Induced flow inside a droplet by static electrical charge

Tapan Kumar Pradhan¹, Theneyur Narayanaswamy Banuprasad², M S Giri Nandagopal¹ and Suman Chakraborty¹,²,* ¹Department of Mechanical Engineering, Indian Institute of Technology Kharagpur, Kharagpur, West Bengal 721302, India ²Advanced Technology Development Centre, Indian Institute of Technology Kharagpur, Kharagpur, West Bengal 721302, India E-mail: suman@mech.iitkgp.ac.in

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Abstract
Introducing controlled fluid motion in a droplet turns out to be of outstanding scientific importance, hallmarkd by a plethora of applications ranging from engineering to biology. While internal mechanisms such as interfacial tension or buoyancy-driven dynamics may trigger fascinating flow structures inside a droplet, controllability of the same without external forcing remains questionabile. On the other hand, in an electrically forced environment, complex fabrication steps and special choices of the ionic liquid are often demanded. Circumventing these limits, here we bring out a new method of flow manipulation inside a sessile droplet by simply deploying a static charge produced by the triboelectric effect. This is physically actuated by charge transfer between the two lateral electrodes within which the droplet is entrained, triggering a strong ionized air current. The flow inside the droplet is generated due to the shear exerted at the interface by the charge-induced ionized airflow around the droplet, a paradigm that has hitherto remained unexplored. The strength of the fluid flow can be controlled by adjusting the supplied static charge. Such unique controllability without sacrificing the physical simplicity opens up new possibilities for flow manipulation in a multitude of applications ranging from droplet microreactors to digital microfluidics.

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(Some figures may appear in colour only in the online journal)

1. Introduction
Actuating, manipulating, and controlling fluid motion in a droplet turns out to be of emerging scientific importance, as attributed to a multitude of applications ranging from engineering to biology. The fundamental premises that control their functionalities include efficient mixing on one side and arresting unwarranted deposition of particulate matter on the other [1–6]. In evaporating droplets, the flow mainly occurs by capillary action [1, 7], surface tension gradient [8–10], or buoyancy forces [11–15]. However, because of their limited controllability, several external actuation mechanisms, including thermal energy [16, 17], have been employed to generate desired flow characteristics within a droplet. Vapor-mediated droplet interaction has also been used to induce or alter flow patterns inside evaporating droplets [12, 18–21].

In recent years, electrical forcing has emerged to be a preferentially alternative mechanism for flow manipulation inside tiny droplets, primarily due to the facilitated integrability of a
droplet with electrical or electronic circuitry from the emerging perspective of lab on a chip technology [22–28]. Despite their obvious merits, reported techniques on electrically driven flow manipulation inside droplets suffer from a number of compelling deficits. In particular, these processes inevitably demand specialized fabrication procedures for the electrodes and surface modification. In addition, it often necessitates changing the composition of fluid to modify the ionic strength, losing the freedom of being applicable to the desired applications on demand.

Overcoming the above constraints, we report here an approach of flow manipulation inside a sessile droplet by simply harnessing a static charge produced by the triboelectric effect. We realize this by charge transfer between the two lateral electrodes within which the droplet is entrained, without demanding any on-chip electrode fabrication. Fluid motion inside the droplet is generated due to the interfacial shear exerted by the charged-induced ionized airflow incipient to the droplet. The strength of the resultant fluid motion can be controlled at will, by altering the supplied charge. The simplicity of the technique, furthered by its elegant controllability, renders this ideally suitable for a wide gamut of applications ranging from droplet microreactors to digital microfluidics.

2. Experimental method

A water droplet of volume (∅) 0.7 µl was placed on a clean glass surface forming an apparent contact angle of ∼36° and a contact line diameter of 2.1 mm. The droplet was placed between two copper electrodes of 0.5 mm diameter having a separation distance of 8 mm. The electrodes were physically separated from the droplet with an air gap on both sides (figure 1). Static charge generated by a Van De Graff generator (VDG) was applied to induce flow inside the droplet. One of the electrodes (electrode 1) was connected to the VDG and the other electrode (electrode 2) was connected to the ground as shown in the schematic. VDG works on the principle of triboelectric effect where friction between two dissimilar materials creates an unbalanced charge on each material. In the present setup (figure 1), friction between the rubber belt and the aluminum plate (present near the lower roller) generates a negative charge on the belt. The negative charge from the belt was transferred to the dome which was then transferred to the droplet by a connecting wire. The electrons from the VDG flow through electrode 1, droplet and the ground electrode (electrode 2). The current (I) in the circuit and the voltage between the two electrodes (∆V) were measured by two multimeters as shown in figure 1. The effect of charge on the flow was assessed by regulating the current over a range of 1 µA – 4.7 µA by adjusting the roller speed.

