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Interplay of electro-thermo-solutal advection and internal electrohydrodynamics governed enhanced evaporation of droplets

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The article experimentally examines and theoretically establishes the influence of electric field on the evaporation kinetics of pendant droplets. It is observed that the evaporation of saline-pendant droplets can be augmented by the application of an external alternating electric field. The evaporation behaviour is modulated by an increase in the field strength and frequency. The classical diffusion driven evaporation model is found insufficient in predicting the improved evaporation rates. The change in surface tension due to field constraint is also unable to explain the observed physics. Consequently, the internal hydrodynamics of the droplet is investigated through particle image velocimetry. The electric field is found to induce enhanced internal advection, which improves the evaporation rates. A scaled analytical model is proposed to quantify the role of internal electrohydrodynamics, electro-thermal and electro-solutal effects. Stability maps reveal that the advection is caused nearly equally by the electro-solutal and electro-thermal effects within the droplet. The model is able to illustrate the influence played by the governing thermal and solutal Marangoni number, the electro-Prandtl and electro-Schmidt number, and the associated electrohydrodynamic number. The magnitude of the internal circulation can be predicted by the proposed model, which validates the proposed mechanism.

1. Introduction

Research on fundamental and applied microfluidic systems have received significant attention in recent times. They hold promise in bio-medical engineering and healthcare [1], automotive industry [2–4], the manufacturing sector [5], detection and quality control and so forth. Droplets are an important microfluidic paradigm which is responsible for the thermofluidic efficiency of several macroscale and microscale thermofluidic systems. Performance of agro-spraying, medicinal sprays and nebulizers [1], coating and spray-painting technologies [6,7], spray cooling systems, automotive engines and gas turbine systems, etc., are strongly dependent on the thermofluidic behaviour of sprays and their constituent droplets [8–10]. Thereby, understanding of the hydrodynamics, thermal and phase interaction of droplets is essential for further improvement and efficient design of the existing system and towards development of new systems.

According to the prevalent regimes, droplets are generally classified into the sessile and the pendant droplets. The evaporation mechanisms of the two types of droplets are grossly different. Evaporation dynamics of complex fluid droplets is a phenomenon which actively involves important hydrodynamics, thermal and species transport characteristics within the droplet. Studies [11–13] have established two distinct modes of evaporation in case of sessile droplets, *viz.* the constant contact angle and constant contact radius modes. Sessile droplet evaporation was studied in a pioneering work by Picknett & Bexon [11], where the dynamics of the contact line had the major focus. The role of the contact line dynamics was further studied thoroughly by Shanahan [14]. The roles of relative humidity, thermal gradient across the substrate and the geometry of the droplet on the evaporation kinetics have also been studied [15].

Of late, a shift of focus has occurred in the field of droplet research, towards the dynamics and behaviour of pendant droplets. Pendant droplets are independent of surface interactions and hence the dynamics of constituent droplets in sprays and atomization processes can be modelled more accurately through the pendant approach. A pioneering work by Godsave [16] introduced the classical D^2 law for the evaporation of pendant droplets. Studies [17,18] have shown that evaporation characteristic of pendant droplets are modulated by tuning the interfacial properties employing salts, or surfactants or colloidal particles. Studies on the role of electromagnetic body forces on pendant droplet behaviour have also been reported [19]. The enhanced evaporation dynamics due to the presence of ferro-convection within droplets of paramagnetic salts in the presence of a magnetic field has been studied [20]. Electric body force also influences the dynamics and transport processes in droplets. Laroze [21] discussed the role of body couple and Kelvin force that are encountered during evaporation of pendant droplet in an electric field environment. Arrest and retardation of evaporation rate from pendant droplets in specially orientated electric field have also been reported [22].

Electrohydrodynamics of droplets has received attention owing to the rich physics involved and the applications, especially in micro-flow devices. Initial works discussed the distortion and deformation of the droplet under the influence of the electric field [23–28]. Theoretical predictions of the different modes of droplet deformation using the classical Young–Laplace, Maxwell, Stokes and Laplace models have also reported [29]. Electrohydrodynamics of droplets find importance in microfluidic bio-medical devices, like therapeutic cell sorter, drug testing chips, gel electrophoresis, platelet segregation, etc. Several intriguing electrohydrodynamic phenomena and applications, like electro-Leidenfrost effect, electro-wetting, electro-spinning, electro-slip-stick phenomenon and electro-propulsion have been explored [30–34]. The electric field also leads to the formation of a Taylor cone at microfluidic junctions and maybe used to generate well-dispersed emulsions [35].

Electro-coalescence of droplets due to an alternating field has been studied in the literature [36]. Hase *et al.* [37] reported that a direct electric field on two identical droplets placed between electrodes can generate droplet oscillation behaviour. Electrohydrodynamics actuated microfluidic jets towards cancer cell diagnostics has also been explored [38]. Evaporation or vaporization of droplets are involved in important applications, like bio-medical systems (spray

based medicines, inhalers, nebulizers, blood culture, DNA patterning, etc.), micro-manufacturing and processing like micro-lithography [5], ink-jet printing and in HVAC systems. The efficiency of internal combustion engines [2–4] also depend on droplet evaporation kinetics. Droplet evaporation is significant for efficient use of pesticides, insecticides, fumigants [1,6] and in microfluidic organ-on-chip devices [39].

