Hydrodynamics of electro-capillarity propelled non-Newtonian droplets through micro-confinements

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Abstract In this article, we theoretically explore the dynamics of droplet motion and its evolution during electro-capillarity propelled actuation within microfluidic systems. The study covers a wide gamut of fluids, wherein we investigate the dynamics of both pseudoplastic and dilatant fluid droplets. It is observed that change in the fluid rheology of the non-Newtonian fluids leads to significant morphing of the droplet dynamics during the actuation and propulsion event when compared to the Newtonian counterparts. We validate the theory using experimental reports on similar systems employing Newtonian droplets. The influence of governing parameters such as the actuation voltage and its transients, dielectric layer thickness on the electrodes and electrode spacing is probed. We also explore the influence of the interfacial properties of the system, such as channel wall friction, droplet wettability, and capillary friction, and establish that the fluid rheology, in conjunction with the interfacial features regulate the electro-actuation and propulsion of the droplets. We further provide theoretical estimates on the optimal design of the electro-actuation system in terms of a proposed electro-interfacial tension parameter. The findings may hold significance towards design and development of microfluidics with electro-actuation systems.

1 Introduction

Lab-on-a-chip devices and other assorted microfluidic systems have started to revolutionize rapid fluidic manipulation and analysis at miniaturized scales and minimized costs. Many such microfluidic devices depend majorly on bio-fluid manipulation, segregation and actuation. The wide scale acceptance of these devices stems from the fact that rapid and accurate testing and analysis with microliter quantities of the fluidic sample and reagents is possible; even in in-situ and outside lab conditions. Thereby, such devices find importance in localized and field trials and testing of important biomedical and engineering testing and on-spot results. Prime utilities of such devices are biomedical analysis of biomarkers such as blood [1–3], tumorous cells [4], urine [5,6], saliva [7,8], lymph [9], cerebrospinal fluid [10], etc. Microfluidics also involves testing and analysis in the food and beverage industry [11,12], chemical testing [13,14], fuel processing [15], etc. Thereby, the science and technology of microfluidics has received immense attention among researchers in the past two decades, and has garnered greater focus especially during the Covid-19 global pandemic [16,17]. Microfluidics devices are typically categorized into continuous flow devices, and discrete flow devices. The latter category is characterized by droplet microfluidics, where single droplets or a train of droplets of the primary or the secondary fluid is manipulated and analysed as per requirement. In such systems, manipulation and propulsion of the droplets through the microfluidic channels becomes a matter of prime importance and concern, as accuracy and control is most essential for proper analysis of the fluids and the dispersed phases, if any. At the same time, generation and fluidic control of such droplets is an area of active research. Droplet generation of a fluid within an immiscible continuous phase is often achieved using microfluidic junctions [18–20], where the channel dimensions [21]; junction angle [22], channel morphology [23], and flow rates are controlled to govern droplet characteristics. It has been extensively shown in the literature that the droplet shearing process is governed strongly by the interfacial tension and viscosity contrasts of the two fluids [24].
In addition to such methods, electric and magnetic field-based droplet generation, control, and actuation is of prime interest and utility to the microfluidics research community. Actuation and control via such fields can be programmed using accurate electronics, which adds accuracy to the fluidic manipulation system. At the microscale, the surface and interfacial forces dominate over viscous, inertial, and gravitational forces; and consequently morphing of the interfacial forces via electric and magnetic stimulus is a useful and convenient method for droplet manipulation. Under the influence of electric field, droplets in microfluidic channels can be propelled [25], sorted, distorted and split [26], coated and encapsulated [27], and transported from one region to another, either by electric body forces or by electro-capillarity [28]. The electric field option is more preferred over the magnetic one as all dielectric and conducting liquids can be controlled by electric field by proper engineering of the electrode-fluid assembly. For magnetic fluids however the magnetic field-based actuation remains the manipulation method of choice. Ferrofluid droplets can be propelled [29], manipulated for analysis [30], sorted and split [31], and transported preferentially [32] within microfluidic regimes by the use of alternating or direct magnetic fields.

Another aspect of significance in microfluidics and droplet manipulation technology is the fluid rheology. As the microfluidic regimes are low-inertial ones, the role of fluid viscoelasticity and non-Newtonian characteristics play a definitive role in determining the droplet dynamics during actuation and transport. Non-Newtonian fluid behaviour has been shown to modulate droplet formation and pinch-off within microfluidic junctions [33,34]. Non-Newtonian fluids also undergo extensional flows within microfluidic channels, leading to droplet formation [35]. Actuation and control of such droplets using electric fields has also been discussed in the literature [36,37]; however, the fluid has been restricted to Newtonian. In reality, microfluidic systems employ different types of bio-fluids, reagents, analytes, complex solutions, etc. which are inherently non-Newtonian. Since the rheology of non-Newtonian fluids depend on the local shear, the actuation induced motion is expected to make the dynamics of non-Newtonian fluid within microchannels differ considerably from the Newtonian counterparts, and the fluid rheology plays a decisive role. In this article, we explore the dynamics of such non-Newtonian droplets during electro-capillary actuation, and show how they behave compared to Newtonian droplets. We investigate the dynamics and motion of the droplet during impulsive start-up, and consequent propulsion when compared to Newtonian droplets. We explore the role of rheology through the flow index in the power law rheology model, and explore both pseudoplastic (shear-thinning) and dilatant (shear-thickening) fluids. We show via in-depth analysis the role played by individual components of the electro-capillary actuation system. The findings may provide significant guidelines towards design and development of microfluidic propulsion systems involving non-Newtonian fluids.

