

Acceleration of flow-focused liquid jets in the presence of a strong electric field

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Abstract. In this paper we investigate acceleration of flow-focused micro-jets under an applied electric field. Such thin and fast jets are needed, for example, for the delivery of protein crystals in X-ray free electron laser experiments. This contribution focuses on analysing the jet acceleration of liquid consisting of 50 % vol water and ethanol mixture, which was focused with nitrogen gas and subjected to 0 - 7 kV electric potential between a submerged electrode and electrode downstream of the nozzle. The analysis is based on the recently published measurements [1] and consists of automatic recognition and capture of jet geometric properties via computer vision. We discovered that micro-jets under strong electric fields are exposed to acceleration up to four orders of magnitude larger than the conventional micro-jets.

1. Introduction

Flow-focused micro-jets produced with a gas dynamic virtual nozzle are now commonly used in serial femtosecond X-ray crystallography (SFX) experiments at X-ray free electron lasers (XFELs). In these experiments, micron-sized protein crystals carried inside a liquid jet, focused by a coaxial gas, flow to the interaction region, where they are exposed to high-intensity X-ray pulses. To match high repetition rate of XFEL pulses the jets have to be fast (to ensure steady supply of pristine crystals), thin (to reduce background scattering so that diffraction from small protein crystals can be measured) and long (to prevent illuminating and damaging the nozzle with intense X-rays).

We started exploring the integration of an externally applied high voltage field with the traditional coflowing gas to make jets thinner and faster, and demonstrated stable jets [1]. The electric field should increase the acceleration of the jet, providing advantages of avoiding the need to further miniaturize nozzles to create faster and thinner jets; and to cater to the demand for a new generation of high-velocity micro jets, essential for megahertz imaging and diffraction techniques [2].

This paper builds upon our previous efforts [1], where flow-focused micro-jets under applied electric fields were produced and analysed for the first time. In that paper, the Taylor cone and the jet shape for different process parameters were described. Here, we expand this study by resolving the jet velocity profiles, gaining further insight of such jets. We assess the experimental outcomes of two stable jetting

regimes of flow-focused micro-jets under high electric potential. Our analysis shows that these jets continuously accelerate until the breakup point is reached.

2. Methods

A complete in-depth description of the experimental setup and post-processing techniques is available in [1]. Here, we provide only a brief overview. The experiment was performed using a vacuum chamber, allowing both atmospheric or near-vacuum pressures. As the liquid we used 50 % volumetric mixture of water and ethanol, while nitrogen was used as the focusing gas. The electric field was applied between an electrode submerged in the sample upstream of the nozzle and the external electrode. Experimental footage was taken by a camera mounted outside of the vacuum chamber via a long-distance working microscopic lens.

The jetting shape snapshots were processed automatically to extract the average jet diameter profile with a purpose-developed computer vision software, using Python libraries Scikit [3] and Numpy [4].

Following the assumption that there is almost no gradient of the jet velocity in the radial direction [5], the velocity was calculated via the continuity equation and using the jet diameter profile.

3. Results

As previously established [1], aerodynamically assisted Taylor cone (TC) jets exhibit an array of TC shapes based on process parameters and fluid properties. In our case, two distinct shapes were selected and analysed. The first shape closely resembles the tip streaming regime, which is characteristic of electrosprays (**Error! Reference source not found. A**). The second matches the shape typically encountered in gas flow-focusing in the jetting regime (**Error! Reference source not found. B**).

In the tip streaming regime, the emerging thin jet is of similar length as the TC, and the transition from the meniscus to the jet is abrupt. On the other hand, the transition from the TC to the jet in the jetting regime is prolonged, and the length of the liquid ligament is significantly elongated.

3.1. Tip streaming regime

In our study the parametric space for a stable tip streaming regime was very narrow. The sample flow rate was limited to 0.2 – 0.27 $\mu\text{l}/\text{min}$, the gas flow rate to 0.9 – 24 mg/min , and the applied voltage around 7 kV. Thus, all experiments were performed at 7 kV.