The velocity of fluid inside the droplet was measured by the micro-particle image velocimetry (µPIV) technique. Fluorescent polystyrene particles of diameter 4 µm were added to the droplet as tracer particles which were illuminated by a mercury lamp. A high-speed camera (Phantom V15) attached to an inverted microscope (Olympus IX71) was used to capture the images at 200 frames per second. Each image has a field of view of 2.65 mm × 2.65 mm. The images were processed by PIVLab [29] to get a velocity field using the FFT algorithm with a 64 × 64 pixels interrogation area. The tiny seeding particles show Brownian motion which is random in nature. The error caused by the Brownian motion was eliminated by averaging the result over five measurements.

3. Results and discussion

When no charge is supplied to the electrode, the internal convection shows a very small flow strength of the order of 1 µm s⁻¹ [18]. The fluid moves unidirectionally towards the contact line which is caused by capillary action to compensate the higher evaporative flux at the contact line [1]. When the charge is applied to the electrode (electrode 1 in figure 1) connected to the VDG, the fluid inside the droplet shows strong convection as shown in figure 2. It shows two circulating loops (see supplementary movie 1) having maximum flow strength of the order of 2.5 mm s⁻¹. No flow is observed when the circuit is disconnected from the ground. Flow is only observed when the charge continuously flows from the VDG to the ground electrode through the droplet.

The negative charge from the VDG is carried to the electrode 1 by a connecting wire. Electrode 1 carries unbalanced electrons and tries to get neutralized. Electrode 2 which is connected to the ground is the easiest path to get neutralized. Hence, the electrons tend to move from electrode 1 to the ground electrode 2. During the process, a collision between the electrons and the air molecules surrounding electrode 1 creates a plasma of negatively charged air. Air molecules acquire negative charge due to electron capture ionization when the molecules are subjected to electron impact [30, 31]. The plasma formation near the electrodes is presented in figure 3(a). The image of the plasma was captured in a closed dark chamber using a digital single-lens reflex (DSLR) camera keeping a high exposure time (15 s). The intensity of the plasma is very less due to low current. The negatively charged air molecules near electrode 1 accelerate towards the ground electrode (electrode 2) to get neutralized which is depicted in figure 3(a). It creates a flow of negatively charged air from electrode 1 to electrode 2, maintaining the continuity of charge flow in the circuit. Static electricity works differently than the normally used DC and AC supplies. In such a low voltage (below the breakdown voltage) of normally used DC and AC, the charge cannot pass between the two electrodes having an air gap of 8 mm. In the case of static electricity, the charged molecules can be transferred between these two electrodes due to a small potential difference similar to electron acceleration in a cathode ray tube.

The schematic of the induced flow inside the droplet using static charge is presented in figure 3(c). When the droplet is placed between the two electrodes as shown in figure 3(b), the droplet experiences the airflow between the electrodes. The flow of air between the two electrodes is confirmed by the smoke flow visualization using an incense stick (see supplementary movie 2). Smoke from the incense stick drifted in the direction of airflow between the two electrodes. The smoke
Figure 1. Schematic of the experimental setup for inducing flow inside the droplet and micro-PIV measurement.

Figure 2. Velocity field inside the droplet near the substrate at $Z = 50 \mu m$ from the substrate surface.

shows an airflow from the electrode connected to the VDG to the ground electrode. The surrounding air current exerts a shear stress at the liquid–air interface of the droplet. The shear stress at the interface induces circulating flow inside the droplet [32] which is well explained in figure 3(c). The flow pattern inside the droplet strongly depends on the airflow pattern near the droplet. Any disturbance in the surrounding air significantly affects the internal convection of the droplet.

The water droplet evaporates at an ambient temperature ($T_\infty$) of 25 °C and relative humidity ($\Omega$) of 0.6. The evaporation time ($t_f$) of the sessile droplet for a small contact angle without any applied charge is given by [33]:

$$t_f \approx \frac{\pi \rho R^2 \theta_0}{16D_v (C_s - C_\infty)}.$$

The droplet has a constant contact line radius ($R$) throughout the evaporation process which is equal to 1.05 mm. The value of the coefficient of diffusivity ($D_v$), saturated vapor concentration ($C_s$), ambient vapor concentration ($C_\infty = \Omega C_s$) and the density of water ($\rho$) are equal to $2.42 \times 10^{-5}$ m$^2$ s$^{-1}$ [34], $2.3 \times 10^{-2}$ kg m$^{-3}$ [34], $1.38 \times 10^{-2}$ kg m$^{-3}$, and 997 kg m$^{-3}$ [34] respectively. The evaporation time ($t_f$) of the water droplet calculated from the above equation is equal to 10.2 min without any applied charge. When the charge is applied, it may add thermal energy due to joule heating or plasma to the droplet affecting evaporation dynamics and internal flow. The maximum possible thermal energy absorbed by the droplet from the applied charge is equal to the total electrical energy lost in the process, which is given by, $W = \Delta V I t_e$. Here, $t_e$ is the evaporation time of the droplet when the source of energy for evaporation comes only from the applied static charge. The thermal energy required for complete evaporation of the droplet is given by, $H = m h_{fg}$. Here, $m$ is the mass of
Figure 3. Plasma formation near the electrodes and flow mechanism. (a) Depiction of ionized air molecules between the two electrodes. (b) Droplet placed between the two electrodes with charge transfer between the electrodes. (c) Depiction of the mechanism of induced flow.