2 Physical domain and mathematical formulation

We first describe the physical domain and formulate the problem at hand, and proceed to the mathematical model. A schematic of the physical domain under analysis has been illustrated in Fig. 1. We consider a microfluidic channel constructed by two parallel plates, which are infinitely long along x and y directions, while separated from each other by a very narrow gap of 2a along the z direction. The plates are formed by sandwiched layers of conducting electrodes and dielectric layers. The bottom plate consists of a single, continuous ground electrode, whereas the top plate contains a linear array of control electrodes. Each control electrode is connected independently to an external circuit, through which a programmable electric field can be generated across any electrode, as desired. The microchannel thus formed is filled with a filler fluid (in this case air) and a microscale droplet is introduced and sandwiched in between. The inner walls of the insulating dielectric layer are coated to provide the requisite wetting regime for the droplet. The equilibrium shape of the droplet depends on the wettability of the top and bottom plates, and forms and equilibrium contact angle \( \theta \).

For simplicity, we assume in this study that both plates have the same wettability at any instant of time, and both dielectric plates have a thickness of \( d \).

To frame the mathematical model, we shall first establish the geometric parameters and physical assumptions. We consider the width of the droplet (along z direction) is \( w \), and is quite large compared to the height of the droplet (2a), and hence hydrodynamic analysis can be done only along the direction of actuation (x direction). We assume that the flow of the droplet is one-dimensional and quasi-steady, and that all properties are constant with space and time, unless explicitly mentioned. In our work, we consider the droplet fluid as a generic non-Newtonian fluid, which conforms to the Ostwald–de Waele power law model of viscosity \( (\mu = \mu_0 \left| \frac{dv}{dy} \right|^{n-1}) \), where \( \mu, \mu_0, n, \) and \( \left( \frac{dv}{dy} \right) \) represent the viscosity, the flow consistency index, the flow behaviour index, and the local shear rate, respectively. Further, as the flow paradigm is in the microfluidic domain, the approximation of small order of magnitude of the governing Weber number \( (We = \frac{\rho U^2 a}{\sigma_{ic}}) \), Capillary number \( (Ca = \frac{\mu_0 \left| \frac{dv}{dy} \right|^n U}{\sigma_{ic}}) \), and Bond number \( (Bo = \frac{\rho a^2}{\sigma_{ic}}) \) holds true. In these expressions, \( U, \rho, \) and \( \sigma_{ic} \) represent the axial velocity (averaged over the channel cross section), the density and surface tension of the droplet fluid, respectively. These essentially imply that the droplet meniscus can be approximated as spherical caps, as illustrated in Fig. 1. The low Bo (< 1) also ensures that the system is surface tension governed and the role of gravitational flattening of the droplet can be neglected.
Fig. 1 Schematic of the physical system under consideration. The overall system is shown in (a). Two long parallel dielectric layers form a micro-confinement, which is filled with a filler fluid (air in present case). A droplet is sandwiched in between the microfluidic confinement. Control and ground electrodes are positioned on the two dielectric layers as shown. An electric field is applied across the electrodes for electro-capillary actuation. In the zoomed view (b), the microchannel height, the spacing of the control electrodes, the thickness of the dielectric layer, and the front and rear contact angles of the droplet are shown. The details are described in Sect. 2. The associated coordinate system has been shown in (a).

Considering the above discussion, the governing equation for the droplet motion under dynamically evolving electro-capillarity actuation can be expressed by appealing to force conservation. At any instant, the inertia of the droplet is balanced by the conservation of the interfacial actuation forces, the viscous forces, and the frictional dissipation at the solid boundaries, and is expressed as

$$ m \frac{dv}{dy} = 2(w + 2a)\sigma lv \left[ \cos(\pi - \theta_f) - \cos(\pi - \theta_r) \right] $$

$$ - \left[ 6wL\mu_0 \left| \frac{dv'}{dy} \right|^n a \left( \frac{\dot{x}_f + \dot{x}_r}{2} \right) \right] - F_r \quad (1) $$

In Eq. 1, $v$, $m$, $x$, and $L$ represent the instantaneous average velocity of the droplet, the mass of the droplet, droplet location, and the instantaneous length of the droplet, respectively. Here, subscripts $f$ and $r$ represent the front and rear of the droplet (refer Fig. 1b). The first term of the RHS of Eq. 1 represents the capillary tension imbalance generated by the electro-capillary actuation, which leads to the axial motion of the droplet. The second term on RHS represents the visco-capillary resistance offered by the fluid due to wall shear and capillary motion, and the third term represents the friction at the wall-droplet interface due to the interaction of adhesion-cohesion forces. This additional frictional force originates from the velocity dependent capillary interactions at the contact line [37], and appears due to the capillarity interactions of the microscopic triple point contact line with the wall roughness. In a contact line of a droplet, the capillary forces manifest as the macroscale (leading to the formation of the apparent contact angle) and the formation of the microscale precursor layer, which extends away from the contact line and forms a microscopic film contact angle. The additional resistance due to the microscale precursor layer...
interacting with the wall roughness is modelled by the third term on RHS of Eq. 1.