A thorough survey of the literature shows that both the evaporation dynamics of droplets and droplet electrohydrodynamics has important implications and physics involved. Reports show that solvated ions enhance evaporation characteristics of aqueous pendant droplets [17,20,22]. This article experimentally and analytically explores the evaporation kinetics of a conducting fluid pendent droplet and its associated electrohydrodynamics. Classical diffusion dominated evaporation models are found to be inadequate in predicting the enhanced evaporation rates under electric field stimulus. Additionally, the change in surface tension and thermo-physical properties were also insignificant towards enhancing the noted evaporation. Consequently, flow visualization employing particle image velocimetry (PIV) was performed to quantify the internal electrohydrodynamics and its role in augmenting the evaporation has been discussed. A scaled analytical model to include the effects of electro-thermal and electro-solutal advection in the evaporation kinetics has been presented. The model predictions are observed to match well with the experimental findings. The present study may find strong implications in electrohydrodynamics modulated transport phenomena in droplets.

2. Materials and methodology

The present experiments were performed using a customized set-up (refer figure 1). It consists of a digitized precision droplet dispensing mechanism (Holmarc Opto-mechatronics, India) which can generate droplets accurate to $0.1\ \mu\text{l}$. A glass chromatography syringe (of capacity $100\ \mu\text{l}$) with a flat end stainless steel needle (22 gauge) is used for dispensing the droplets. Pendent droplets of volume $15 \pm 0.5\ \mu\text{l}$ were suspended from the tip of the needle. The evaporation process has been recorded using a charge-coupled device (CCD) camera, with a microscopic lens assembly (Holmarc Opto-mechatronics Pvt. Ltd.), at 10 fps and resolution of 1280×960 pixels. A light emitting diode array (DpLED, China) was used for illumination. The set-up was enclosed within an acrylic chamber to eradicate fluctuations due to atmospheric conditions. Digital hygrometer and thermometer were used to measure humidity and temperature near the droplet (using a sensing probe approx. 5 cm away from the droplet). For the whole set of experiments, the temperature was $28 \pm 3^\circ\text{C}$ and humidity was $62 \pm 3\%$.

Figure 1 also illustrates the arrangement of the electrodes (two stainless steel sheets, $20\text{ mm} \times 10\text{ mm}$). The electrodes are placed (4 mm apart) to impart the electric field horizontally across the droplet (inset figure 1). The droplet is positioned at the centre of the electrodes to ensure minimal distortion of the field. The electrodes are connected to a programmable AC power source (Aplab, India) of controllable frequency range of 50–500 Hz with 0.1 Hz resolution, and field strength ranging from 0 to 275 V, and with 0.1 V resolution. Experiments with a water droplet, at the zero-field, but with the electrodes in a position were performed as control experiments and validated against reports in the literature [17,20,37] and good agreement was noted. Thereby, solely the presence of the electrodes was ascertained to be unimportant to the evaporation process.

The selection of salt solution has been made based on previous experiments by the present authors [17,20]. Solutions of anhydrous sodium iodide (NaI) (Sigma Aldrich, India) of concentrations 0.05, 0.1 and 0.2 M were employed. The images of the evaporation process were processed in the open source software IMAGEJ. The droplet shape was fit to the Young–Laplace equation and the instantaneous equivalent spherical diameter for the pendant droplet was deduced. PIV has been done to qualitatively and quantitatively determine the internal hydrodynamics of the evaporating droplet using neutrally buoyant fluorescent polystyrene seeding particles (diameter approx. $10\ \mu\text{m}$; Cospheric LLC, USA). A 532 nm continuous wave laser (Roithner GmbH, Germany) and a plano-convex cylindrical lens have been used for

Figure 1. Schematic of the experimental set-up. It consists of (a) a CCD camera mounted on a two-axis movable frame and attached with a microscopic lens assembly (b) solid-state laser module mounted on the stand, with plano-convex cylindrical lens (not illustrated) for generating light sheet (c) programmable AC power source with frequency and field strength controller (d) data acquisition computer (e) precision droplet dispenser mechanism and backlight controlling module (f) droplet dispensing mechanism controller and programmer and (g) power source and control unit for the laser. (h) The inset (top-left corner) shows the zoomed-in views of the arrangement of electrodes and the needle of the glass syringe. (Online version in colour.)

generating a light sheet (approx. 0.5 mm thick). The light sheet has been focused at the droplet mid-plane for the two-dimensional velocimetry.

The laser power employed for the velocimetry is approximately 10 mW. The PIV is conducted during the initial stage of evaporation (within the first 5 min) so that the evaporation induced increase in salt concentration does not lead to errors. A CCD camera is used to capture the PIV frames at 30 fps, at resolution of approximately $150 \text{ pixels mm}^{-1}$, and for 90 s. The open source code, PIVlab, has been employed for the analysis. A cross-correlation algorithm with four-pass window interrogation, with interrogation windows of 64, 32, 16 and 8-pixel widths has been used. Standard noise reduction algorithms have been used to improve the signal-to-noise ratio. The velocimetry is performed for 1000 consecutive frames, and the spatially averaged velocity for each frame is determined. The spatio-temporal averaged velocity field for all the frames is also determined, and this is the mean velocity field within the droplet.

