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Influence of spray conditions on droplet charge per unit volume for electrospray deposition

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ABSTRACT

In electrospray deposition (ESD), the droplet charge per unit volume is an important parameter for optimizing a coating condition. Previous research has investigated the electrostatic charge on droplets both theoretically and experimentally. Recently, an additive solvent technique was proposed that yields smooth organic thin films, and several types of high-performance organic devices were demonstrated. However, investigating just the droplet charge per unit volume is insufficient to understand the advantages of the additive solvent. In the present work, we investigate the influence on the droplet charge per unit volume of several parameters such as applied voltage, liquid flow rate, and spraying distance to optimize the charging conditions even at a high liquid flow rate, which leads to a high deposition rate. The escaped droplet charge per unit volume is estimated from the relationship between spraying distance and droplet charge per unit volume. In addition, we also estimate how an additive solvent and a polymer affect efficient charging in ESD. The droplet charge per unit volume becomes high upon using the additive solvent, even at a low concentration (10 vol%). The additive solvent technique is found to be a useful tool for realizing high the droplet charge per unit volume, which results in a large spray diameter. However, because of its low concentration in the solvent, the polymer has little effect on the droplet charge per unit volume.

1. Introduction

Liquids atomized electrostatically have been widely used for several applications such as electrospray-ionization mass spectroscopy (Banerjee & Muzudar, 2012; Dole et al., 1968; Ho et al., 2003), negative-ion generators for air purifiers and air conditioners, electrostatic spray-painting processes, and electrospinning methods for fabricating nanofibers (Doshi & Reneker, 1995; Huang et al., 2003; Li & Xia, 2004). Currently, interest has grown in using electrospray deposition (ESD) methods as novel coating methods for organic thin-film devices, and several types of organic devices have already been demonstrated (Hu et al., 2013; Koishikawa et al., 2013; Yamauchi et al., 2014). A significant advantage of such methods is that an organic multilayer structure can be formed by using a simple experimental setup without a vacuum chamber (Ali et al., 2012; Fukuda et al., 2013). However, a serious problem with such methods is that a smooth organic thin film is difficult to form because of the fast evaporation of the solvent and the narrow spray diameter. Recently, Ju et al. (2009) demonstrated that an additive solvent technique solves this problem by showing that a soluble solvent for semiconducting polymers combined with an additive solvent with the high dielectric constant allows smooth organic thin films to be fabricated.

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To date, this technique has been used to fabricate organic light-emitting diodes, organic photovoltaic cells, and organic thin-film transistors (Fukuda et al., 2011a; Zhao et al., 2014). In addition, a practical application of the ESD process has been demonstrated in the form of a roll-to-roll printing system that achieves high throughput over a large coating area (J.-H. Kim et al., 2010; J.S. Kim et al., 2010). Typically, the ESD technique involves dispensing liquid at a slow flow rate through a glass capillary or a stainless-steel needle with an inner diameter that ranges from several tens to hundreds of microns. Because a certain range of applied voltage and flow rate is acceptable for the ESD process, multinozzle (Jung et al., 2010) and multiplexed (Deng & Gomez, 2007) systems have also been investigated for fast deposition over a large area.

Another approach involves basic research into the mechanism by which sprayed droplets are ionized, the mechanism by which the Taylor cone forms, and the movement of sprayed droplets under the influence of an electrostatic field due to high voltage (Gañán-Calvo & Montanero, 2009; Hartman et al., 2000; Zhong et al., 2009). In addition, the droplet velocity (Hartman et al., 2000) and the diameter (Gañán-Calvo et al., 1997; Kim et al., 2011) have been estimated by using a phase Doppler anemometer and a high-speed charge coupled device (CCD camera). The droplet diameter can be controlled to some extent via the flow rate of the liquid and the applied voltage, and it ranges from hundreds of microns down to several tens of nanometers (Jaworek & Sobczyk, 2008). Furthermore, the droplet charge per unit volume has been investigated to obtain efficient ionization of the liquid by applying a high voltage. The charge density at the Rayleigh limit (Q_R) can be estimated from (Rayleigh, 1882)

$$Q_R = 2\pi(16\gamma\epsilon_0 r^3)^{1/2}, \qquad (1)$$

where γ, ϵ_0, and r are the liquid surface tension, the dielectric constant of vacuum, and the droplet radius, respectively. The Rayleigh limit affects the conditions for efficient ESD and depends on the surface tension, the dielectric constant, and the droplet radius.

A small droplet (i.e., diameter $< 1\ \mu m$) is necessary to fabricate a multilayer structure with a smooth surface, and high droplet charge per unit volume results in efficient droplet fission at the syringe tip (Grimm & Beauchamp, 2002). These facts indicate that efficient charging is important for fabricating organic thin-film device by ESD. However, a fast liquid flow rate results in insufficient droplet charge per unit volume, making a stable electrospray difficult to achieve. In addition, the effect on the droplet charge per unit volume of adding a polymer to the liquid is unknown, and the droplet charge per unit volume of the mixed solvent has not been investigated, even though the additive solvent technique is important for fabricating organic thin films. Therefore, further investigations into charged droplets are required to develop practical ESD methods.

In the present work, we investigate the droplet charge per unit volume of sprayed droplets to understand which parameters are important in the ESD process. In particular, we investigate the relationship between process parameters and droplet charge per unit volume. In addition, we also estimate the droplet charge per unit volume of the mixed solvent containing the semiconducting polymer to understand the effect of the polymer and additive solvent.

2. Experimental

Figure 1 shows a schematic of the configuration of the ESD setup. A high voltage was applied by an Element ETM3-20K01PN1 high-voltage source to a stainless-steel needle fixed to the tip of a glass syringe (Hamilton, 1750). The inner diameter of the stainless-steel needle was 150 μm, and a syringe pump (Suruga Seiki, KZL06050) was used to control the flow rate of the liquid in the glass syringe. The scanning speed of the syringe pump was varied from 10 to 140 μm/s, and the liquid flow rate was estimated from the scanning speed and the diameter of the syringe. The calculated flow rates ranged from 71 to 990 nL/s. Since humidity adversely affects electrospray conditions (Bodnár & Rosell-Llompart, 2013), dry air was injected into the glove box in which the ESD system was set up. Consequently, all experiments were conducted at

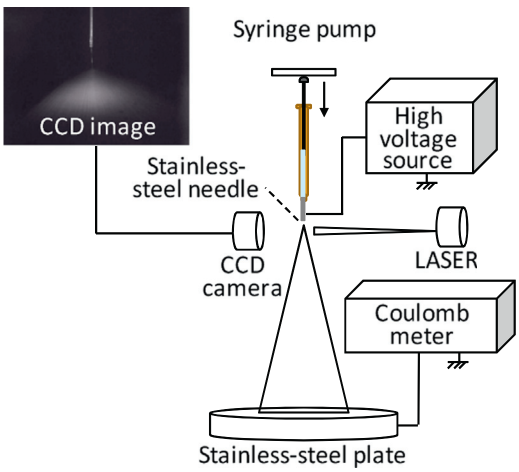

Fig. 1. Schematic of ESD setup including the system for measuring the charge density per unit volume of airborne droplets.