Novel Method for Micro- and Nano- Particle Preparation by Electrohydrodynamic Atomization

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In recent years, there has been an increasing interest in the fabrication of micro- and nano- particles for controlled drug delivery purposes. Methods such as spray drying and emulsion methods have been well studied. Yet, disadvantages such as high fabrication temperature for the spray drying method and low encapsulation efficiency for emulsion methods have yet to be overcome. Electrohydrodynamic atomization has recently been shown to be suitable for the preparation of nearly monodisperse polymeric particles[1].

Electrohydrodynamic Atomization

The electrohydrodynamic process was first investigated in the late nineteen hundreds[2,3,4]. In 1952, Vonnegut and Neubauer[5] suggested that, when a certain instability mode is chosen by carefully manipulating the electrical potential applied to the nozzle, the electrohydrodynamic atomization process is able to produce highly monodisperse droplets.

The most common electrohydrodynamic atomization process for the fabrication of monodisperse polymeric particles involves a two step process. In the first step, a solution containing an organic solvent and a polymer solute is sprayed through a nozzle which is maintained at a high electrical potential. The electrical potential difference between the nozzle and a grounded needle placed directly below the nozzle creates an electrical field that causes the formation of the Single Taylor Cone/Single Jet, in which a liquid cone is formed at the tip of the nozzle and a thin jet is ejected from the apex of the cone. The jet breaks into nearly monodisperse droplets. If the nozzle is subjected to a sufficiently high potential, multiple jets are observed. The Single Taylor Cone/Single Jet mode was chosen for particle fabrication because the droplets thus formed are less polydisperse. Subsequent to droplet formation, organic solvent evaporates and solid particles are then formed and collected.

To change the size of the particles, the first step is to change the size of the droplets. There are many methods to change the size of the droplets, but most of them require modifications to the physical system or changes in the properties of the solution being atomized. Such methods are unwieldy in industrial applications. To overcome these problems, the present study aims at designing a new method for the preparation of micro- and nano-particles.

Experimental Apparatus

Two separate electrodes are used to produce the electrical field. The nozzle is a 29 gauge spinal tap needle from Becton Dickinson with the sharp tip cut and filed to form a flat tip. It has an outer diameter of 340 microns. It is connected to a Glassman high voltage DC power supply. The second electrode is in the form of a ring that is placed slightly above the outlet of the nozzle, with the nozzle going through the center of the ring. The ring electrode is made from copper tube with outside diameter of 2mm and formed into a ring with a diameter of 40mm. The center of the ring electrode is placed 10mm from the tip of the nozzle. It is connected to a separate Glassman high voltage DC power supply. This arrangement enables the nozzle electrical potential and the ring electrical potential to be varied independently. The ground needle is also a 29 gauge spinal tap needle which is located 100mm below the tip of the nozzle (Figure 1). Ijsebaert[6] has previously made use of the ring electrode in his study of the production of aerosols, but the ring electrical potential was set to a constant and was thought only to have the effect of focusing the spray.

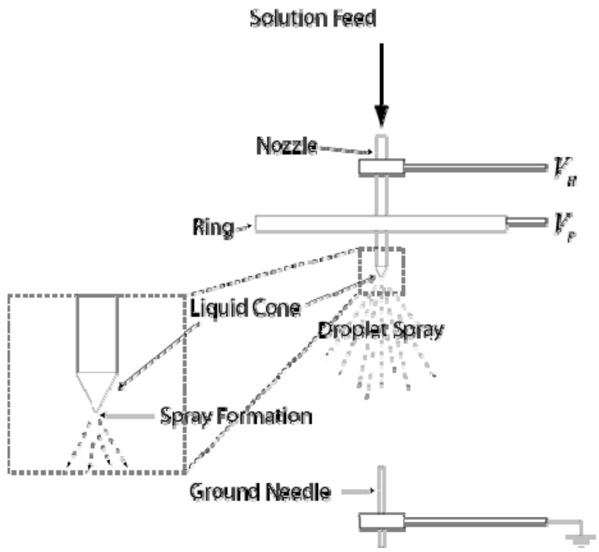

Figure 1: Schematics of the electrohydrodynamic atomization setup.

Phenomenological Observations

For phenomenological observation, a Nikon D2H digital single lens reflex camera is used with a 105mm macro lens. To facilitate the observation, backlighting is used.

To obtain the Single Taylor Cone/Single Jet mode of electrohydrodynamic atomization, the nozzle electrical potential is first set to a constant value of, say, 8kV: and the ring electrical potential is then raised until the required mode is obtained. A low ring electrical potential results in the multiple cone mode. A somewhat higher ring electrical potential produces the Single Taylor Cone/Single Jet mode. An even higher ring electrical voltage produces the gravity dripping mode. By slightly varying the ring electrical potential while keeping the nozzle voltage constant, we observed that the Single Taylor Cone/Single Jet mode can be maintained. As the ring electrical potential is decreased, the cone angle of the Taylor Cone is also reduced. The range of cone angle for which the Single Taylor Cone/Single Jet can be obtained ranges from about 10 degree to about 80 degree (Figure 2).