3. Results and discussion

(a) Evaporation characteristics under the influence of an electric field

The evaporation of water and salt solution droplets under zero-field have been used as the control case. The presence of different electric field constraints across the pure water droplet has no influence over the evaporation dynamics. The evaporation behaviour of water droplet is observed to follow D^2 law [16]. Considering the instantaneous droplet diameter D , the initial droplet diameter D_0 , and time t , the law is expressed

$$\frac{D^2}{D_0^2} = 1 - k \frac{t}{D_0^2}, \quad (3.1)$$

where k represents evaporation rate. The evaporation rate for the water droplet conforms to reports in the literature [17,37]. Figure 2*a,b* illustrates the evaporation behaviour for different droplets for different field frequency and strength. Figure 2*a* shows that increase in frequency from 75 to 225 Hz (at 150 V) improves droplet evaporation rates. Likewise, enhancement in electric field strength (from 0 to 225 V (at 150 Hz)) also augments the evaporation rates (refer figure 2*b*).

The effect of increasing salt concentration has been reported by the present authors [17] and has not been discussed in this article. It has been shown that the saline droplet experiences internal thermo-solutal advection, which increases with the concentration of salt. Due to the internal

Figure 2. The evaporation behaviour of different saline droplets for (a) variation of field frequency at 150 V and (b) variation of field strength at 150 Hz. (Online version in colour.)

Figure 3. Time-lapse array for the droplets (0.2 M solution) for (a) frequency variation at 150 V and (b) field strength variation at 150 Hz.

circulation and the solutal Marangoni advection at the droplet surface, the vapour diffusion layer in contact with the surface of the droplet is also sheared. This shear replenishes the otherwise stagnant diffusion layer with ambient air, leading to improved evaporation [17]. The time lapses arrays in figure 3*a,b* qualitatively illustrate the role of the external field on the droplet. The higher field strengths are found to be more potent towards enhancing the evaporation compared with higher field frequencies. The potential mechanisms for improved evaporation could be due to modulated surface tension due to the Maxwell stresses generated at the droplet interface by the electric field, and the role of electric field induced internal thermo-hydrodynamics of the droplet.

(b) Role of surface tension and diffusion dominated evaporation kinetics in electric field environment

The surface tension of a fluid–gas interface may strongly regulate the evaporation rate of the liquid molecules to the ambient gas phase. The surface tension of conducting fluids such as salt solutions can be modulated by an electric field due to Maxwell stresses at the surface. The additional stress components at the surface arise due to Lorentz forces on the solvated ions,

Figure 4. (a) The effect of electric field stimulus on the surface tension of 0.2 M NaI solution. ST_f represents the variation of frequency at 150 V, whereas ST_v represents the effect of field strength variation at 150 Hz and (b) the comparison of predictions from the diffusion is driven evaporation kinetics [40] with the experimentally observed evaporation rates. (Online version in colour.)

which remain unbalanced in case of the ions adsorbed at the surface. Figure 4a illustrates the nature of variation of the surface tension of 0.2 M NaI solution for increasing field frequency (at fixed 150 V) and increasing field strength (at fixed 150 Hz). The pendant drop method has been used to determine the surface tension for the fluid. The results have been further validated by equating the weight of the detached fluid droplet to the surface force at the needle tip during detachment.

It can be observed (figure 4) that the surface tension increases marginally with the addition of salts (0.2 M). The surface tension of the water sample studied has been noted as 72.2 mN m⁻¹. With increasing field frequency or field strength, the surface tension is observed to increase, and then saturate. The increase in surface tension is within approximately 5–6% only. The increment in surface tension due to the addition of salts has also been reported in the literature [17]. Further, the electric field induces additional Maxwell stresses at the surface of the droplet due to the solvated ions adsorbed to the interface, which leads to a change in surface tension. Improved surface tension could lead to increment in evaporation rate in order to quickly minimize the surface energy, which is energetically favourable for the droplet. However, the observation of increased surface tension is not potent enough to induce enhanced evaporation along the observations in figure 1 and hence is not a strong mechanism to explain the present findings.

For pure fluids, diffusive evaporation to the surrounding gas phase is the dominant mechanism for pendant droplets. The classical diffusion driven model [41] can determine the evaporation rate for pendant droplets to the ambient medium. It models the evaporation as a quasi-steady process, which occurs at the liquid–gas interface. A thin film or shrouding layer of vapour is formed adjacent to the liquid’s outer layer. The vapour diffuses to the ambient gas due to a difference in vapour concentration between the vapour shroud and the ambient. Let Y_s and Y_∞ represent the mass concentration of the vapour phase within the shrouding diffusion layer and in the surrounding ambient phase. Then, Spalding’s heat and mass transfer numbers (B_T and B_M) are expressible as [40]

$$B_M = \frac{Y_s - Y_\infty}{1 - Y_s} \quad (3.2)$$

and

$$B_T = (1 + B_M)C_{pf}Sh/C_{pg}NuLe - 1. \quad (3.3)$$

C_{pf} , Sh , C_{pg} , Nu and Le represent the specific heat of the fluid, the Sherwood number, the specific heat of the surrounding gas, the Nusselt number and the Lewis number, respectively. The Spalding mass transfer number is defined as the non-dimensionalized difference of vapour concentration between the droplet surface and the far ambient. The Spalding heat transfer number is defined as the ratio of heat energy transported from the droplet to the ambient (due to evaporative flux and by virtue of temperature difference between the droplet and the far ambient) to the heat transfer across the vapour film shrouding the droplet. The Sherwood number represents the ratio of mass transfer rate due to convection to the mass transfer rate due to diffusion; the Nusselt number is the ratio of heat transfer due to convection to the conduction heat transfer and the Lewis number represents the ratio of thermal diffusivity to species diffusivity. Based on either equation (3.2) or (3.3), the mass flux due to evaporation can be deduced as [40]

$$\dot{m} = 2\pi\rho_g D_v R \ln(1 + B_M)Sh = \frac{2\pi\lambda R}{C_{pf}} \ln(1 + B_T)Nu. \quad (3.4)$$