As the viscosity of the surrounding fluid (air in this case) is orders of magnitude smaller than that of a liquid droplet, the additional drag force due to the frictional force can be modelled based on experimental observations [38] in the form

\[ F_r = 2\sigma_{lw}(w + 2a)BCa^q \]  

In Eq. 2, \( B \) and \( q \) are variables obtained from regression analysis on experimental data. The rationale behind modelling the capillary friction at the wall as function of the \( Ca \) can be provided based on the mechanism involved. During the motion of the contact line, the microscopic precursor layer traverses over the microscopic indentations and irregularities of the solid boundary; and this brings to the forefront the presence of a slip-modulated frictional force at the three phase contact point. At this region, the surface tension controls the local acceleration of the liquid–vapor interface, whereas the local viscosity governs the retardation of the control line. Due to the acceleration–retardation interplay, the interface executes locally oscillatory behavior while traversing over the microscopic irregularities of the solid walls, leading to capillarity induced additional frictional force. The effective traversal velocity is governed by the order of \( \sigma_{lw}/\mu_0 \left( \frac{d\nu}{d\theta} \right)^n \), or in other words by the order of \( \sim Ca \). Consequently, the frictional kinetics is modelled through Eq. 2. Although the original form of Eq. 2 has been reported for a liquid–liquid system [38], the equation is empirical in nature and dependent on the values of \( B \) and \( q \) only. Hence, it can be used for any combination of fluids as the empirical structure of the model can be adjusted simply by altering the values of \( B \) and \( q \).

Another important consequence of the capillarity at the contact line is the motion induced dynamic contact angle of the droplet. As the deformable droplet begins to move from its initial equilibrium position (Fig. 1), the shape of the droplet distorts and the front and rear contact angles, \( \theta_f \) and \( \theta_r \), appear. As the contact line traverses along the solid–liquid interface, the capillary tension and local viscous retardation leads to morphing of the droplet shape locally at the front and the rear, which leads to time dependent dynamic evolution of the \( \theta_f \) and \( \theta_r \). The temporal evolution of either of these angles can be expressed in terms of the initial equilibrium contact angle \( \theta_0 \) as

\[ \frac{d\theta}{dt} = F(\theta) + Ca \left\{ \ln \left( \frac{K}{\theta_d} \right) + \ln Ca \right\} \]  

Here, \( K \) is the constant associated with the wall slip model [37] and \( \theta_d \) is the length scale of average wall roughness. Equation 3 is valid for \( Ca \sim 1 \) and greater. The function \( F(\theta) \) is expressed further as

\[ F(\theta) = \int_0^\theta \frac{d\varphi}{f(\varphi)} \]  

Where,

\[ f(\phi) = \frac{2\sin\phi\left[q^2(\phi^2 - \sin^2\phi) + 2q[\phi(\pi - \phi) + \sin^2\phi] + (\pi - \phi)^2 - \sin^2\phi \right]}{q(\phi^2 - \sin^2\phi)(\pi - \phi) + \sin\phi\cos\phi + (\phi - \sin\phi\cos\phi)\left[(\pi - \phi)^2 - \sin^2\phi \right]} \]  

In Eq. 5, \( q \) represents the ratio of the viscosities of the two fluids (droplet and filler fluid) within the capillary system. It is noteworthy that Eq. 5 is predominantly aimed towards a Newtonian fluid. A power law fluid formulation is in essence a generalized non-Newtonian fluid. It is the most generic non-Newtonian formulation which only models the shear dependent flow features of the fluid. The model does not take into account any viscoelastic features, which means the model does not contain formulations of the coefficients of normal stresses at the fluid interface. It only expresses the shear stress in terms of the shear rate as a nonlinear function. Hence, the formulation is just a nonlinear representation of a Newtonian fluid and does not encompass any viscoelastic phenomena. Accordingly, the stresses near the contact line remains non-viscoelastic, as no additional viscoelastic stresses are introduced by the model. Thereby, the power law model (a generalized Newtonian model) can be incorporated with Eq. 5.

