cooley, 1900). Zeleny conducted the electrospray experiment with ethanol and photographed a cone–jet in 1914 (Zeleny, 1917). In 1960s, Taylor developed a mathematical description of the EHDA process, simulating the conical shape of the liquid phase in the presence of an electrical field that became known as Taylor cone later on (Taylor, 1966). In the 1980s, Fenn and coworkers performed a series of studies that eventually made electrospray capable of introducing dissolved analytes into the gas phase for mass analysis (Fenn, 1989). John Bennett Fenn won the Nobel Prize in Chemistry in 2002 because of his contribution to electrospray ionization for analyzing biological macromolecules (Grayson, 2011).

EHDA is a well-practiced technique for generating very fine droplets with mono-dispersed size from a liquid under the influence of electrical forces. Though the applications of EHDA to many fields are numerous, including raindrops in thunderclouds, combustion, crop spraying, and electrospray ionization mass spectroscopy, this technique has started attracting a lot of attention since 1990s for producing and processing micro-/nanoparticulate materials in a rich variety of applications. In the present work, we begin with describing the basic setup and mechanism of EHDA, followed by a discussion of controlling composition, size, shape and secondary structure, collection, and deposition of micro-/nanoparticulate materials and their applications in the biomedical fields.

2. Electrohydrodynamic atomization: principles and basic setup

The basic setup for EHDA consists of several major components: a syringe pump, a syringe, a metal needle serving as nozzle, a high voltage power source and a grounded substrate serving as a collector (Fig. 1a). Some EHDA setups employ a closed chamber where air/nitrogen flow transfers particles toward collecting filters. The use of the chamber reduces the evaporation of solvents and facilitates formation of smaller particles with smoother surface morphology (Xie et al., 2006b, 2008b).

EHDA (electro refers to electric energy; hydrodynamic refers to fluid dynamics applied to liquids; and atomization refers to making bulk liquid into fine droplets) is a process where a liquid jet breaks up into fine droplets under influence of an external

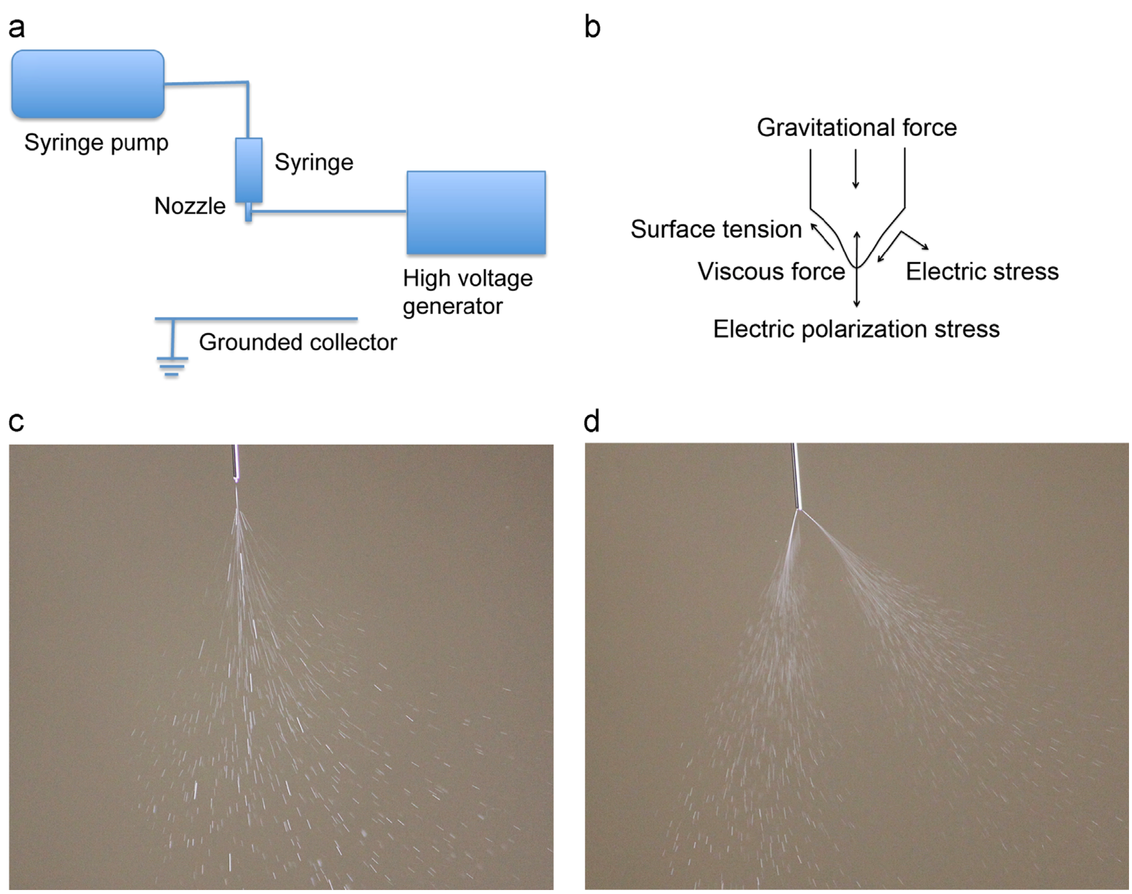

Fig. 1. Schematic illustration of (a) basic setup of EHDA and (b) major forces on the spraying cone (reprinted from Hartman et al., 1999, with permission from Elsevier). Photographs showing (c) a single-cone spraying mode (also called stable cone–jet mode) and (d) a multiple-cone spraying mode. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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