D_v , λ and R represent the diffusion coefficient of the vapour with respect to the ambient phase, the thermal conductivity of the gas phase and the instantaneous radius of the pendent droplet, respectively. Figure 4b illustrates a comparison between the model predictions (equation (3.4)) and the experimentally observed evaporation rates. The model predicts the evaporation rate for water droplet accurate to $\pm 10\%$ (not illustrated in the figure, however, consistent with literature reports [17,41]). The experimental evaporation rate for water is determined as approximately $0.0011 \text{ mm}^2 \text{ s}^{-1}$, and of 0.2 M saline solution under the zero-field condition is approximately $0.0017 \text{ mm}^2 \text{ s}^{-1}$. It can be observed that the theory predicts the same value for the evaporation rate for all the cases in figure 4b. This is an expected phenomenon since the diffusion driven model predicts evaporation kinetics based on the gas side conditions, which remains unaltered for all the cases. Therefore, the diffusion driven evaporation kinetics model cannot predict the evaporation rates with field stimulus. Consequently, the mechanism of enhanced evaporation in all probability rests within the droplet and hence the internal transport phenomenon of the droplet during the evaporation needs to be probed.

(c) Influence of internal electrohydrodynamics on the evaporation kinetics

The internal hydrodynamics of the droplet during evaporation has been observed using PIV. PIV studies were carried out for 0.2 M NaI droplets, for permutations of field strengths of 75 and 225 V, and a frequency of 75 and 225 Hz. The PIV set-up is validated using the internal velocity of evaporating water droplets, and the interior of the droplet is observed to exhibit zero advection (with minor drift motion), and in agreement with reports [17,20,41]. Figure 5a–d illustrates the time-averaged velocity field for different electric field configurations. For the salt solution droplet at the zero-field condition, internal circulation exists (not illustrated, but reported earlier by present authors [17,20]). The spatio-temporal averaged circulation velocity for this case

Figure 5. Time-averaged means velocity contours within 0.2 M NaI solution droplets with different field constraints, as (a) 75 V and 75 Hz (b) 75 V and 225 Hz (c) 225 V and 75 Hz and (d) 225 V and 225 Hz. (e) Illustration of the comparison of theoretical velocities (of the spatio-temporally averaged velocities) with respect to the velocities predicted from the present mathematical model. (Online version in colour.)

is approximately 0.185 cm s^{-1} (refer figure 5e). This value is in agreement with previous reports [17,20,41]. Figure 5a,b illustrates the velocity field for frequency variation at 75 V and figure 5c,d illustrates the same for variation of frequency at 225 V.

The plane of circulation for salt solution under zero-field corresponds to the plane of the paper (the axis of circulation passing perpendicularly through the paper, and nearly through the centre of the droplet). Based on the PIV studies, it can be inferred that electrohydrodynamic flows within the droplet are active, and the external field tends to enhance the strength of the average internal flow (refer figure 5e). The internal advection within a droplet evaporating in a quiescent ambience can occur due to two possibilities, *viz.* the thermal and solutal gradients generated within the droplet. It is noteworthy that the solutal gradient is possible only in case of complex fluid (salt solutions, colloids, surfactant solutions, etc.), binary fluid droplets, etc. The evaporation process leads to cooling of the droplet's interior compared to its surface, which generates a thermal gradient across the droplet. Likewise, the pendant shape promotes unequal evaporation along the

droplet; leading to thermal gradients generated along the droplet surface as well. This gives rise to a minor (for water droplets) thermal advection within the droplet as well as thermal Marangoni circulation at the droplet interface (due to the temperature dependence of surface tension) [17,41].

Along similar lines, saline droplets exhibit solutal advection and solutal Marangoni circulation (due to the solute concentration dependence of surface tension). Solvated ions due to the addition of salts are preferentially adsorbed–desorbed at the droplet surface, which is evident from the modulated surface tension (refer figure 4a). The evaporation from the surface leads to a concentration gradient between the droplet's interior and interface. Additionally, the non-uniform evaporation from the pendant shape leads to solutal Marangoni circulation at the droplet interface [17,20]. The advection currents generated within the droplet and at the surface of the droplet develops shear at the liquid–gas interface. This shears the vapour diffusion layer surrounding the droplet and replenishes the otherwise stagnant layer with ambient air. This prevents the diffusion layer from saturating with vapour and improves the evaporation rate [17,20,41].

