ELECTROHYDRODYNAMIC COULTER COUNTING

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ABSTRACT

A new paradigm of Coulter counting is developed using electrohydrodynamic liquid jets in lieu of solid state pores as the sensing aperture. The electrohydrodynamic jet was successfully used to detect micron-sized particles through resistive pulse sensing. Similar to conventional Coulter counting, the relative current change is proportional to the particle-to-jet volume ratio. Compared to its conventional counterpart, the new Coulter counting technique with a liquid sensing aperture has a major advantage of being non-clogging to impurities and agglomerates.

KEYWORDS: Coulter counting, particle detection, electrohydrodynamic jet, cone-jet

INTRODUCTION

This paper reports a new paradigm of Coulter counting (resistive pulse sensing) using electrohydrodynamic liquid jets in lieu of solid pores as the sensing aperture (Figure 1a). The Coulter counter detects and characterizes particles by the modulation of electrical current through a small fluidic aperture (Figure 1b).[1] Commercial Coulter counters and state-of-the-art nanopores are typically based on solid-state apertures which have fixed diameters and suffer from clogging. The electrohydrodynamic Coulter counting technique reported here has the potential to circumvent these drawbacks in dynamic range and life span by exploiting the cone-jet transition to produce tunable liquid jets down to 10 nm,[2] and using these thin jets emitted from much larger off-the-shelf nozzles as the sensing aperture.

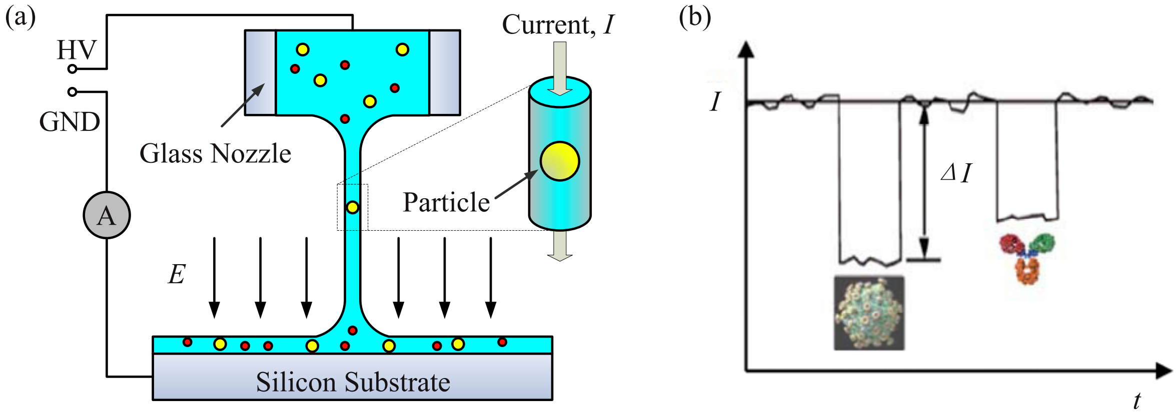

Figure 1: Schematic of (a) an electrohydrodynamic Coulter counting system in which particles are detected by (b) the modulation of electrical current (b) flowing through an electrified liquid jet.

EXPERIMENTAL

The experiments setup is schematically shown in Figure 1a. A steady *double*-cone-jet was generated by applying a high voltage between a 100 µm-ID glass nozzle (New Objective) and a conductive silicon substrate. The actual setup was rotated 90 degrees with the jet being horizontal, so that a steady-state thin film could be set up on the silicon substrate by gravity. Polystyrene particles of 3, 5, or 7 µm diameter (Thermo Scientific) were washed with DI water by three cycles of centrifugation, and subsequently dispersed in ethylene glycol doped with salt to a conductivity of 3, 10 or 30 µS/cm. The flow rate was controlled by a syringe pump. The current through the electrohydrodynamic jet was measured with an oscilloscope, and synchronized high-speed imaging was captured by a high speed camera (Phantom v7.3).

RESULTS AND DISCUSSION

A proof-of-concept for electrohydrodynamic Coulter counting is shown in Figure 2. Polystyrene particles of 7 µm diameter were dispersed in ethylene glycol with a conductivity of 10 µS/cm. When the colloidal dispersion was electrified at the tip of a 100 µm-diameter glass nozzle, a thin jet with a diameter of approximately 12 µm issued from the nozzle. A liquid bridge with a length of 140 µm formed between the nozzle (held at 4 kV) and the counter electrode (ground); see Figure 2b. The current through the electrohydrodynamic jet was measured with an oscilloscope (Figure 2a). The oscillogram in Figure 2a indicated the passage of a non-conducting particle through the conducting jet, which was confirmed by synchronized high-speed imaging in Figure 2b. For the representative case shown in Figure 2b, when the particle entered the jet at time t_1, the non-conducting particle started to block the current flowing in the jet (t_2); the current depression peaked at t_3, and relaxed backed to its equilibrium value around t_4 when the particle exited the jet. The jet continued to oscillate for a few cycles after the particle passage. The relative current change was experimentally measured as

