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## Corona discharge in electro spraying



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## ABSTRACT

The paper presents investigations of current–voltage and light emission characteristics of electro-spraying of various liquids in atmospheric air. The spectroscopic measurements have shown that the onset of corona discharge coincides with the onset of electro spraying, with the voltage increasing. The emission intensity in selected spectral lines during electro spraying depends on spraying mode, discharge power and a kind of liquid. In the specific experimental conditions in air, mainly the  $N_2$  second positive system, which is visible as violet faint light, has been recorded. The emission intensity of other gaseous species, which could be product of electro sprayed molecules decomposition or dissociation, was at very low level for the voltages applied, i.e., for glow or onset streamer discharges. From the measured light emission spectra of discharges from capillary nozzle and liquid jet, the dependence of the amplitude of selected spectral lines on capillary–nozzle voltage has been determined and it was found that this relation can be approximated by a third-degree polynomial function. This approximation has been supported by theoretical considerations. The presented results support the hypothesis that faint electrical discharges (glow, onset streamers) usually occur during and are inherent to electro spraying.

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## 1. Introduction

Electrohydrodynamic spraying, also called electro spraying, is the process of liquid atomisation by electrical forces acting on the meniscus of a liquid flowing from capillary nozzle. Due to these forces, the meniscus assumes nearly conical shape, and from the apex of this cone, a thin liquid filament flows, which finally disperses into fine droplets. Electro spray device consists usually of a capillary nozzle maintained at high electric potential and grounded counter electrode, for example, a substrate. Instead of connecting the nozzle to a high voltage supply, an extractor electrode, placed co-axially with the nozzle, is used in order to induce the electric field and steer the spraying process. In this case, the extractor is maintained at high potential while the nozzle is grounded. The electric field produced by extractor electrode induces an electric charge on the surface of meniscus and jet, which is necessary for liquid atomization. In recent years, electro spraying has attracted attention of many aerosol research laboratories due to its potential wide range of applications in nanotechnology and biotechnology, for example, for thin film deposition, novel materials synthesis, monosized nanoparticles production, fuel combustion, drug inhalation, electric propulsion for spacecraft, mass spectrometry, etc.

Due to high electric potential applied to electro spray nozzle and low radius of curvature of the capillary, meniscus and jet, the ionization processes in the gas surrounding the nozzle can be expected. These processes, leading to various forms of electrical discharges, have been extensively studied in the past [1,4,5,9,10,17–19,21,32,36–38,52,55,56,58,61,64,65,69,73,89,92], but mainly for metal electrodes. The phenomenon of corona discharge from the surface of conducting liquid, including liquid jet from electro spray nozzle or a charged droplet, has been investigated by few authors using different tools, such as, current–voltage characteristics, recording of current waveforms, steady or streak photography, mass spectrometry, light intensity measurements, or optical-emission spectroscopy. However, there are still many controversies about gas ionisation processes in the vicinity of liquid jet maintained at high electric potential, and the effect of this ionisation on electro spraying process. The results and conclusions on this subject are frequently contradictory as presented by different authors.

There is, however, no doubt that at certain circumstances, for various, mainly polar liquids, the gas in the vicinity of capillary nozzle can be ionised. The main controversy is, under which conditions, the electrical discharge can occur, and whether it can disturb the process of electro spraying, particularly in the cone-jet mode. This question is of great importance, in particular, in the case of electro spraying used for surface coating, dispersion of fragile substances, or in mass spectrometry. When electro spraying

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is used, for example, for the processing of biological or other fragile samples, ion bombardment or free radicals produced in the discharge, could damage these samples and make the measurement process useless. Moreover, the gaseous ions produced in electrical discharge could generate spurious signals at mass-spectrometer detector, leading to false results.

There are still unanswered doubts about the problem of electrical discharges from the surface of conducting liquid sprayed by electrohydrodynamic method. The key question to answer is whether the electric discharge is actually an intrinsic property of electro spraying, accompanying all electrohydrodynamic processes at the outlet of capillary nozzle maintained at high potential. The second question is up to what extent this discharge disturbs the electro spraying process. In our former papers we have studied these phenomena by photographing the glowing surface of liquid meniscus at high potential [40], and measuring the spectral lines emitted from the surface of liquid jet [38].

The aim of this paper is to investigate quantitatively the current–voltage and light emission characteristics of electro spraying for various liquids in air atmosphere. To this goal, we have measured the light emission spectra of gaseous discharges in the vicinity of capillary nozzle and determined the dependence of the amplitude of selected spectral lines on the voltage applied to the nozzle. Although our results cannot be considered as conclusive ones, they certainly provide new data for better understanding the role of electrical discharges in electro spraying process, and prove the hypothesis that faint electrical discharge (glow, onset streamers) is usually present during electro spraying.

