Experimental and Numerical Investigation of Electrohydrodynamic Modes of Viscoelastic Polymeric Solutions

Amirreza Panahi, Ahmadreza Pishevar, Mohammadreza Tavakoli

Department of Mechanical Engineering, Isfahan University of Technology, Isfahan 84156-8311, Iran

Abstract

The main aim of this study is to explore the role of viscoelastic properties of polymeric solutions on mode transitions in electrospray process. By adjusting the applied electric potential between the nozzle and the collecting substrate, various electrohydrodynamic (EHD) modes were photographed by a high-speed camera. Then, the effect of operating parameters on the drops size in dripping mode and the jet profile in jet mode was investigated. By categorizing the EHD modes of each viscoelastic solution into dimensionless operating maps, it can be seen that by increasing the solution concentration or flowrate, the extents of dripping mode and beads on string structure dwindle, while the jet stabilizes in a wider range of electric capillary numbers. Furthermore, contrary to deionized (DI) water, when the applied voltage escalates, the stick jet mode is observed where the jet sticks to the outer surface of the nozzle, and the asymptotic thickness of the jet falls. In the second part of this research, several numerical simulations were conducted to simulate the behavior of an electrified viscoelastic jet. First, an electrified DI water jet was simulated, and the obtained jet profile was compared to the experimental data. Afterwards, the proposed algorithm was used to simulate viscoelastic electrified jets, where the effect of Weissened number (Wi) on the jet profile was examined. In agreement with the experimental results, by increasing the solution concentration, the asymptotic profile of the jet is reached at a smaller length from the nozzle, while the final thickness of the jet is slightly reduced.

I. Introduction

Electrohydrodynamic (EHD) phenomena have attracted increasing attention over the past decades. Electrospaying and electrospinning are the two main EHD applications. Electrospay is a method of producing a continuous stream of monodisperse droplets in ambient air. This is achieved by exerting the potential difference between a nozzle that is ejecting a continuous stream of droplets and a substrate positioned directly below the nozzle. Various means have been proposed for producing monodisperse droplets. Electrospay has advantages over other methods due to the flexibility it allows for droplet size and distribution. It can be used in thin film deposition, microencapsulation, inkjet printing, semiconductor devices, sample analysis, polymer particle production, and nanoparticle preparation.

Applied potential difference can produce various EHD modes in the electrospaying of a fluid with specific physical properties. These proposed EHD modes are categorized according to the shape and behavior of the fluid that is ejected from the nozzle. For instance, EHD modes using deionized (DI) water have been classified in several studies, and a brief description of each mode is presented here. Initially, in the absence of electric field, a dripping mode is observed for electrospay tests with DI water. If the voltage is increased, the droplet frequency surges, while the droplet size diminishes. Moreover, droplet

1 Author to whom correspondence should be addressed: apishe@cc.iut.ac.ir
frequency in this mode is relatively low, and the droplet size is greater than the diameter of the nozzle. Then, if the potential difference is increased, the micro-dripping mode, in which the droplet size is smaller than the diameter of the nozzle, is observed. This mode can be used for monodisperse aerosol generation. In the dripping and microdripping modes, the main drop is occasionally followed by a miniscule satellite drop. Next, the spindle mode is reached when a thin filament of fluid is detached from the nozzle. After a larger potential difference is brought to bear, a stable jet is generated by the application of tangential electric force on the jet surface. This tangential force accelerates the jet toward the substrate. The cone-jet mode can be easily identified through the presence of a conspicuous conical meniscus stretched upon its apex at the end of the nozzle. Depending on the physical properties of the fluid, the apex of the conical meniscus might be straight, oblique, convex, or concave. Finally, the multijet mode is identifiable by the presence of several thin jets on the annular rim of the cone. Further increases of the strength of the electric field leads to a higher number of jets, which in turn implies thinner jets.

Other work has explored the effects of different spraying fluids and changes in polarity. These modifications of the test circumstances lead to severe changes in the EHD modes produced and their stability domains. A complete review of electrospray and its fundamentals was published by Ganán-Calvo.

The EHD modes introduced have led many researchers to study and model the dynamic response of fluids to electric fields applied to them. Initially, most studies were focused on perfect dielectric fluid or perfect conductive fluid models. It was not until the pioneering work of Taylor and Taylor and Melcher that the leaky dielectric model became known as a way to model fluid deformation through the accumulation of electric charges on the two-phase flow interface. Ever since the introduction of this model, numerical and theoretical studies pertinent to this model have become ubiquitous in the literature. Saville summarized the main concepts and equations for this model in a 1997 review article. Moreover, in several previous studies, this model was used to simulate the deformation of a drop within a specified electric field strength and the well-known oblate and prolate deformations and conditions under which these deformations were reached were thoroughly discussed. Due to its ability to exert tangential force on the fluid interface, the leaky dielectric model has been utilized for cone-jet simulation and electrically controlled droplet generation.