We can see that changing the ring electrical field slightly does not change the mode of the spray from the Single Taylor Cone/Single Jet mode. Maintaining a Single Taylor Cone/Single Jet is important in the fabrication of particle since literature[1] has shown that particle fabricated with this spray mode has a lower size distribution compared to other spray mode.

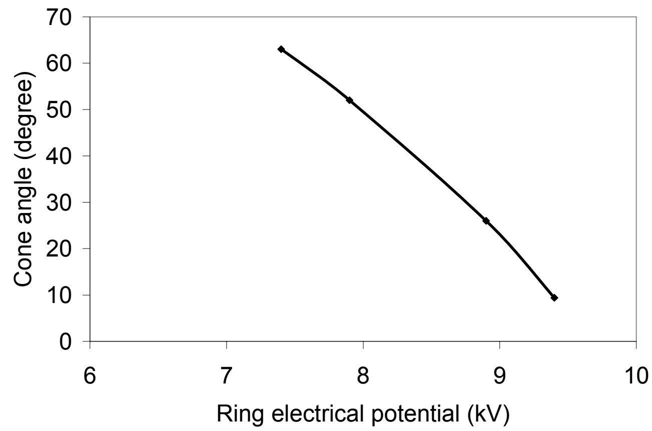

Figure 2: (Left) A typical electrohydrodynamic atomization in the Single Taylor Cone/Single Spray mode. (Right) The changes in the cone angle of the Taylor cone when the ring electrical potential is varied while the nozzle electrical potential is kept constant at 8kV. The flow rate of the solution is 6ml/h.

Phase Doppler Particle Analyzer

A Phase Doppler Particle Analyzer was used to determine the droplet size in the spray. The Phase Doppler Particle Analyzer used was obtained from Aerometrics and it has a measurement range of 0.5 to 200 microns. For pure dichloromethane, results have shown that increasing the electrical potential on the ring while maintaining a constant electrical potential on the nozzle increases the size of the droplets (Figure 3).

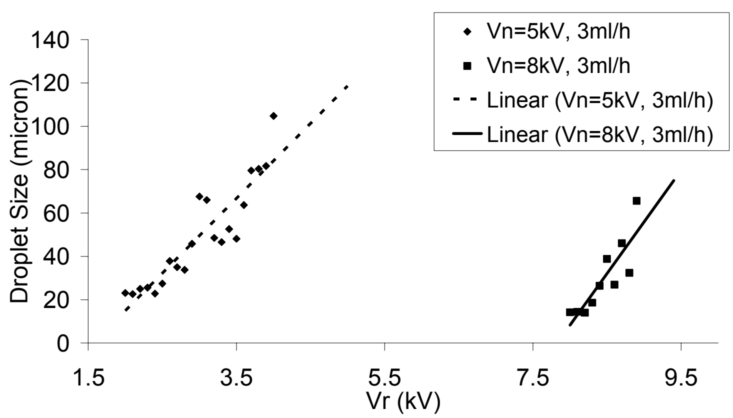

Figure 3: Variation of droplet size when the ring voltage is varied while the nozzle voltage is fixed. Two different sets of data are shown, the line on the left represents changes in droplet size when the nozzle electrical potential is kept at 5kV, while the line on the right represents changes in droplet size when the nozzle electrical potential is kept at 8kV. Linear represents lines of linear fit of the experimental data.

Scanning Electron Microscopy

If polycaprolactone is used as the solute, particles may be fabricated. The particles are collected on a rotating drum located about 10 cm below the nozzle. The rotating drum is grounded and replaces the needle as the counter electrode. Simulation has shown that swapping the grounded needle with the grounded drum has minimal effect on the electrical field strength near the nozzle. Scanning electron micrographs were used to determine the size of the particles. Results have shown that the size of the particles increases with increasing ring electrical potential (Figure 4.).

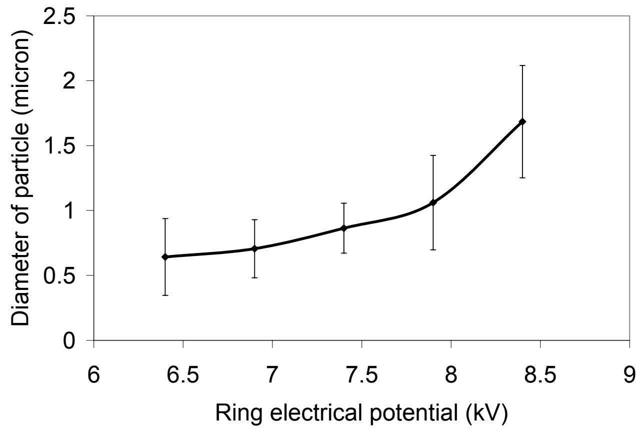

Figure 4: Changes in particle size when the ring electrical potential is varied. The ring electrical potential is varied while the nozzle electrical potential is kept constant at 8kV. The flow rate of the solution is 3ml/h.

Concluding Remarks

The use of a ring electrode has been shown to have advantages as compared to the conventional methods of size control in electrohydrodynamic atomization. Without any modification to either the physical system or the solution, we are able to change the size of the particles in real time by simply changing the electrical potential applied to the ring electrode. A clear trend can be observed in which decreasing the electrical potential of the ring can generate particles with smaller diameters, down the to nanometer range.

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