## 2. Electro spray-corona controversies

Zeleny [94–97] was probably the first who observed corona discharge occurring during electro spraying and studied the discharge from the surface of conducting liquid (water). Later, this phenomenon has drawn attention of many investigators [25,60,72] who have contributed to better understanding this phenomenon via providing new experimental data. The interest in this subject has grown in 1970's because of electro spray application to mass spectrometry [23], but many controversies about the role of electrical discharge during electro spraying have, hitherto, not been answered unambiguously. There are two main points of view in this subject: the first is that electric discharge disturbs electro spraying process, and the second that electric discharge is inherent to electro spraying and is favourable for electro spray stabilization. The second opinion states that electrical discharge onsets at nearly the same voltage as cone jet mode commences. Both of these opinions pay only little or no attention to the type of discharge accompanying electro spraying and its effect on electro spraying mode.

The fact, which is acceptable perhaps by all authors, is that liquid can be electro sprayed in cone-jet mode if the onset potential of corona discharge is higher than the potential at which this mode is formed [14,16,29,31,33,34,79,82,87,91,95]. It will be shown in the following sections that 'corona discharge' can assume various forms, similarly to those generated from a metal point. Discharges, which lead to disturbing the process of electro spraying are pre-breakdown streamers, intermittent streamers, spark, arc, or burst pulses, whereas glow discharge or onset streamers does not disturb this process. In the former case, when those types of discharge are generated before the cone-jet mode is formed, the current–voltage characteristics are non-linear and the cone meniscus is distorted, preventing stable jet formation [6,25,50,88]. Such situation can occur for water or other polar liquids in air, and visible discharge occurs at the same or lower potential as the meniscus starts to break-up [25,91].

The destabilization of cone-jet mode by electric discharges causes changes in droplet size distribution from monodisperse to bimodal or even polydisperse [7,8,53,71], however, the exact type of discharge has not been reported in those papers. On the other hand, many authors assume that after the onset of electric discharge (presumably glow), the space charge of gaseous ions near the liquid surface stabilizes the electric field close to this surface, and further increase in supply voltage may not cause instabilities, but only causes an increase in the discharge current and glow brightness – until electric breakdown occurs [2,3,6–8,15,41,45,50,51,53,63,66,68,81,94]. This is because an increase in electric field reduces the radius of curvature of the tip of cone meniscus to one micron or below, until the electric field at this point becomes higher than a critical electric field for gas ionisation [44,79]. Moreover, the space charge of gaseous ions in the vicinity of the meniscus causes the conical meniscus to remain in equilibrium [44,51,54].

Many authors have shown that corona discharge can be favourable for the generation of cone-jet mode under conditions different from those own to "classical" Taylor cone. Tang and Gomez [88] observed a second type of stable cone-jet mode in the presence of corona discharge, and called the mode "corona-assisted cone-jet mode" (for water). Following these authors, the "corona-assisted cone-jet mode" operates at lower voltages and for flow rates one order of magnitude smaller than the normal cone-jet mode. No visible glow was evidenced by those experiments although the discharge was classified as pulseless glow or Townsend discharge [88].

On the other extreme, Jaworek and Krupa [41] have also observed a stable cone-jet mode stabilized by corona discharge in air, but for higher voltages (>25 kV), by larger electrode distances (50 mm) and for higher flow rates (100–200 mL/h). Due to large interelectrode distance only glow discharge was visible at the meniscus and jet surfaces, without transition to streamers, until electric breakdown occurred. However, large droplets, of the size in the range from 30 to 70  $\mu\text{m}$  were produced by this mode. A similar stable electro spraying mode has been observed by Borra et al. [6,8] for higher voltages. The authors called this mode the "cone-jet-glow mode". In this "cone-jet-glow mode", the size of droplets was not affected by the nozzle diameter, but mainly by the electric field at the liquid surface, which depends on the radius of curvature of meniscus and jet. Ku and Kim [54], and Kim et al. [50] also reported on a specific electro spraying mode, which they called the "corona-stabilized electro spraying mode". In that case, glow discharge was localised near the liquid surface and remained almost unchanged with increasing voltage until breakdown streamers occurred. After that, the electro spray became unstable, turning to spindle or other "irregular" modes.

From these experiments, it can be therefore concluded that the glow-stabilized cone-jet mode can be generated for higher liquid flow rates (35–200 mL/h) and higher voltages (20–30 kV) than the classical, discharge-free cone-jet mode, which is typically produced for a flow rate of 0.1–10 mL/h and by a few kV, depending on the distance between the nozzle and counter electrode. However, droplets produced by higher voltages are larger (for example, 50–150  $\mu\text{m}$  by Borra et al. [6], or 30–70  $\mu\text{m}$  by Jaworek and Krupa [41–43]) than those at lower ones.

Similar to electric discharges from a metal electrode, the type of discharge in electro spraying also depends on the polarity of electrode (capillary in the case of electro spraying). It has been found by many authors [15,82] that electro spraying at positive polarity is less disturbed by corona discharge than that at negative. The length of jet from the apex of the Taylor cone to the point of its breakdown is shorter for negative polarity than for positive one, but this difference decreases with increasing jet velocity (increasing flow rate),

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