The jet velocities along the jet for 9 different combinations of gas and sample flow rates are shown in **Error! Reference source not found. (top)**. The starting point is defined as the meniscus-to-jet transition. Interestingly, the jet velocity increase is independent of the gas and sample flow rate combinations at a constant electric potential. A linear approximation over all data points between 0 and 20 μm (where jet breakup does not influence the velocity profiles) yields an almost perfect linear correlation ($R^2 = 0.9991$) of $v_j(x) = 0.0856x + 1.229$, where v_j denotes the axial velocity of the jet. The jet's acceleration can then be calculated from the material derivative of its velocity. Since the problem is radially symmetric and non-transient, we retain only the axial component of the convective term.

$$\vec{a} = \frac{\partial \vec{v}}{\partial t} + (\vec{v} \cdot \nabla) \vec{v} \rightarrow a(x) = \frac{v_j^2(x_2) - v_j^2(x_1)}{2(x_2 - x_1)} \quad (1)$$

Calculating the axial jet acceleration using Equation (1), we find that it is equal to $2.15 \times 10^5 \text{ m/s}^2$, or more than four orders of magnitude larger than the Earth's gravity.

3.2. Jetting regime

In the case of the jetting regime, a stable jet was observed for the following parameter ranges: liquid flow rate between 0.5 – 1.5 $\mu\text{l}/\text{min}$, gas flow rate between 0 – 25 mg/min , and applied voltage above 4 kV. At the lower applied voltage and gas flow rate, the jet was stable, but the TC was often not. It flickered and pulsed, making accurate measurements over a large dataset challenging.