The Weissenberg number (Wi), a dimensionless value that is regularly used in problems of viscoelastic fluids, is inherently large in electrospray problems. As reported in previous numerical studies, measures have to be taken to deal with the high Wi problem (HWNP). Different techniques have been proposed to stabilize numerical solutions in cases of high Wi values. The inconsistent streamline upwinding (SU) method, a special case of the Petrov–Galerkin formulation, can be used to stabilize numerical solutions for convection-dominant problems. Other stabilization methods, such as adding weak-form stabilization terms, have been proposed by Behr et al. and Coronado et al. for the Oldroyd-B model. Additionally, the SU method, together with the log-conformation method (LCM), can be used to stabilize the numerical solution of the Oldroyd-B model for viscoelastic fluid flow at high WIs.

The LCM reformulation, initially proposed by Fattal and Kupferman, solves the logarithm of the conformation tensor. In this way, the positive definiteness of the conformation
tensor is preserved, and the extensional components of the deformation field behave additively. The LCM reformulation has been used to solve several sophisticated problems of viscoelastic fluid flow, including lid-driven cavity stokes flow\textsuperscript{33, 34}, flow past a confined circular cylinder\textsuperscript{35-37}, flow past a sphere in a cylindrical tube\textsuperscript{38, 39}, abrupt contraction\textsuperscript{40, 41}, viscoelastic flow in a curvilinear microchannel\textsuperscript{42} and viscoelastic extrudate swell\textsuperscript{43, 44}.

The methods introduced in the literature to properly model the moving interface in two-phase flows include the volume-of-fraction method, the level-set method, and the phase-field method. These methods are widely used in both Newtonian and viscoelastic two-phase flows, however, the phase-field method is used in our simulations due to the improved numerical convergence.

The phase-field method has been the main subject of several studies, where it has been introduced as a versatile tool in multi-phase flow modeling. Two main types of this model include the Allen–Cahn and Cahn–Hilliard equations. The governing equations for both types have been delineated and investigated in the literature\textsuperscript{45, 46}; however, this article largely focuses on the Cahn–Hilliard equation, especially in conjunction with the Navier–Stokes equation\textsuperscript{47-49}. On top of that, the Cahn–Hilliard equation has been successfully coupled with different viscoelastic fluid models in previous works\textsuperscript{50-52} that investigate the applicability of the phase-field model to solving multi-phase non-Newtonian fluid flow.

Many studies have conducted numerical and experimental examinations of the role of viscoelasticity in flow behavior. As an illustration, the atomization mechanism of a charged viscoelastic liquid sheet was investigated in one work by solving viscoelastic constitutive equations in a perturbed state\textsuperscript{53}. The electrospinning of polyisobutylene-based solutions was investigated theoretically by Carroll and Joo\textsuperscript{54} using the method of linear instability analysis. Additionally, the atomization of polymer solutions was numerically modeled\textsuperscript{55} and experimentally studied\textsuperscript{56}, along with the role of the physical and rheological properties of fluid on the Sauter mean diameter of droplets. Li et al.\textsuperscript{57} used Oldroyd-B and leaky dielectric models to investigate viscoelastic jet axisymmetric and non-axisymmetric instabilities. In 2010, several electrospray tests were conducted for highly viscous solutions of sodium alginate, and empirical equations were suggested for the diameter of produced droplets\textsuperscript{58}. The dispensing mechanism of polyacrylamide (PAA) Boger fluid in the dripping mode was examined\textsuperscript{59}, where the surface tension and gravity forces were balanced in the neck region to obtain the diameter of the drop. Moreover, much research has been done on viscoelastic, electrified\textsuperscript{60, 61} or free-falling\textsuperscript{62} liquid jets. The beads on a string structure, which is a rather peculiar and interesting behavior of viscoelastic fluid, was numerically modeled by Li and Fontelos\textsuperscript{63} and Turkoz et al.\textsuperscript{64}. Recently, viscoelastic fluid electrospray was investigated by Yu et al.\textsuperscript{3} to optimize the fluid used in inkjet printing. They used polyethylene oxide with different molecular weights at various solution concentrations to independently control the elasticity and viscosity of the solution. Because of the different physical and rheological properties that were explored, new EHD modes were observed, and operating maps for every solution were provided.

The main aim of this study is to explore the role of viscoelasticity on mode transitions of EHD. For this reason, the electrospray of a dilute PAA aqueous solution at different concentrations is examined experimentally, and transitions in modes of EHD are classified with dimensionless operating maps. Furthermore, new EHD modes
for viscoelastic dilute solutions are introduced and observed with high-speed photography. The images acquired in this way are used to examine the effects of various test conditions on droplet diameter and the viscoelastic jet profile. The process is also investigated numerically by solving the constitutive equations for viscoelastic electrified jets. In our case, due to the small characteristic length of the problem, the Wi is rather large. As a result, an HWNP, a complication encountered in the solution of viscoelastic constitutive equations, is addressed and the implementation of the LCM reformulation is clearly described to rectify the problem. To validate the overall results of the current work, the jet profiles obtained through numerical simulations are compared with their corresponding experimental data, and a good agreement is seen between the two data sets.

