



# Performance study of a twin-head electrospray system

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## ABSTRACT

A twin-head electrospray (THES) aerosol generation system with one spray head producing positively charged particles and the other producing negatively charged ones has been constructed and its performance has been experimentally evaluated in this study. The charge-enhanced coagulation between oppositely charged droplets generated by the THES was verified by the measuring the resultant particle size distributions via a scanning mobility particle sizer (SMPS), the imaging of dye-labeled coagulated particles via a fluorescence microscope and the chemical composition characterization of generated particles via high-efficiency liquid chromatography (HPLC). The characterization time of charge-enhanced coagulation in our study's generator was estimated to be in the order of  $10^{-5}$  s under the tested operational conditions. The size evolution due to charge-enhanced coagulation was studied for particles with initial sizes ranging from 10 to 710 nm. Compared with those prior to the coagulation, the size distributions of particles upon coagulation became broader. Further, regardless the initial charge levels, the peaks of resultant particle size distributions were about 1.3 times larger than the corresponding ones before coagulation, indicating the collision of two oppositely charged particles. As expected, the number of charges on the resultant particles was significantly reduced in the THES generation system. However, only a small fraction of resultant particles carried zero electrical charge. Our study also evidenced that a maximal 70% transmission efficiency can be achieved for the THES system. It is therefore concluded that the twin-head ES technique makes it feasible to take unique advantage of electrosprayed particles without any charge reduction scheme.

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## 1. Introduction

Because of its capability to generate un-agglomerated, monodisperse particles with diameters ranging from nanometers to micrometers when operated at the so-called cone-jet mode, electrohydrodynamic atomization (or electrospray; ES) has received much attention in the past few decades. The application of electrospray has been proposed in a wide range of areas, for example, particle/thin film coating, material synthesis, suspension dispersion, drug delivery, and particle encapsulation (Jaworek, 2007; Wyslouzil et al., 2009; Kim et al., 2010; Stride et al., 2010; Trotta et al., 2010). In the electrospray process, a DC electrical field is established between the spray head and the reference electrode. The balance between electrical force on the surface of liquid meniscus at the spray head exit and the surface tension of the liquid enables the formation of a liquid cone and a jet-spray emission from the cone apex. The breakup of the liquid jet

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converts into droplets, resulting in the atomization of the liquid. Because of the presence of the DC electrical field electrospayed droplets are highly charged to the same polarity, which prevents the particles from the agglomerating (due to columbic repulsion). A simple electrospray system consists of a capillary and a grounded reference electrode. In addition to the simple capillary electrospray systems, the spray heads in dual-capillary and tri-capillary configurations have recently been explored for more sophisticated particle applications, for example, generating particles with multiple coating layers or containing multi-functional components (Mei and Chen, 2007; Lee et al., 2010; Kim and Kim, 2011; Lee et al., 2011).

Particle coagulation is one of the inter-particle phenomena often encountered in the cases of aerosols in high concentration. Particle coagulation can be enhanced by attractive electric forces among particles. With the high charge level associated with electrospayed particles, charge-enhanced coagulation of particles generated by two spray heads with one at positive high voltage and the other at negative high voltage has been proposed in various applications. The concept of using two electrospray heads to generate counter-charged droplets was first proposed in 1965 to study the collision of charged droplets (Schneider et al., 1965). Electrostatic atomization was used in this previous study to generate droplets with the diameters of approximate 180  $\mu\text{m}$ . The experiment confirms that the collision rate of oppositely charged droplets was much higher than that of droplets carrying like charges. Langer et al. studied the encapsulation process between charged liquid droplets and solid particles (Langer and Yamate, 1969). Their work again used electrostatic atomization to generate droplets with diameters larger than 1  $\mu\text{m}$ . A high percentage of encapsulated particles were observed for droplets with diameters larger than 10  $\mu\text{m}$ . The authors however found that a majority of charged droplets were deposited on the electrically grounded plate instead of entering the encapsulation chamber through orifices, due to the relatively small orifice compared to the scale of the grounded metal plate.

The possibility of utilizing bipolar coagulation of particles generated by two electrospray heads in the powder production has further been explored (Borra et al., 1999; Camelot et al., 1999). Each of two electrospray heads in their system generated monodisperse droplets from one reaction precursor. The coagulation of precursor droplets with positive and negative charges created a good mixing of chemical compounds in the microscale. The droplets made by the bipolar coagulation could then serve as micro-reactors to form powders with the desired composition. Supermicrometer-sized droplets are favorable in these studies because of the powder production. More, the studies were primarily focused on the micro-mixing process and on the micro-reactor. Limited information on the system performance and evaluation of the coagulation process has been reported in the literature.

The dual spray system, having one head for electrospin/electrospray to generate fibers/particles and the other for electrospray to reduce the charges on generated fibers/particles, has been proposed for nano-mat formation and for bio-aerosol generation (Morozov and Vsevolodov, 2007; Morozov, 2011). For nano-mat formation, the electrospin head was used to generate polymer fibers and the electrospray head for generating ions by spraying a volatile solvent to reduce the electrical charges on electrospun fibers. Electrospun fibers were not immediately neutralized after being deposited. Opposite charges were accumulated on both side of the nano-mat. As a result, generated nanofibers collapsed into a film because of electrostatic attraction. By replacing the electrospin head with an electrospray one, active bio-aerosol was dispersed and neutralized by the ions generated from the other electrospray head when spraying volatile solvents. It was found that the biological activity of bio-aerosol is mostly preserved in the above electrospray, as compared to that neutralized by an ion source via corona discharging. Park et al. also studied the interactions between fibers and particles generated by electrospin and electrospray heads in opposite high voltages (Park et al., 2008). They observed that the stream of fibers changed the moving direction towards oppositely charged droplets. However, the coagulation between oppositely charged droplets generated by the same system was hardly observed in their study. It is because most of charged droplets were deposited on the surfaces of counter-charged nozzles.

Although twin-head ES systems have been previously investigated, all the previous studies were focused on specific applications. No systematic investigation on the system performance and droplet coagulation process involved in the systems has been previously reported. More, all previous studies only investigated the cases of particles with initial sizes in the supermicrometer range. In this study, a twin-head electrospray system, enabling to carry out such a study, has been designed. The performance of designed twin-head ES system has been systematically evaluated for particles with initial diameters ranging from 10 to 700 nm.

## 2. Twin-head electrospray system and experimental methods

### 2.1. Design of twin-head electrospray system

A schematic diagram of the designed twin-head electrospray systems is shown in Fig. 1a and b. The studied spray system consists of two separate spray chambers with an identical dual-capillary electrospray head in each, and a particle coagulation chamber. The design of the spray heads enables us to use them either in the single-capillary or coaxially-aligned dual-capillary configuration. The inner diameter (ID) of the outer capillary was fixed at 0.046". The outer and inner diameters of the inner capillary were at 0.03125" and 0.008", respectively. The distance between the ES head exit and the entrance of the coagulation chamber was adjustable. An outer case made of transparent plexiglass was used to hold the spray heads. The plexiglass case also served as the insulation between the spray head at the high voltage and the orifice disk at the electrical ground. Air flow, as aerosol carry gas, was introduced into each spray chamber via the inlet tube

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