## Author's Accepted Manuscript

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 PII:
 S0021-8502(17)30307-5

 DOI:
 https://doi.org/10.1016/j.jaerosci.2017.12.008

 Reference:
 AS5234

To appear in: Journal of Aerosol Science

Received date:4 August 2017Revised date:16 December 2017Accepted date:19 December 2017

Cite this article as: Zhentao Wang, Lei Xia, Lin Tian, Junfeng Wang, Shuiqing Zhan, Yuanping Huo and Jiyuan Tu, Natural Periodicity of Electrohydrodynamic Spraying in Ethanol, *Journal of Aerosol Science*, https://doi.org/10.1016/j.jaerosci.2017.12.008

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#### ACCEPTED MANUSCRIPT

### Natural Periodicity of Electrohydrodynamic Spraying in Ethanol

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Abstract: Electrohydrodynamic (EHD) spraying could be regular and periodic. The cycles under different spraying modes, consisting of drop or/and jet initiation, growing, deformation, oscillation and separation, were experimentally visualized. Repetition frequencies of these cycles were obtained from offline analysis of time-resolved images captured by a high-speed digital camera with zoomed lens. It was observed that the frequency increased with electric Bond number and a sharp jump occurred when spraying transited from dripping to micro-dripping or the spindle mode. The frequency was evidently affected by flow rate in the dripping mode; however, flow rate dependency became relatively weak in the pulsated-jet mode. The frequency with a single capillary was generally higher than that in a double-capillary system. Value of the sharp jump in frequency decreased with an increase in flow rate. An electrohydrodynamic spraying classification was developed using electric Bond number  $(B_{\rm F})$ , which was a critical parameter to benchmark the transition where sharp jump in frequency occurred. When  $B_{\rm F} \leq$ 0.20, spraying was in the low frequency region with values less than a few Hertz. Spraying mode was dripping with capability to generate highly identical drops. The critical Electric Bond number  $(B_E)$  corresponding to transition from dripping to spindle mode appeared to be independent of the flow rate, and was in the range from 0.20 to 0.25. In the high frequency region, uniform and very fine drops could be produced, and spraying was in the pulsated-jet mode (jetting or whipping). The repetition frequency in different spraying modes could further our understanding on spraying behaviors and be beneficial to many practical applications. Key words: Electrohydrodynamic spraying; Periodic characteristics; High-speed microscopy; Spraying modes; Electric Bond number

#### 1. Introduction

Electrohydrodynamic spraying describes a physical process of drop or/and jet ejecting from tip of a metallic capillary subject to electrical forces. This process is frequently utilized to generate mono-dispersed drops and control the spraying modes to acquire desired jet or/and drop patterns and sizes (Cloupeau & Prunet-Foch, 1990; Jaworek & Krupa, 1999; Gañán-Calvo et al., 2013; Maktabi & Chiarot, 2016). Following the pioneer work of Rayleigh and Zeleny from late 1890 to 1920s (Zeleny, 1908, 1917, 1920), a number of experimental and theoretical studies have been performed to better understand the various spraying modes and disintegration mechanisms in the electrohydrodynamic spraying process. According to published literatures, typical spraying modes, such as dropping, micro-dripping, spindle, procession, pulsated-jet, cone jet and multi-jet mode, were classified according to the drop or/and jet geometric form and disruption pattern (Jaworek & Krupa, 1999; Verdoold, 2014). Electrohydrodynamic spraying has been utilized in a variety of applications such as paint and print spraying (Grace & Marijnissen, 1994), film coating (Miao et al., 2002), medical protein production (Gomez, Bingham, Juan & Tang, 1998), ion sources in mass spectrometry (Scalf et al., 2000), fuel injection (Gan et al., 2016), particles collection (Jaworek et al., 2013), and space propulsion (Lenguito & Gomez, 2014).

In most of the existing literatures, Electrohydrodynamic spraying was often described as random and irregular, resulting in uncontrollable drops of varying sizes and charges. However, a few qualitative observations have indicated that the electro-spraying process could somewhat be regular and periodic based on snapshot photos, handwritten sketches and stroboscopic technologies (Kim et al., 2011). By utilizing the natural periodic oscillation of an electrically stressed meniscus at a capillary tip, Sample and Bollini (1972) successfully obtained uniform

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