Next, we address the contact line velocity formulation in Eq. 1. Due to the different magnitude and evolution dynamics of contact angles at the front and rear, the contact line velocity at the front and rear are different. These may be evaluated based on the displacement of the front and rear contact lines, \( x_f \) and \( x_r \). In order to accurately map the displacement transients, we appeal to the conservation of volume of the droplet. During the motion of the droplet, the shape of the droplet distorts (elongation or contraction, depending on the relative magnitude of the contact line velocities) from its initial equilibrium shape; however, the volume of fluid is conserved. We first determine the volume \( V_\theta \) of the spherical caps at the two ends of the droplet (Fig. 1b) as function of the respective contact angle as [37]

\[ V_\theta = \frac{\pi a^3}{3\sec\theta} \left( \frac{2 + \sin\theta}{\left(1 + \cos\theta\right)^2} \right) \]
The distance $H_\theta$ of the centroid of the spherical caps from the respective triple contact line is evaluated as

$$H_\theta = \frac{(3 - 2 \sin \theta - \sin^2 \theta)}{4(2 + \sin \theta)} a \sec \theta \quad (7)$$

From volume conservation, we can write

$$V_{\theta_f} + V_{\theta_r} + 2a\omega L(\theta_f, \theta_r) = V_{\theta_i} + 2a\omega L_i = V_i \quad (8)$$

In Eq. 8, subscript $i$ represents the initial equilibrium conditions, and $L(\theta_f, \theta_r)$ is the instantaneous length of the temporally evolving droplet excluding the spherical caps at the end. We evaluate the spatial locations of the center of the front and the rear edges of the droplet based on the average velocity of the centroid of the whole droplet. The displacements are evaluated as \[37\]

$$x_f = \int v dt + \frac{a\omega L^2 - V_{\theta_f} H_{\theta_f} - V_{\theta_r}(L - H_{\theta_r})}{V_i} \quad (9)$$

$$x_r = \int v dt - \frac{a\omega L^2 - V_{\theta_r} H_{\theta_r} - V_{\theta_f}(L - H_{\theta_f})}{V_i} \quad (10)$$

Differentiation of Eqs. 9 and 10 yields the contact line movement of the centroid of the droplet $v'\frac{dy}{dt}$ term. As the droplet traverses in contact with the channel walls, the viscous stresses are confined in the near wall region. In this regime, the local velocity $v'$ can be expressed in terms of the velocity of the centroid of the droplet $v$ by appealing to the cubic order boundary layer approximation as

$$v' = \frac{3}{2} \eta - \frac{1}{2} \eta^3 \quad (11)$$

In Eq. 11, $\eta = y/\delta$, where $y$ is the local $y$-coordinate taken with respect to the top or bottom plates, and extending to the centre of the droplet, and $\delta$ is the approximate thickness of the near-wall viscous shear regime. The viscous shear is experienced at both the top and the bottom walls, and a factor 2 is introduced in the viscous friction formulation of Eq. 1 to account for the friction at both surfaces. Thereby, at the wall, the local shear rate can be evaluated as

$$\text{Lim}_{y \to 0} \frac{dv'}{dy} = \frac{3v}{2\delta} \quad (12)$$

Here, further approximation of $\delta = cx(Re)^{-1/2}$ is employed, where $c$ is a constant and $Re = \rho Lv/\mu$ is the local droplet Reynolds number.

We employ the power law model in the formulation, and obtain

$$\delta = cx\sqrt{\frac{\mu_0}{\rho Lv}} \left(\frac{dv'}{dy}\right)^{\frac{n}{2}} \quad (13)$$

Substitution of Eqs. 13 in 12 yields

$$\left.\frac{dv'}{dy}\right|_{y=0,x=L} = 3v^{3/2} \sqrt{\frac{\rho L}{2eL \mu_0}} \left(\frac{dv'}{dy}\right)^{\frac{n}{2}} \quad (14)$$

Further algebraic arranging yields

$$\frac{dv'}{dy} = \left(\frac{9\rho v^3}{4c^2L\mu_0}\right)^{\frac{1}{2n+2}} \quad (15)$$

We employ Eq. 15 in Eq. 1 to simplify the local shear rate term in the visco-capillary drag component; however, without any loss of physical generality as Eq. 15 is a function of the non-Newtonian parameters and the essence of the power law fluids is conserved. Although the set of Eqs. 11–15 are quasi-Newtonian in nature, it holds good for the power law model for non-Newtonian fluids. The power law model in essence is a generalized Newtonian model, where the viscous response is a function of the local shear, and no other additional viscoelastic parameters are present. Accordingly, it describes a generic Newtonian fluid and thereby conforms to the set of quasi-Newtonian equations.

The solution for Eq. 1 requires the initial condition to be prescribed in terms of the actuating electric field stimulus. For electro-capillary actuation, the electrical stimulus is applied to each control electrode following a pre-programmed sequence, such that the droplet is actuated in a slip-stick fashion along the length of the channel. Such actuation of droplets within microfluidic regimes is also commonly known as ‘digitized droplet motion’. Thereby, we require the expression for the initial velocity $v_0$ (which acts as the initial condition for Eq. 1) in terms of the electrical actuation parameters. In order to achieve motion from rest, the droplet has to overcome the static friction at the walls and the interfacial tension at the three phase contact line. Consequently, an impulsive voltage signal has to be provided to the electrodes at time $t = 0$. Unless an impulsive voltage is provided, the acceleration will not be instantaneous and the droplet cannot overcome the friction at the walls and begin motion. Once the electric field is established, the droplet and the electrode-insulation layers for a parallel plate capacitor system, with effective capacitance of $C$ and resistance of $R$. At approximately $t = 2.3RC$, the charging of the capacitive system attains $\sim 90\%$ of its rated capacity. It is during this time interval that the droplet experiences the impulsive starting velocity by overcoming the visco-capillary
resisting forces. Once this start-up velocity is attained, the droplet traverses to the next electrode by conservation of inertia, where the impulsive motion and subsequent actuation process repeats, until the droplet is transported to its destination.