The value of $t_e$ is equal to 443 min. The evaporation time of the droplet due to only electrical power from VDG is much more than the normal evaporation time of the droplet evaporating in a room environment extracting energy from the substrate. Hence, the effect of the heat of plasma or electrical energy on the evaporation of the droplet is neglected.

VDG has disadvantages for many applications due to its low current. However, low current is advantageous in the present application, because it leads to low total energy, keeping the droplet unaffected. Hence, it avoids rapid evaporation of the droplet. Due to very low thermal energy, the thermal effect like thermal Marangoni convection on the fluid flow inside the droplet can be neglected.

Other possible flow mechanisms caused by electrical energy may contribute to the observed flow are electrohydrodynamics, electrophoretic, and electroosmosis and electrophoresis. Electrohydrodynamics (EHD) causes quadratic flow structure inside the droplet [22], whereas only two circulating loops were observed in the present experiment. Hence, it is expected that the flow is not due to the EHD effect. Electrophoretic effect is due to electric body force caused by Joule heating. However, from the above time scale analysis of evaporation, it is found that the supplied electrical energy hardly causes any thermal effect. An approximate velocity magnitude for electro-osmotic flow can be given by the Helmholtz–Smoluchowski equation, $u_{eo} = \frac{\varepsilon \zeta E}{\mu}$. The value of electric permittivity of water ($\varepsilon$), viscosity of water ($\mu$), and zeta potential of water-glass interface ($\zeta$) are equal to $6.96 \times 10^{-10}$ F m$^{-1}$ [34], $8.9 \times 10^{-4}$ Pa.s [34], and $-62.2$ mV [35] respectively. The voltage across the electrodes is 32 V which gives average electric field strength ($E$) of $4 \times 10^3$ V m$^{-1}$. The velocity scale is equal to 0.2 mm s$^{-1}$ which is negligible as compared to the observed value. Polystyrene particles (diameter = $4 \mu$m) are used in the fluid for micro-PIV measurements which may show electrophoretic movement in the electric field. Considering the particle size to be much higher than the EDL, the electrophoretic velocity can be given by, $u_{ep} = \frac{\varepsilon \zeta p E}{\mu}$. The approximate value of zeta potential of the polystyrene particle-water ($\zeta_p$) is equal to $-79$ mV [36]. The characteristic electrophoretic speed is in the tune of $0.25$ mm s$^{-1}$, which is significantly less as compared to the observed flow velocity. In addition, by varying the salt concentration of the droplet, no significant alteration in the flow structure is observed. These observations implicate that electroosmotic, electrophoretic and electrohydrodynamic effects are inconsequential in dictating the flow behavior. Rather, the shear-mediated interactions with the externally ionized airflow govern the essential physics of interest.

We next study the effect of charge on the flow strength at different charge flows which is presented in figure 4. The flow strength is characterized by the area average of velocity

Figure 4. Variation of flow strength inside the droplet with charge flow.
magnitude. With an increase in charge flow in the circuit, the negatively ionized air molecules increase in number. This leads to an increase in the flow rate of air surrounding the droplet. Hence, the flow strength inside the droplet increases with an increase in charge flow between the two electrodes. It is also observed from the plot that the uncertainty (error bar) increases with an increase in current flow. It is expected that the increase in air flow rate increases the fluctuation in the air velocity and becomes more chaotic.

The flow induced by static electricity can also be used as a method to manipulate the deposition pattern of a drying droplet. The deposition pattern of a drying droplet strongly depends on the flow pattern inside the droplet [37]. The coffee ring deposit of a droplet of fountain pen ink is shown in figure 5(a) which occurs due to the unidirectional capillary flow caused by evaporation [1]. When the droplet is dried with the simultaneous application of static charge to the droplet, the coffee ring breaks and forms a deposition throughout the droplet base area (figure 5(b)). The breaking of the coffee ring occurs due to the flow circulation (figure 2) caused by the applied charge. The circulation suppresses the unidirectional capillary flow and disturbs the particle convection towards the contact line.

4. Conclusion

We have suggested a simple method of inducing controlled flow inside a droplet, mediated by static electric charges. The method essentially features the interfacing of an external electrical system with a droplet-based fluidic system, mediated by the shear stress of surrounding air. This imposes no fabrication constraints on the fluidic system, and puts no limitation on the nature of the fluid. Also, there is no change in the composition of fluid during the process. Biological samples can also be used in the droplet as there is no heating and composition change during the flow manipulation.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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Conflict of interest

All the authors declare no competing financial interest.

ORCID iD

Suman Chakraborty https://orcid.org/0000-0002-5454-9766

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