Journal of Aerosol Science 76 (2014) 148–162

Contents lists available at ScienceDirect

Journal of Aerosol Science

journal homepage: www.elsevier.com/locate/jaerosci

An electrospray aerosol generator with X-ray photoionizer for particle charge reduction

Qiaoling Liu, Da-Ren Chen*

Particle Engineering Laboratory, Department of Mechanical and Nuclear Engineering, School of Engineering, Virginia Commonwealth University, 401 W. Main Street, Richmond, VA 23284, USA

ARTICLE INFO

Article history:
Received 8 November 2013
Received in revised form
12 May 2014
Accepted 4 June 2014
Available online 26 June 2014

Keywords:
Electrospray
Charge reduction
Soft X-ray
Nanoparticles

ABSTRACT

A prototype single-capillary electrospray (ES) aerosol generator having a soft X-ray photoionizer for charge reduction has been constructed and its performance under various configurations has been investigated in this study. The prototype essentially consists of spray and charge reduction chambers having an orifice disk plate as the partition. Two soft X-ray irradiation directions (i.e., 90° and 180° relative to the spray direction) and two disk plates (with the orifice diameters of 0.25 in. and 1.25 in.) were tested for the prototype configuration. In the investigation, the spray current as a function of ES voltage was recorded. It is shown that the presence of soft X-ray irradiation in the close proximity of the electrospray process resulted in the increase of applied voltage needed to operate the spray while the spray current at the cone-jet mode remained unchanged. The fluorescence analysis was applied in this study to quantify the transmission efficiency and the charge fractions of particles exiting from the prototype generator. A maximal 47% transmission efficiency could be achieved by the prototype having the setup of 180° irradiation and use of 0.25 in. D-orifice plate, and operated at carrier gas flow rate of 8.0 lpm. Under the above setup and operation the charge fraction of particles with the sizes ranging from 30 to 300 nm was further characterized. It is found that the measured charge fractions of particles exiting from the generator is in good agreement with the calculated Fuchs bipolar charge factions for the cases having the particle sizes less than 110 nm. The deviation from the Fuchs charge fraction increases as the particle size increases. It may be because (1) more ions were needed to reduce the charge level on particles of large sizes; (2) spatial non-uniformity of aerosol concentration and ions in charge reduction chamber; and (3) because of the direct photoionization of particles under the X-ray irradiation. At last, the size measurement of ES particles via a scanning mobility particle sizer (SMPS) shows the good quality of monodispersity for particles generated by the prototype when operated at the cone-jet mode.

1. Introduction

Electrospray (ES) or electrodydrodynamic atomization is a technique capable of converting liquids/solutions in the bulk phase into droplet phase with the droplet sizes ranging from nanometers to micrometers (Zeleny, 1914; Chen et al., 1995;

* Corresponding author. Tel.: +1 804 828 2828.
E-mail address: dchen3@vcu.edu (D.-R. Chen).

http://dx.doi.org/10.1016/j.jaerosci.2014.06.003

Loscertales et al., 2002; Jaworek, 2007; Morozov and Vsevolodov, 2007; Lee et al., 2011). The electrospray process had been applied to the surface coating (Hines, 1966; Paul, 1985; van Zomeren et al., 1994; Matsumoto et al., 2005; Fukuda et al., 2011), agricultural treatments (Coffee, 1964), emulsion (Nawab and Mason, 1958; Marín et al., 2007; Lin et al., 2010) or super-micron aerosol production, fuel spraying (Jones and Thong, 1971; Deng et al., 2007; Yuliati et al., 2012), micro-encapsulation (Langer and Yamate, 1969; Xie et al., 2008), ink-jet printers (Tomita et al., 1986), and colloid micro-thrusters (Huberman et al., 1968; Si et al., 2007). More recently, new applications have been explored. Examples are (1) using the electrospray as ion sources for mass spectrometry (ES MS) for the macromolecular detection (Yamashita and Fenn, 1984; Thompson et al., 1985; Fenn et al., 1989; Cole, 1997; Smith et al., 1997; Dulcks and Juraschek, 1999; Loo, 2000; Hautreux et al., 2004; Sterling et al., 2010), (2) monodisperse nanoparticle generation (Chen et al., 1995; Suh et al., 2005; Kim et al., 2010), (3) biomolecule detection using gas-phase electrophoretic mobility molecular analyzer (GEMMA) (Kaufman et al., 1996; Kaufman, 1998; Koropchak et al., 1999; Scalf et al., 1999; Bacher et al., 2001; Allmaier et al., 2008), (4) enhancement of droplet mixing by inter-electrospray (Dunn and Snarski, 1991; Snarski and Dunn, 1991; Dunn et al., 1994), (5) targeted drug delivery by inhalation (Tang and Gomez, 1994; Ijsebaert et al., 2001), (6) micro-mixing for drug power production (Borra et al., 1999), (7) ceramic nanoparticle preparation by electrospray pyrolysis (Lenggoro et al., 2000; Oh and Kim, 2007), (8) preparation of nano-structured ceramic thin films (Chen et al., 1999; Hosseinmardi et al., 2012), (9) electrospray gene transfection (Chen et al., 2000; Zeles-Hahn et al., 2011), and (10) dual-jet electrospray for the nano-encapsulation and enhancement of targeted lung delivery of nano-medicines (Chen and Pui, 2000). The general reviews of the electrostatic atomization processes and applications are covered in the books written by Michelson (1990) and by Bailey (1988) and paper of Salata (2005). More advanced discussion on the spray modes in the process are presented in the works of Cloupeau and Prunet-Foch (1989, 1990, 1994), and Jaworek and Krupa (1999). Among all the operational modes, the cone-jet electrospray has been widely studied because of its production of monodisperse particles.