(d) Role of electro-thermal advection

The internal advection is caused by either thermal gradients or solutal gradients. Hence, it is important to determine the governing mechanism behind such internal advection. It is assumed that only the surface tension varies due to the thermal and solutal gradients established within the droplet during evaporation. Considering the electro-thermal effect as the dominant mechanism behind internal advection, the energy balance can be expressed as

$$\dot{m}h_{fg} = k_{th}A \frac{\Delta T_m}{R} + \rho C_p U_c A \Delta T_m + \rho C_p V_f A \Delta T_m. \quad (3.5)$$

In equation (3.5), the left-hand side represents the energy associated with the evaporative mass flux. This is balanced by the terms on the right-hand side, which represent the conduction heat transport, convective transport due to internal flow and the convective transport due to the electro-thermal flows, respectively. Here $\dot{m}$ and h_{fg} represent the evaporative mass flux and the enthalpy of vaporization, respectively. The variables k_{th} , A , ρ , ΔT_m , C_p , U_c and V_f represent the thermal conductivity of the fluid, the area of cross section of the droplet, the density of the fluid, the temperature difference between the interface and the bulk of the droplet, the specific heat capacity of the fluid, the mean internal circulation velocity due to thermal advection, and the mean internal circulation velocity due to electric field driven advection, respectively.

The advection velocity due to the thermal gradient is $U_c = \sigma_T \Delta T_m / \mu$, where σ_T and μ represent the gradient of surface tension with temperature change, and viscosity of the fluid [17,41]. The solvated ions are preferentially adsorbed–desorbed to the liquid–gas interface. The thermal advection leads to a system of moving charges within the liquid. In the presence of an electric field, the ions give rise to a collective body force, leading to an additional electro-thermal advection component, leading to enhanced circulation velocity. The electro-thermal advection velocity V_f is obtained by balancing the electrohydrodynamic forces in the system. The electromagnetic force on a charged system due to imposed fields is as

$$\mathbf{F} = q\mathbf{E} + (\sigma_e \mathbf{E} \times \mathbf{B}) + \sigma_e (\mathbf{v} \times \mathbf{B}) \times \mathbf{B}, \quad (3.6)$$

where $\mathbf{F}$, q , $\mathbf{E}$, $\mathbf{B}$, σ_e and $\mathbf{v}$ represent the electromagnetic force, the net charge within the fluid, the electric field strength, the magnetic field strength, the electrical conductivity of the fluid and the velocity of the charged particles, respectively. In the present case, no magnetic field is involved, and equation (3.6) reduces to

$$\mathbf{F} = q\mathbf{E}. \quad (3.7)$$

The force per unit volume (f) is hence expressed as

$$f = \rho_e E, \quad (3.8)$$

where ρ_e is the charge per unit volume, and $\rho_e = zeN \mathcal{A} \tilde{u}$, e and N represent the valence of the solvated ions, the magnitude of elementary charge and the number density of solvated ions. The electric body force leads to inertia within the system as

$$\rho_e E = \rho a, \quad (3.9)$$

where ' a ' represents the acceleration due to inertial circulation within the droplet. The acceleration can be scaled as $\sim V_f/t$ and the time elapsed can be scaled as $t \sim R/V_f$, where R is the instantaneous radius of the droplet. Substituting the scaled acceleration in equation (3.9) yields

$$V_f = \sqrt{\frac{\rho_e ER}{\rho}}. \quad (3.10)$$

V_f can be simplified in terms of the electric field potential ($V = ER$) and the volumetric charge density as

$$V_f = \sqrt{\frac{eNzV}{\rho}} = \frac{v}{R} \sqrt{\frac{eNzVR^2}{\rho v^2}} \quad (3.11)$$

and

$$V_f = \frac{v}{R} \sqrt{E_{HD}}. \quad (3.12)$$

In equation (3.12), v represents the kinematic viscosity and E_{HD} represents the Electrohydrodynamic number (refer equation (3.11)). Upon substitution of all the obtained expressions in equation (3.5), it is expressible as

$$\rho \dot{R} A h_{fg} = k_{th} A \frac{\Delta T_m}{R} + \rho C_p A \Delta T_m \frac{\sigma_T \Delta T_m}{\mu} + \rho C_{pf} A \Delta T_m \frac{v}{R} \sqrt{E_{HD}} \quad (3.13)$$

and

$$\rho \dot{R} R h_{fg} = k_{th} \Delta T_m \left[1 + \rho C_p \frac{R \sigma_T \Delta T_m}{k_{th} \mu} + \rho C_{pf} \frac{v}{k_{th}} \sqrt{E_{HD}} \right]. \quad (3.14)$$

The terms in equation (3.14) can be further simplified and rearranged as

$$\rho \dot{R} R h_{fg} = k_{th} \Delta T_m [1 + Ma_T + Pr \sqrt{E_{HD}}], \quad (3.15)$$

where Pr and Ma_T represent the Prandtl number and thermal Marangoni number, respectively. For stable circulation, the Ma must be above a critical value (approx. 80) [41] and hence equation (3.15) can be simplified as

$$\rho \dot{R} R h_{fg} = k_{th} \Delta T_m [Ma_T + Pr \sqrt{E_{HD}}]. \quad (3.16)$$

The second term within brackets (r.h.s. of equation (3.16)) represents the effect of the electric field on the thermal diffusion and is termed as the electro-Prandtl number. The effective electro-thermal Marangoni number governing the process can be expressed as $Ma_{T,e} = Ma_T + Pr \sqrt{E_{HD}}$, which represents the net modulation of the thermal Marangoni advection due to the presence of the electric field.