During the impulsive electric field actuation, a differential electric field is set up between the front and the rear contact lines of the droplet, leading to electric field induced modulation of the interfacial tension. This differential interfacial tension leads to force imbalance, which manifests as the electro-capillary motion of the droplet. From classical mechanics viewpoint, the inertia of the droplet is conserved and can be equated to the net interfacial force imbalance across the droplet. The fraction of second time-lag that exists between the impulsive voltage increase and the sudden motion of the droplet has been neglected in the analysis for ease of mathematical analysis. Combining the discussion in the previous paragraph with this premise, we can mathematically express the initial start-up velocity of the droplet as

$$v_0 = \int_{t=0}^{2.3RC} \frac{2(w+2a)\{(\sigma_{sv} - \sigma_{sl})f - (\sigma_{sv} - \sigma_{sl})r\}}{m} dt$$

Equation 16

We further simplify the interfacial tensions in terms of the local equilibrium static contact angle \(\theta\) by appealing to the Young’s equation, given as

$$\sigma_{sv} - \sigma_{sl} = \sigma_{lv} \cos \theta$$

Equation 17

When the electric field is applied across the electrodes, the conducting fluid droplet and the electrode-insulator assembly assumes the role of a parallel plate capacitor (refer to Fig. 1). The capacitance \(C\) of the system is expressed as \(C = \frac{\varepsilon A_{sl}}{d}\); where \(A_{sl}, \varepsilon, \) and \(d\) represent the solid–liquid interfacial area, the dielectric permittivity and thickness of the insulation layer, respectively. The electrostatic energy is stored at the solid–liquid interface due to the capacitive nature of the system. From the point of view of conservation of energy, this leads to an effective reduction in the solid–liquid interfacial energy, as described by the classical Young-Laplace-Lippmann equation for electrowetting. The modulation of the interfacial energy can be described as

$$A_{sl}\sigma_{sl}(V) = A_{sl}\sigma_{sl}(0) - \frac{1}{2} CV^2$$

Equation 18

Here, \(V\) represents the applied electric field.

Equation 18 is employed in Eq. 17 to obtain the effective change in the static contact angle of the system under electric field actuation as

$$\cos \theta(V) = \cos \theta(0) + \frac{\varepsilon}{2d\sigma_{lv}} V^2$$

Equation 19

Equation 19 forms an expression akin to the Young-Laplace-Lippmann equation, which describes the change in the contact angle due to electric field, leading to morphed hydrophobicity at the wall, and thereby leading to directional electro-actuation in the direction of the electrode array. Equation 19 allows modelling the change in the contact angle in presence of electric field. It is the sudden change in contact angle that leads to the force imbalance, and leads to impulsive droplet motion. Both Eqs. 19 and 3 in conjunction decide the effective contact angle under electric field stimulus, and the effective behaviour of the front and rear contact angles under such stimulus, and the nature of motion due to frictional behaviour at the wall and the instantaneous capillary number of the droplet. We substitute Eqs. 17–19 in Eq. 16 to obtain the expression for the initial velocity of the droplet in terms of the time-dependent voltage actuation

$$mv_0 = \int_{0}^{2.3RC} 2(w+2a)\sigma_{lv} \frac{\varepsilon}{d} V(t)^2 dt$$

Equation 20

In Eq. 20, the electric field actuation signal is modelled in terms of a capacitive charging system as

$$V(t) = E(1 - \exp(-t/RC))$$

Equation 21

Here, \(E\) represents the peak value of the EMF applied across the electrode system. With the aid of the expressions in Eqs. 2–15, and the initial conditions imposed by Eqs. 20 and 21, the governing Eq. 1 is solved computationally. The transient Eq. 1 is solved numerically using an interactive time-marching integration scheme, and the displacement, velocity and acceleration of the droplet dynamics is obtained for different parameters.