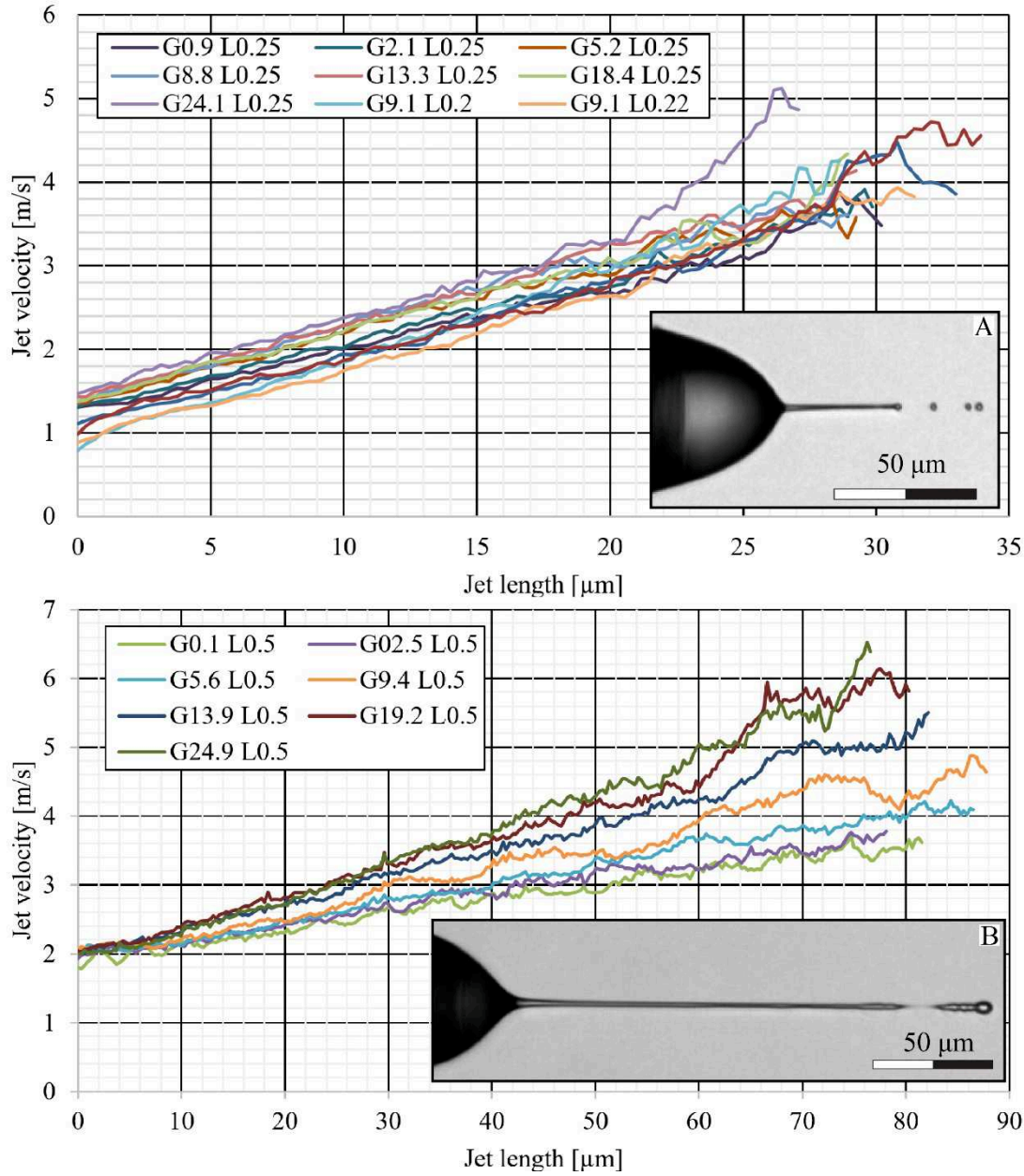


Figure 1: Jet velocity profiles for gas and sample flow rate combinations for the tip streaming regime (top) and jetting regime (bottom). A typical jet and TC shape for the corresponding regimes are shown in details A and B. Legend: G represent the gas flow rate in mg/min, and L sample flow rate in $\mu\text{l/min}$.

In **Error! Reference source not found.** (bottom), the jet velocity in the tip streaming regime is plotted, where the applied voltage and liquid flow rate were fixed at values where the TC was spatially stable. By increasing the liquid flow rate the jet velocity increased while the jet diameter remained almost unchanged. Increasing the flow rate above 1.5 $\mu\text{l/min}$ destabilised the delicate equilibrium in the TC enough to collapse the jetting regime completely. Thus, we fixed the liquid flow rate to 0.5 $\mu\text{l/min}$. To compare both regimes in a similar way, we set the applied voltage at 5 kV, which allowed the gas flow vary between 0 and 25 mg/min.

Analogous to the tip streaming regime, the velocities along the jet in the jetting regime increase linearly except that here they also depend on the gas flow rate. In particular, increasing the gas flow rate leads to an increase of jet acceleration. Using Equation (1), we could determine the limiting accelerations

of the jet. Linear approximations were fitted up to a jet length of 60 μm , where jet breakup does not influence the velocity profile. For low gas flow rate, case G0.1 L0.5 the calculated velocity is $v_j(x) = 0.0222x + 1.8943$, with $R^2 = 0.976$. For high gas flow rate, case G24.9 L0.5, the velocity profile is equal to $v_j(x) = 0.0493x + 1.8332$, with $R^2 = 0.994$. The calculated accelerations for the two limiting cases are thus $5.68 \times 10^4 \text{ m/s}^2$ and $1.68 \times 10^5 \text{ m/s}^2$, respectively.

4. Conclusion

In this paper, we present the analysis of the experimental acceleration data of flow-focused micro-jets under an applied electric field together with a short overview of the experiment and post-processing techniques. We observed and studied two jetting regimes and showed that the velocity increases over the longitudinal axis of the jet at a constant rate. The acceleration was calculated by fitting linear functions over the velocity profiles, which matched with very high coefficients of determination. In the jetting regime, the acceleration depends on the gas flow rate, while in the tip streaming regime it does not. In the latter, the gas flow rate mainly served as a stabilizer for the TC.

Noting that the velocity profile for conventionally focused liquid jets eventually flattens, one can appreciate the added focusing force, which continues to accelerate the liquid ligaments until the very end, where it is up to four orders of magnitude higher than the acceleration g due to gravity.

5. Acknowledgements

Funding for this research is provided by the Centre for Free-Electron Laser Science (CFEL) under the project: Innovative methods for imaging with the use of x-ray free-electron laser (XFEL) and synchrotron sources: simulation of gas-focused micro-jets, and Slovenian Grant Agency (ARIS) within Program Group P2-0162 and Project J2-4477. This work is also partly supported by the Cluster of Excellence ‘CUI: Advanced Imaging of Matter’ of the Deutsche Forschungsgemeinschaft (DFG) – EXC 2056 – project ID 390715994.

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