Figure 6a,b illustrates the effective contribution of thermal diffusion, thermal advection and electro-thermal advection (the terms on r.h.s. of equation (3.13)), with variation of frequency and field strength. The contribution due to thermal diffusion is insignificant (approx. 1–2%) when compared to the other mechanisms and hence not shown. At zero-field, the thermal advection dominates over the thermal diffusion within the droplet. In the presence of electric field, the contribution of the electro-thermal advection term is typically 40–45%, with reduction in the thermal advection component. The scaling analysis reveals that a large proportion of the internal electrohydrodynamics is caused by the electro-thermal effects of the conducting fluid. Now the genesis of the advection needs to be probed.

Thermal advection may be a typical Rayleigh advection due to the thermal gradient induced density gradients within the system. It could also be due to the thermal Marangoni advection,

Figure 6. The contribution of advection and electrohydrodynamic advection components in thermal transport with (a) frequency variation at 150 V and (b) field strength variation at 150 Hz, and in species transport with (c) field strength variation at 150 Hz and (d) frequency variation at 150 V. (Online version in colour.)

which is prominent in droplets and liquid films. The advection velocity due to buoyancy effects is expressible as [41]

$$u = \frac{g\beta\Delta T_R R^2}{\nu}, \quad (3.17)$$

where g and β represent the acceleration due to gravity, and the volumetric thermal expansion coefficient of the fluid, respectively. The temperature difference ΔT_R within the droplet, which leads to buoyant advection effect, is expressible as [17,41]

$$\Delta T_R = \sqrt{\frac{\nu \dot{R} h_{fg}}{g\beta R^2 C_p}}, \quad (3.18)$$

where, $\dot{R}$ is the rate of change of droplet radius. The Rayleigh number Ra is expressed as

$$Ra = \frac{R^2}{\alpha} \sqrt{\frac{\dot{R} h_{fg} g\beta}{C_p \nu}}. \quad (3.19)$$

To determine the dominant cause of the internal thermal advection, the approach of Rayleigh–Marangoni advection by Nield [42] and Davis [43] is adopted. The relative dominance of the Rayleigh and Marangoni convection in a conjugate system is found. Their strength is deduced with respect to their respective critical values (determined from stability analysis). The mathematical expression for the same is as

$$\frac{Ra}{Ra_c} + \frac{Ma_T}{Ma_{T,c}} = 1, \quad (3.20)$$

where Ra_c and $Ma_{T,c}$ represent the critical values of the Rayleigh and thermal Marangoni number, respectively. When plotted on the Ma–Ra plane, the relative dominance of the two modes can be determined using the criteria by Nield [42] and Davis [43]. The critical Ra is 1708 (as per

Figure 7. Stability plot for thermal Ma against Ra to determine the dominant mode of internal thermal circulation, with a variation of (a) field frequency (b) field strength. The two lines represent the stability regions by Nield [42] and Davis [43]. The entries in the legend without any field parameters represent zero-field cases. (Online version in colour.)

Chandrasekhar's stability criteria), while the critical Ma is approximately 80 [44]. Further, Davis put forward a regime of marginally stable internal advection, for critical Ma of approximately 53–54. Figure 7a illustrates the map of thermal Marangoni number (Ma_T) and Ra with a variation of field frequency (at 150 V) and figure 7b illustrates the same for variation in field strength (at 150 Hz).

For pure water, the point lies in the zone of unstable advection, which signifies that the droplet exhibits no internal hydrodynamics, and is experimentally observed. In the plot, the region below the Davis line represents region of no internal advection, the region between the Davis and Nield lines represents the zone of partially stable and intermittent internal circulation, and the region above the Nield [42] line represents the zone of stable internal circulation. The presence of salt in the system enhances the values of both Ma_T and Ra (figure 7a,b); however, the regime still lies in the region of unstable circulation. The increase of either field frequency or strength shifts the regimes upwards and rightwards, but the advection still remains within the region of unstable circulation. This contradicts the experimental observations, where the field induced internal advection persists over time, signifying stable or partially stable circulation. Thereby, the Ma_T versus Ra approach is not physically consistent and a different approach is essential to describe the mechanism.

Figure 8. Stability plot for electro-thermal Marangoni number ($Ma_{T,e}$) against Ra with a variation of (a) the field strength and (b) field frequency. The different lines cutting across the plot represent the iso- E_{HD} lines, which represent the various saline droplets and electric field constraints non-dimensionally and act as stability lines for the electro-thermal Ma system. The entries in the legend without any field parameters represent zero-field cases. (Online version in colour.)

The effective electro-thermal Marangoni number, $Ma_{T,e}$ versus Ra for different salt concentration and field constraints have been illustrated in figure 8a,b. At zero-field, the $Ma_{T,e}$ and Ma_T are equal, and hence lies in the region of unstable advection. With an increase in field parameters, all the points shift to the region of partially stable advection. This is in agreement with experimental observations, where significantly stable circulation behaviour is noted with electric field. This also cements the hypothesis that electro-thermal Marangoni advection is a key component behind the augmented internal advection, which leads to enhanced evaporation. In addition to the stability lines by Nield [42] and Davis [43], iso- E_{HD} lines have also been illustrated in the plot to show the behaviour of the electrohydrodynamics within the droplets for different

field constraints. It is observable that with increasing E_{HD} at a constant Ra , the iso- E_{HD} lines shift upwards, signifying increasing contribution to internal circulation by the electro-thermal Marangoni effect. It is also noteworthy that the separation between the iso- E_{HD} lines reduces with its increasing magnitude. This signifies that with increasing field strength or frequency, the strength of the internal advection gradually saturates, where stable circulation is consistent.