3 Results and discussions

3.1 Dynamics of non-Newtonian droplet during electro-capillary actuation

We start the discussions with the dynamics of the droplets during electric field induced motion. For a typical case, we use the following realistic parameter values in the simulations: \(2a = 0.3\) mm, \(\rho = 1000\) kg/m\(^3\), \(\sigma_{sl} = 0.0072\) Nm\(^{-1}\), \(\mu_0 = 0.00089\) Pas, \(\theta_0 = 105^\circ\), \(L_i = 1.5\) mm, \(B = 2.5\), \(q = 0.3\), \(w = 1.5\) mm, \(l_d = 500\) mm, and \(K = 0.3\). We explore the role of rheology by studying the system for different values of \(n (= 0.25, 0.5, 0.75, 1, 1.5, 2, 2.5, \text{and} 3)\). To investigate the dynamics of the droplet during the actuation and propulsion events, we determine the displacement, velocity and acceleration signatures of the droplets with time. Figure 2 illustrates the displacement, velocity and acceleration of different shear-thinning fluid droplets, and their comparison with Newtonian fluid \((n = 1)\) droplet. For the Newtonian droplet, the average velocity over the time of observation is noted to be \(\sim 33.8\) mm/s, and is in close agreement with experimental observations in the literature [28], where the experiment with same parameters
as the current simulation showed average velocities of \( \sim 30 \text{mm/s} \). The analysis with the Newtonian droplets therefore is in agreement with experiments from the literature, and thereby validates the current mathematical formulation in the limiting case of Newtonian cases. There are no reports in the literature regarding similar experiments with non-Newtonian droplets, however, with the limiting case of validation of the Newtonian fluids, we believe that the current formulation predicts well over a large range of exponent \( n \). In addition, the power law model has been used here, which represents a generalized Newtonian fluid with shear dependent viscous response. Since the model is essentially a generalized Newtonian model, the experimental validation of the \( n = 0 \) case essentially justifies validity of the viscosity response model for all \( n \).

We first discuss the dynamics of the shear-thinning droplets with respect to the Newtonian ones. The rate of displacement or velocity of the Newtonian droplets (Fig. 2a, b) decreases with time, showing a sudden initial motion due to the imposed field. This initial impulse or jump is caused by the rapid charging of the electrodes within the initial time frame, which initiates the electro-capillary motion and the droplet rapidly slides by overcoming the local static friction and interfacial tension. Once the electro-capillary migration initiates, the frictional forces and the interfacial tension at the contact line of the droplet balance the inertia, which causes the velocity to decay out and attain a constant value with time. Such behaviour may not be very useful in case of sustained propulsion of the droplet at constant flow rate, as there is considerable difference between the impulsive start-up velocity (\( \sim 85 \text{mm/s} \)) and the steady-state velocity (\( \sim 35 \text{mm/s} \)). Further, when manipulating a train of droplets over a linear array of electrodes, such transients will lead to pulsatile flow behaviour, which will hamper analysis and detection within microfluidic devices. With change in the fluid rheology towards more shear-thinning behaviour (\( n = 0.75 \)), we note that the degree of reduction in velocity beyond the initial impulse reduces significantly. For \( n = 0.5 \), the velocity of the droplet attains a constant value, equal to the initial impulsive velocity, and consequently the displacement of the droplet becomes linear with time. With further change in the pseudoplastic nature to \( n = 0.25 \), we note that the velocity of the droplet consistently increases beyond the initial impulse, and the displacement shows a run-off nature.

The shift in the velocity history curve of the pseudoplastic fluids from the Newtonian case occurs due to the shear-thinning behaviour during the motion. When the initial electrical impulse is provided, the electro-capillarity at the contact line leads to sudden impulsive start of the droplet motion. This regime is short-lived, and only governed by the electric field and the interfacial tension, as the viscosity plays no role in overcoming the interfacial tension and static friction. This is evident from the fact that all the fluids show the exact same impulse peak velocity when the same electric field is applied. Once the impulsive motion begins, the fluid experiences viscous resistance during motion within the microfluidic passage. This viscous resistance is dependent on the shear at the wall, and for non-Newtonian pseudoplastic fluids, the viscous stress at the wall reduces as the index of the shear rate reduces. Consequently, for \( n = 0.25 \), the low viscous shear leads to a run-off like behaviour of the droplet, while the high shear leads to subsequent decay in viscosity for \( n = 1 \) fluids.

From a microfluidics application perspective, both behaviours are unwarranted, as decaying as well as run-off velocities may hamper any analysis and manipulation techniques in devices. From a utilitarian standpoint, the fluids close to \( n = 0.5 \) behaviour offer the most versatile and perfect droplet actuation and manipulation conditions due to the constant velocity history. Another important observation is noted from the acceleration plots (Fig. 2c). After the initial instantaneous acceleration, the droplet experiences constant acceleration for the \( n = 1 \) droplet. However, as the \( n \) reduces, the acceleration behaviour becomes largely nonlinear with time beyond the initial peak acceleration. From a mechanics point of view, this leads to nonzero values of the rate of change of acceleration, or jerk. Consequently, the pseudoplastic droplets, it is expected that the motion of the droplet will not be smooth, but exhibit fluctuating and local jerk-like motion during the actuation event.

We next focus on the hydrodynamics of the shear thickening fluid droplets (Fig. 3a–c). The initial impulsive rise in velocity due to the electro-capillary actuation leads to sudden increase in shear at the droplet-channel wall interface, which leads to sudden increase in the local viscosity of the shear thickening fluids. Due to this behaviour, the velocity rapidly decreases beyond the impulse peak, and the displacements history is largely deteriorated compared to the Newtonian counterpart. Consequently, we observe that although the actuation of such droplets using electric impulse is similar to that of Newtonian or shear-thinning droplets, subsequent propulsion is very slow paced due to sudden decay of velocity to a minimal constant value. However, it is noteworthy that the acceleration history (Fig. 3c) is unlike the shear-thinning droplets, and similar to the Newtonian droplets. The impulsive arrest of acceleration signal ensures that the droplet motion is steady and jerk-free.