A typical electrospray setup is in the point-to-plate configuration, i.e., a spray nozzle (i.e., single-, dual- or tri-capillary) facing a plate (with/without an orifice at the center). To operate the ES, a DC electrical field is established between the spray nozzle and the plate. Spray liquid is fed into the nozzle either by syringe pump or gravity force. Cone-jet meniscus is then formed once the surface tension of liquid at the nozzle exit is balanced with the applied electrical force. The breakup of the liquid jet produces droplets.

Because of the presence of DC field in the ES, generated particles are highly charged in the same polarity. The charge level on electrosprayed particles depends on the particle size and the charge distribution is usually broader than the associated particle size distribution (De Juan and Fernández de la Mora, 1997). The point-to-plate setup of ES systems makes it easy to place particles on surfaces since the collection surfaces can serve as one of the ES electrodes. For other aerosol applications which require keeping particles gas-borne, the reduction of electrical charges on electrosprayed particles is necessary in order to further transport them by carrier gas flow (because of particle loss by electrostatic and space charge effects).

Several charge reduction techniques have been applied to lower the charge level of ES-generated particles. They include the use of radioactive sources, corona discharge, and UV/soft X-ray irradiation (Chen et al., 1995; Ebeling et al., 2000; Hontanon and Kruis, 2008; Modesto-Lopez et al., 2011). Ions in both polarities (bipolar ions) are created in all the above-listed cases. Highly charged particles are mixed with bipolar ions in the cases, reducing their carried charges by the diffusion charging mechanism. Radiation sources such as ^{210}Po/^{241}Am of alpha sources have been effectively applied to produce bipolar ions. Unfortunately, the increasingly rigorous regulations in the use of radioactive material make it difficult for the future use of such sources. Even, in many countries, purchase or use of radioactive material is either under rigorous control or impossible. Corona discharge has been utilized as an alternative for bipolar ion generation. The complication in using such a technique involves the need of using high voltage power supplies with two polarities, the balance of ion concentration in both polarities (i.e., requiring the careful monitoring) and the potential of ozone production (i.e., undesirable due to its adverse heath effect). The effort in searching for alternative techniques to produce bipolar ions with symmetrical ion number concentration is thus continuous in the research community.

The modern advance in the development of UV and soft X-ray radiation sources has made them easily accessible to researchers. Photoionization by either a UV light or soft X-ray source thus provides other alternatives for bipolar ion generation. With the higher intensity than UV light and lower intensity than a hard X-ray, the soft X-ray photoionizer has been proposed as a particle neutralizer or particle charger in literatures (Kulkarni et al., 2002; Shimada et al., 2002; Han et al., 2003; Lee et al., 2005; Jiang et al., 2007a; Kim et al., 2011). When a particle-laden stream is irradiated by a soft X-ray, direct photoionization of gas molecules and particles takes place at the same time (Shimada et al., 2002; Kulkarni et al., 2002; Jiang et al., 2007a, b), resulting in generating bipolar ions and effectively reducing the charges on particles. Compared with radioactive sources with a short lifetime (such as ^{210}Po) by which the concentration of bipolar ions produced decreases as they age, a soft X-ray source performs with a stable ion production rate during its entire lifetime and generates bipolar ions with nearly the same electrical mobility (Liu et al., 1986; Lee et al., 2005). More, commercialized soft X-ray sources can be easily purchased without any user license when compared with the order of radiation sources. Soft X-ray sources are also easily operated when compared with the requirement to operate the corona discharge. The compact sizes of these soft X-ray sources make them portable, easy for the safety protection and convenient to be included in various applications where the charge reduction is essential. Note that the irradiation intensity of soft X-ray can be readily varied by changing its distance from the irradiation zone or partially blocking of its light. Soft X-ray sources are thus considered in many recent studies as a good alternative candidate for particle charge reduction (in place of radiation sources).