(e) Role of electro-solutal advection

The thermal Marangoni advection of evaporating droplets is common for fluids with temperature-dependent surface tension, e.g. water, alcohols and colloids [45]. The surface tension of many fluids is also strongly dependent on the concentration of solvated solutes or of other miscible fluid molecules. Hence, saline droplets will also exhibit the solutal Marangoni effect and the change in salt concentration due to evaporation further modulates this advection behaviour. Similar to the thermal Marangoni effect, the shape of the pendent induces non-uniform evaporation, leading to concentration gradient along the droplet surface, leading to solutal Marangoni advection on the surface. Additionally, solvated ions are preferentially adsorbed to the droplet interface compared to the bulk of the droplet (evident from the change in surface tension of the fluid). The dynamic change in solute concentration due to evaporation, within the droplet bulk as compared to the droplet's interface, gives rise to the solutal internal advection. The literature shows [17,45] that in saline droplets, the augmentation of evaporation is dominated by the solutal advection compared to the thermal advection. Consequently, the role of the electro-solutal advection also needs to be probed.

As discussed, solvated ions preferentially adsorb–desorb to the liquid–gas interface compared to the bulk liquid. Hence, the interfacial concentration and the bulk concentration of ions in a droplet are unequal, giving rise to solutal advection. Evaporative flux occurs from the droplet surface, which alters the surface concentration in a stronger manner than the bulk concentration. This aggravates the existing concentration difference, which further improves the solutal advection inside the droplet. The dynamic bulk concentration is determined by conservation of the total species. The product of V (instantaneous volume) and C (instantaneous bulk concentration) must be conserved ($VC = \text{const.}$), which is used to find the instantaneous bulk concentration from the instantaneous volume of the droplet. The dynamic interfacial concentration is determined from the temporally evolving surface tension, since the surface tension is dependent on solute concentration (refer figure 4) [17]. The temporal variation of the surface tension is quantified by fitting instantaneous droplet shape profiles to the Young–Laplace equation.

The transient evolution of surface tension is obtained from experimental data and fit to a mathematical expression. Likewise, the dependence of surface tension on the salt concentration and field constraints is obtained mathematically. The two equations are used to determine the dependence of interfacial concentration with time. These relationships were deduced for the initial 15–20 min to ensure that the Worthington number does not drop below approximately 0.6 [44], ensuring that the drop shape analysis is error-free. With time, the Worthington number ($Wo = V/V_0$, where V_0 is the initial volume) reduces and the shape fitting exercise is prone to errors. The dynamic bulk and interfacial concentrations have been illustrated in figure 9a and the difference between them in figure 9b. Under any field constraint, a difference exists between the bulk and the interfacial ionic concentrations, which establishes that solutal advection is present within the droplet. The strength of the advection increases with time due to the increasing difference between the concentrations.

The mass transport due to evaporation can be deemed responsible for the changes in species diffusion transport, solutal advection and electro-solutal advection (only in the presence of electric field) within the droplet. Mathematically, this can be scaled as

$$\dot{m} = DA \frac{\Delta C_m}{R} + U_{c,m} \Delta C_m A + V_{f,c} \Delta C_m A, \quad (3.21)$$

Figure 9. (a) The temporal variation of the dynamic and bulk concentration within the droplet (0.2 M NaI) for different field strengths (75 and 225 V) and frequencies (75 and 225 Hz). The term ‘int’ represents the dynamic interfacial concentration, and ‘bulk’ represents the dynamic bulk concentration. (b) The difference between the bulk and interfacial concentration for different field constraints. (Online version in colour.)

where $\dot{m}$, D , ΔC_m , $U_{c,m}$ and $V_{f,c}$ represent the rate of mass evaporation from the droplet, the diffusion coefficient of the salt with respect to water, the solute concentration gradient within the droplet, the velocity of internal solutal advection and the velocity of electro-solutal advection due to electric field, respectively. Further, $U_{c,m} = \sigma_c \Delta C_m / \mu$, where σ_c represents the rate of change of surface tension with the change of solute concentration [17,45]. Similar to the electro-thermal analysis, the $V_{f,c}$ is deduced as

$$V_{f,c} = \frac{D}{R} \sqrt{E_{HD}}. \quad (3.22)$$

Substituting the expressions for the advection velocities in equation (3.21) yields

$$\rho \dot{R} R = D \Delta C_m + \frac{\sigma_c (\Delta C_m)^2 R}{\mu} + v(\sqrt{E_{HD}}) \Delta C_m \quad (3.23)$$

and

$$\frac{\rho \dot{R} R}{D \Delta C_m} = (1 + Ma_s + Sc \sqrt{E_{HD}}), \quad (3.24)$$

where Ma_s is the solutal Marangoni number and Sc is the Schmidt number. For Marangoni advection to manifest, $Ma \gg 1$ [41], and hence the expression is simplified to

$$\frac{\rho \dot{R} R}{D \Delta C_m} = Ma_s + \sqrt{Sc_{sp}} \sqrt{E_{HD} Sc_i}. \quad (3.25)$$

The term $(Ma_s + \sqrt{Sc_{sp}} \sqrt{E_{HD} Sc_i})$ behaves as the effective electro-solutal Marangoni number ($Ma_{s,e}$). Equation (3.24) has been remodelled to express the Sc as composed of the Sc_{sp} , representing the species transport Schmidt number and the Sc_i , representing the ionic Schmidt number. The Sc_{sp} governs the strength of the momentum diffusion due to internal circulation to the mass diffusion of the ions. The Sc_i is the ratio of the ionic mobility (m_{ion}) to the diffusivity of the ions (D_{ion}), and is important during the electro-solutal advection.