Further, to characterize the electro-actuation behaviour of the different fluid droplets and to optimize the design parameters in realistic systems, we propose an electro-tension parameter, expressed as

\[
\beta = \frac{E^2 \varepsilon}{d} \tag{22}
\]

The parameter \( \beta \) has unit of N/m, and behaves akin to tension per unit length, and is thereby named accordingly. Essentially, \( \beta \) represents the capacitive calibre of the electro-actuation system, and its ability to propel the droplet from rest. In order to understand the role of the parameter, we quantify the displacement of the droplets before the impulse voltage reduces to 80% of
its peak value. The consideration of 80% is only a design parameter, and any value may provide similar insights. We plot this displacement ($X_{80\%}$) as a function of $\beta$ in Fig. 4 for the different non-Newtonian fluids. It is noted that the $X_{80\%}$ parameter initially increases with $\beta$ and then reduces, showing the existence of an optimal design regime. This optimal value of $\beta$ lies in the neighbourhood of $\sim 0.045$ N/m for the $n < 1$ fluids, while the $X_{80\%}$ is almost independent of $\beta$ for $n > 1$ fluids. Thereby, the rheology of the fluid has a direct link towards design and development of the system. Essentially, for best actuation and propulsion characteristics, the dielectric layer material (manifested through $\varepsilon$) and its thickness must be selected such that for the range of electric fields utilized, the value of $\beta$ remains close to the optima.

3.2 Influence of governing parameters

(I) Electric field strength and dielectric layer thickness
As seen from the preceding discussions, for a fixed selection of the dielectric material, the parameter $\beta$ is dependent on the electric field strength and the thickness of the dielectric layer. Consequently, these are two important design parameters whose influence shall be probed further. In Fig. 5, we illustrate the role played by the electric field strength ($E$) on the evolution of velocity of the different fluid droplets. As the control case, we first discuss Fig. 5b, wherein we focus on Newtonian droplets. Based on the value of $E$, we notice two different distinct regimes, wherein a transition in dynamics is observed between 80 and 100 V actuation voltage. In the regime up to $\sim 80$ V, the Newtonian droplets do not show the impulsive start-up behaviour. At 60 V, the start-up is gradual, and the droplet velocity increases beyond the peak, while at 80 V, the start-up is more rapid, however, the droplet velocity attains saturation at the peak. From 100 V onwards, a new regime is noted. The start-up event is more impulsive and the peak velocities are very high, albeit with the reduction of velocity beyond the peak.

However, it is noted that despite the decreasing velocity, the overall displacement is higher for higher electric field, caused by the initial high velocity regime. Eventually, the velocity curves for different $E$ merge as the system attains steady state. In the event of $n > 1$ droplets (Fig. 5c), the phenomena of reduction in velocity after the impulse peak is more pronounced. This is
Fig. 3 Transient dynamics of Newtonian and shear thickening fluid droplets \((n > 1)\) during electro-capillary actuation at 80 V. The plots show a displacement, b velocity, and c acceleration.

Fig. 4 Displacement of the droplet before undergoing 80% reduction of the peak voltage with respect to the electro-tension parameter \(\beta\), for a \(n < 1\) and b \(n > 1\) fluids.
more evident from the fact that the transition of regime from gradual start-up to impulsive start-up now occurs after 60 V. In addition, the steady-state velocities are attained earlier, and the magnitude is lower than the $n = 1$ case. In case of the $n < 1$ droplets (Fig. 5a), a reverse trend is noted. Here, the impulsive start-up regime is absent up to 120 V, and for all voltages, the start-up is gradual, followed by increase in the velocity. Consequently, in these cases, a steady-state velocity is not achieved and the droplets are expected to show run-off behaviour. For constant velocity propulsion (not accounting for the velocity magnitude), the electric field strength required for $n < 1$ fluids is higher ($> 120$ V) than the $n = 1$ fluids ($\sim 80$ V), which in turn is higher than that essential for $n > 1$ fluids ($\sim 60$ V).

Next, we discuss the influential role played by the thickness of the dielectric layer and illustrate its effects in Fig. 6. As a control system, we first consider the case of $n = 1$ droplets (Fig. 6b). It is observed that with increase in the dielectric layer thickness, the strength of the impulsive peak reduces significantly. This behaviour is essentially caused by the reduced capacitance of the system with increasing layer thickness, which reduces the electro-actuation calibre. Focusing on the $n > 1$ fluids (Fig. 6c), it is observed that the overall response of the fluids remain very similar to that of $n = 1$ fluids, for different dielectric layer thickness cases. The major difference occurs in the time taken by the velocity curves to converge, which is hastened in case of the $n > 1$ fluids. The dynamics of the $n < 1$ fluids (Fig. 6a) however is different, and the presence of the impulsive start-up behaviour is observed to be dependent on the layer thickness. The impulse start-up peak is noted to exist for very low values of the layer thickness. With increase in the thickness, the regime shifts to gradual start-up with increasing velocity condition. Consequently, the layer thickness is a strong operating parameter especially for $n < 1$ fluids. For applications that require constant velocity propulsion, there exists an optimum dielectric layer thickness ($\sim 1.6$ micron in the present case) and this thickness is a function of fluid rheology.  