$$\frac{\Delta I}{I} = \frac{I(t_1) - I(t_3)}{I(t_1)}. \qquad (1)$$

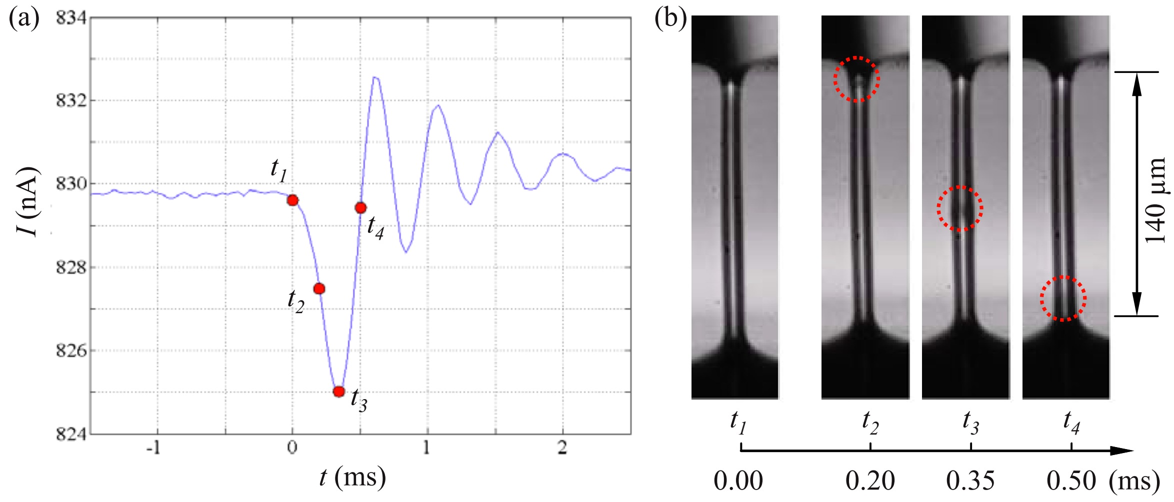

Figure 2: (a) Oscilloscopic and (b) microscopic measurements confirming the passage of a 7 µm particle in an electrohydrodynamic jet with a diameter of 12 µm and a length of 140 µm. The dashed circles in (b) indicate particle location at times specified in (a). The jet continued to oscillate for a few cycles after the particle passage.

Using electrohydrodynamic Coulter counting, particles with diameters (d_p) of 3, 5 or 7 µm were successfully detected using ethylene glycol jets with conductivity of 3, 10 or 30 µS/cm (Figure 3). The jet diameter (d) ranged from 7 µm to 13 µm, and the jet length L was kept at 140 µm. The double-cone-jet enabled a liquid sensing aperture (Figure 2b) smaller than commercial Coulter apertures, the smallest off-the-shelf aperture being 20µm in diameter.[3] Similar to conventional Coulter counting, when a non-conducting particle with a volume V_p passes through the electrohydrodynamic jet with a volume V_j, the relative change in current I scales approximately as the particle-to-jet volume ratio[4],

$$\frac{\Delta I}{I} \sim \frac{V_p}{V_j} = \frac{2d_p^3}{3Ld^2}. \qquad (2)$$

This scaling law holds when Ohmic current dominates and the particle is not too large compared to the diameter and length of the sensing jet. Results from electrohydrodynamic Coulter counting are consistent with this scaling law (Figure 3), for volume ratios and liquid conductivities each spanning one order of magnitude.