Figure 6c,d illustrates the different modes of species transport described by equation (3.21). Comparing with figure 6a,b, it is observed that the degree of contribution of the electro-thermal advection (with respect to the zero-field thermal advection) is similar to the degree of contribution of the electro-solutal contribution (with respect to the zero-field solutal advection). Hence, it can be theorized that both the electro-thermal and electro-solutal advection contributes largely to the improved internal circulation, and increase in the evaporation rates. This is in stark comparison to the equivalent magnetohydrodynamics within droplets, where the magneto-solutal effect dominates over the magneto-thermal effect [20]. The stability of the electro-solutal Ma advection is

Figure 10. The phase map of the (a) thermal Ma against the solutal Ma in the absence of electric field (b) effective electro-thermal Ma against the effective electro-solutal Ma with electric field under variant field constraints. The thermo-solutal phase map is adapted from the literature [46]. The entries in the legend without any field parameters represent zero-field cases. (Online version in colour.)

determined from an analogous stability map of the $Ma_{T,e}$ versus $Ma_{s,e}$, (refer figure 10). Figure 10a illustrates the phase map for the Ma_T versus the Ma_s , for zero-field case.

All the points in figure 10a are to the right of the iso-Lewis number line, $Le = 0$ (not illustrated in the figure) [46]. This represents that the circulation within the droplet due to the solutal gradient is a stable and persistent. However, the relative roles played by the electro-thermal and electro-solutal Marangoni effects are not clear from this illustration. Figure 10b illustrates an effective thermal Marangoni number versus the effective solutal Marangoni number for field strength and frequency variations. With the electro-diffusive term incorporated in both numbers, the points shift further away from the origin, towards the right and upwards. This indicates increment of both the thermal and solutal Marangoni advection towards even more stable circulation with field constraints. This nature of the shift is also evident from the iso- E_{HD} lines, which have been included to map the stability of the field mediated advection.

The relative increment in the $Ma_{T,e}$ with respect to the Ma_T and the relative increment of the $Ma_{s,e}$ with respect to the Ma_s are of similar magnitudes. In the zero-field case, the solutal Ma is the dominant cause of advection compared to the thermal Ma [17]. However, as revealed by the present analysis, the augmentations in the effective electro-solutal Ma and the effective electro-thermal Ma are similar in magnitude. Hence, in the case of electrohydrodynamics mediated

improvement in evaporation, both the electro-thermal and electro-solutal advection play nearly equal important roles. Since both the electro-thermal and electro-solutal advection are noted to be of similarly dominant, the internal advection velocity can be deduced as a mean of both approaches (equations (3.12) and (3.22)). The mean advection velocities determined from the theoretical approach have been compared against representative experimental observations (refer figure 5e). Good agreements have been noted, which further strengthens the proposed mechanisms.

4. Conclusion

We study and discuss the effects of electric field on the evaporation characteristics of pendent droplets of conducting fluids. For the first time in the literature, improved evaporation of conducting fluid droplet due to the presence of an electric field has been experimentally studied and theoretically verified. The evaporation rate of saline pendent droplets was observed to increase with the application of an external electric field. The magnitude of improved evaporation rate holds a direct relationship with the increase of field strength and frequency. All the droplets under different field constraints conform to the classical D^2 law, which signifies that the vapour side is unaffected by the electric field. Analysis reveals that the improved evaporation rate cannot be explained by the changes in surface tension, as contradictory behaviour is observed. The classical diffusion driven evaporation model is also found to fail in predicting the evaporation rate constant under field constraints.

PIV study reveals that the thermo-solutal advection within the droplet is largely augmented by the electric field. This enhanced internal circulation velocity thus strongly shears the droplet interface, which further shears the vapour diffusion layer shrouding the droplet. This replenishes the vapour diffusion layer with ambient air, thereby improving the evaporation of the droplet. Theoretical scaling models based on electro-thermal and electro-solutal Marangoni advection has been proposed. The models reveal that the electro-thermal and electro-solutal Marangoni effects, and the internal electro-thermo-solutal advection lead to the improved circulation. It is shown that the electro-thermal and electro-solutal advection lies within the regions of stable advection on the respective stability maps. The spatio-temporally averaged internal advection velocities are deduced from the scaled model and good agreements with experiments have been noted. The work may find strong importance in design and development of microscale electrohydrodynamic systems.

Data accessibility. All data are presented and provided within this paper.

Authors' contributions. P.D. conceived the idea and V.J. performed the experiments and analysed the data. P.D. and V.J. wrote the article. P.D. also worked on the mathematical model. Both authors contributed to the manuscript and gave final approval for publication.

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