(II) Solid–liquid frictional and wetting characteristics

The frictional behaviour at the solid–liquid interface plays a vital role in the efficiency of the electro-capillary actuation and propulsion. The frictional behaviour is morphed by tuning the parameters $B$ and $q$ in Eq. 2. In the previous discussions, the values of $B$ and $q$ have been taken as 2.5 and 0.3, respectively. We illustrate the effects of different values of $B$ in Fig. 7a, b, and that of $q$ in Fig. 7c, d, on the dynamics of the droplet motion. Figure 7a, b shows that with increasing $B$, the dynamics of the droplets shifts from increasing velocity (or run-off) condition towards a velocity reduction condition. Such behaviour is caused by the direct increase in the dynamic friction at the solid–liquid interface with increase in $B$. Figure 7c, d illustrates that with increasing value of $q$, the opposite trend occurs. With increase of $q$, the dynamics of the droplets shift from the regime of velocity reduction towards the regime of increasing velocity (or run-off). Increased value of $q$ increases the dependence of the dynamic friction on the $Ca$ of the system. The increase in $q$ leads to effective reduction of the frictional force due to higher microscale slip of the contact-line over the roughness at the solid–liquid interface. The role of the initial contact angle is illustrated in Fig. 8, where the effective higher interfacial slip in case of hydrophobic surfaces ($\theta_0 = 150$; Fig. 8a, c) leads to enhanced velocity signature of the droplets (post-actuation regime) compared to the low slip in case of hydrophilic ($\theta_0 = 50$) surfaces.

4 Conclusions

To conclude, we have shown theoretically the aspects of dynamics of droplet motion during electro-capillary actuation and propulsion within microfluidic confinements. We have also studied the evolution of motion during electro-capillarity for a generic range of non-Newtonian fluids. In this work, we focus on a wide gamut of fluids, and investigate the dynamics of both shear-thinning and thickening fluid droplets. This is done keeping in mind the varied applications of microfluidics, where biological fluid and chemical droplets, which are essentially non-Newtonian, are often employed.
Fig. 6 Influence of the thickness of the dielectric layer on the velocity of the droplets for $a\ n = 0.5$, $b\ n = 1$ and $c\ n = 1.5$ fluids. The thicknesses studied are 4.0, 3.2, 2.4, 1.6, and 0.8 microns.

Fig. 7 Role of the solid–liquid frictional characteristics on the velocity signature of the different droplets for parameters $a\ B = 1.25$, $b\ B = 5$, $c\ q = 0.15$, $d\ q = 0.45$. For the displacement characteristics of $n < 1$ fluids, refer fig. S1, and for velocity characteristics of $n > 1$ fluids, refer fig. S2 (supporting information).
Fig. 8 Role of initial contact angle on the droplet electro-capillarity velocity. Here, a and b represent the acceleration for $n < 1$ fluids, for a $\theta = 150^\circ$, and b $\theta = 50^\circ$. The dynamics for $n > 1$ fluids is shown for c $\theta = 150^\circ$, and d $\theta = 150^\circ$. The corresponding acceleration plots are illustrated in figure S3 (supporting information).

for analysis. The theoretical model shows that change in the fluid rheology of the droplets leads to significant morphing of the hydrodynamics during the actuation and propulsion when compared to the Newtonian fluid counterparts. The theoretical estimate of the Newtonian droplet displacement is validated using literature reports on experiments. We have explored the role played by governing parameters such as the actuation voltage and its transients, dielectric layer thickness on the electrodes and electrode spacing.

We show that optimal values of such parameters exist for sustained propulsion of the droplets, free from run-off and jerk conditions. We also show that the interfacial properties of the system, such as channel wall friction, droplet wettability, and capillary friction, play dominant roles in the hydrodynamics. Our theory is able to establish that the fluid rheology, in conjunction with the interfacial features regulate the electro-actuation of the droplets. We propose that the optimal design of the electro-actuation system can be achieved in terms of an electro-interfacial tension parameter, which can be tuned to obtain optimal propulsion behaviour. The overall study reveals that non-Newtonian effects in fluids may significantly alter droplet actuation and transport behaviour under electrical stimulation. This is true especially for shear-thinning fluids, where the dynamics of the droplets, such as displacement, impulsive actuation, and response to operating parameters such as dielectric layer thickness, etc. are grossly dependent on the exponent $n$, and different from non-Newtonian droplets. It was noted that as the fluids became more shear-thinning in nature, actuation and control may become difficult due to the increasing impulsive reaction of the droplet to electric stimulus. The finding from this study may hold importance towards design and development of electro-actuated micro-nano-fluidic devices.

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Author contribution statement

All authors have contributed equally to the work.

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Declarations

Conflict of interest We declare that we do not have any conflict of interest with respect to this research work.

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