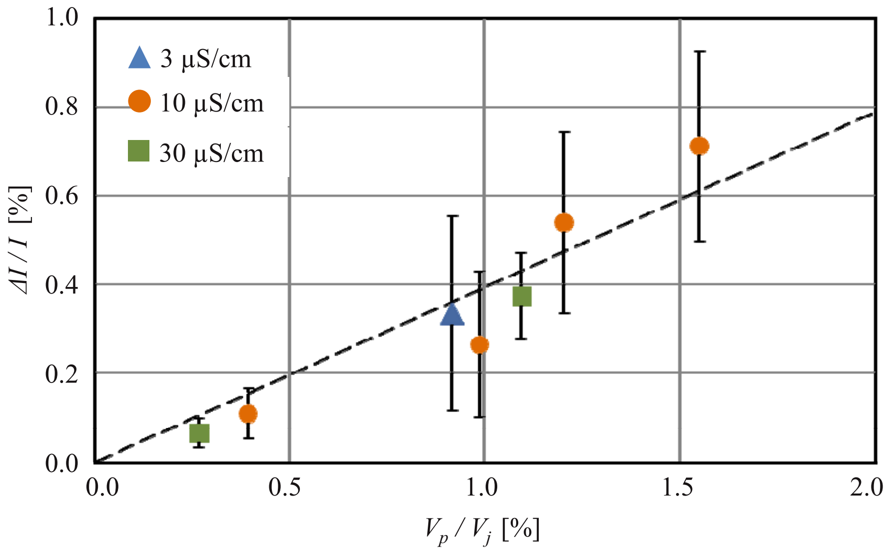

Figure 3: The relative change of electrical current is approximately proportional to the particle-to-jet volume ratio. Particle diameters (d_p) of 3, 5 or 7 µm were detected using ethylene glycol jets with a conductivity of 3, 10 or 30 µS/cm. The jet length was 140 µm. The applied voltage was 4 KV for the conductivity of 3 µS/cm and 10 µS/cm, and 2.7 KV for the conductivity of 30 µS/cm. The flow rate ranged from 100 µL/h to 300 µL/h.

A main advantage of the electrohydrodynamic Coulter counting technique over its conventional counterpart is that, by introducing an adaptive liquid aperture, the clogging problem for solid-state apertures is eliminated. An example is shown in Figure 4 with the passage of a large debris 30 µm in length in a 14 µm-diameter jet .Even though the longitudinal axis of the debris turned from streamwise (0.1 ms) to spanwise (0.3 ms), the debris passed through the electrohydrodynamic jet without difficulty. More importantly, the electrohydrodynamic jet system quickly relaxed back to its equilibrium configuration (0.5 ms). Such a large debris would have been detrimental to a solid state pore with a diameter only half of the debris size. The clogging problem is likely the main limiting factor for commercial Coulter counters which use solid-state pores (the minimum diameter of off-the-shelf apertures is 20 µm[3]).

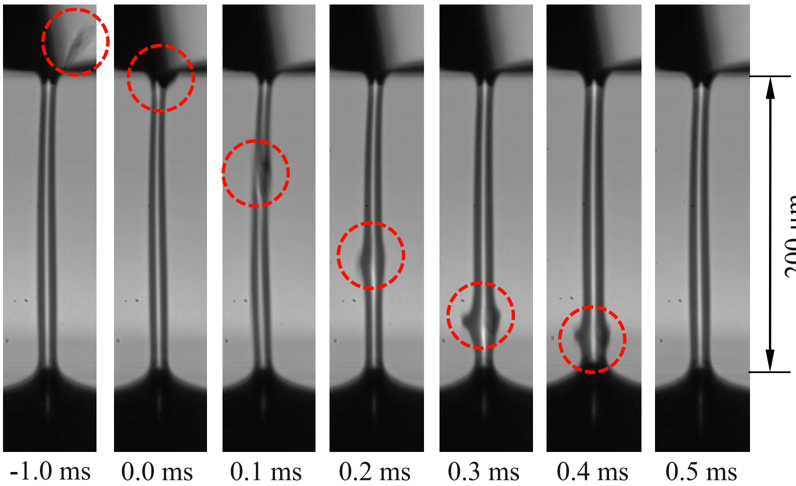

Figure 4: The passage of a large debris twice the size of the equilibrium jet diameter. The dashed circles indicate the debris location. The ethylene glycol jet with 14 µm diameter and 200 µm length was established at 5 KV. When a large debris with a length of approximately 30 µm entered the jet, the double-cone-jet self adjusts to pass the debris in approximately 0.5 ms, despite that the particle turned transverse to the jet at 0.3 ms. After the passage of the debris, the jet quickly relaxed back to its equilibrium shape at 0.5 ms.

CONCLUSION

In this paper we demonstrated the proof of the concept for Coulter counting using an electrohydrodynamic liquid jet. Micron-sized particles were detected with the relative change in resistive current linearly proportional to the particle-to-jet volume ratio. We have essentially reproduced a conventional Coulter counter, but without solid channels/pores which are prone to clogging. Electrohydrodynamic Coulter counting offers a more affordable means of producing tunable, non-clogging “nanopores” and “micropores” with a diameter between 10 nm and 10 µm, a range that is not reachable with conventional aperture manufacturing process and too costly with most reported processes for artificial nanopores. Work is under way to utilize micron- and submicron-scale electrohydrodynamic jets, in which surface charge convection may dominate over bulk charge conduction,[5] to count nanoparticles and macromolecules.

ACKNOWLEDGEMENTS

The work is supported by an NSF CAREER Award (CBET-08-46705).

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