The rest of this paper is organized as follows: the problem is formulated in Section II. In Sections III and IV, the experimental setup and the results of the experiments and simulations are discussed. Finally, the paper is concluded in Section V.

II. Governing Equations

Here, we assume that the system under consideration consists of two immiscible, incompressible fluids. One, the polymeric solution, behaves as a viscoelastic fluid, while the other one, the air phase, behaves as a Newtonian fluid. In the following subsections, the equations used to model the current system are given step by step descriptions. Subsequently, the aforementioned equations are coupled in fluid flow equations. The governing equations in this section are delineated in axisymmetric coordinates, with the z axis considered the symmetry axis.

A. Viscoelastic Constitutive Equations

The flow behavior of viscoelastic fluid can be explained by generalized Newtonian, linear viscoelastic or non-linear viscoelastic models. The Oldroyd-B model, a simple non-linear viscoelastic model, is chosen as our governing equation to model the flow behavior of viscoelastic fluid. Assuming a constant polymeric viscosity (no shear thinning), which makes it a perfect fit for modeling Boger fluids, this model is written as follows:

\[ \tau + \lambda_1 \tau^v = \eta_0 (\dot{\gamma} + \lambda_2 \dot{\gamma}^v) \]  

where \( \lambda_1, \lambda_2, \eta_0 \) and \( \dot{\gamma} \) denote relaxation time, retardation time, total viscosity, and shear rate, respectively. The total viscosity is defined as the sum of polymeric and solvent contributions to viscosity, and the shear rate is defined by the velocity gradient:

\[ \eta_0 = \eta_p + \eta_s \]  

\[ \dot{\gamma} = 0.5 (\nabla u + (\nabla u)^T) \]  

The \( \tau^v \) variable is the upper convected Maxwell derivative of stress and is defined as follows:

\[ \tau^v = \frac{\delta\tau}{\delta t} + (u, \nabla)\tau - (\nabla u)^T \cdot \tau - \tau \cdot (\nabla u) \]  

It should be noted that the definition of the upper convected Maxwell derivative for shear rate is identical to Eq. (4). By splitting the total stress and relating relaxation and retardation times, we produce the following:

\[ \tau = \tau_p + \tau_s \]  

\[ \lambda_2 = \left( \frac{\eta_s}{\eta_p + \eta_s} \right) \lambda_1 \]  

If Eqs. (5) and (6) are substituted into Eq. (1), the following relationships are obtained:

\[ \tau_s = \eta_s \dot{\gamma} \]  

\[ \tau_p + \lambda_1 \tau_p^v = \eta_p \dot{\gamma} \]  

Eqs. (7) and (8) show that Newtonian and polymeric stress equations can be solved and added independently to the Navier–Stokes equation. From this point forward, the index of \( \lambda_1 \)
The general form of conformation and log-conformation tensors is similarly defined. Furthermore, a symmetrical positive definite tensor, the conformation tensor can be decomposed as follows:

$$\sigma = R\lambda RT^T$$

(16)

where $R$ is an orthogonal tensor made by the eigenvectors of $\sigma$, and $\lambda$ is a diagonal tensor made by eigenvalues of $\sigma$. Next, $N$, $\Omega$, and $B$ are decomposed by the tensor $R$ and its transpose:

$$N = R\begin{pmatrix} 0 & n_{12} & n_{13} \\ -n_{12} & 0 & n_{23} \\ -n_{13} & -n_{23} & 0 \end{pmatrix} R^T$$

(17)

$$\Omega = R\begin{pmatrix} 0 & \omega_{12} & \omega_{13} \\ -\omega_{12} & 0 & \omega_{23} \\ -\omega_{13} & -\omega_{23} & 0 \end{pmatrix} R^T$$

(18)

$$B = R\begin{pmatrix} b_{11} & 0 & 0 \\ 0 & b_{22} & 0 \\ 0 & 0 & b_{33} \end{pmatrix} R^T$$

(19)

Tensor $M$ is defined as follows:

$$R^T (\nabla u^T) R = M = \begin{pmatrix} m_{11} & m_{12} & m_{13} \\ m_{21} & m_{22} & m_{23} \\ m_{31} & m_{32} & m_{33} \end{pmatrix}$$

(20)

Eqs. (17)–(20) are used to decompose the velocity gradient. For more details, readers are referred to Appendix 1. If we substitute Eq. (13) into Eq. (10) and simplify the result, following Fattal and Kupferman, a log-conformation constitutive equation is obtained:

$$\frac{\partial \psi}{\partial t} + (u.\nabla)\psi - (\Omega \psi - \psi \Omega) - 2B = \frac{1}{\lambda} (e^{-\psi} - I)$$

(21)

where $\psi$ is the log-conformation tensor. The conformation and log-conformation tensors are related by the eigenvectors of the conformation tensor:

$$\psi = R \log(\lambda^{\sigma}) R^T$$

(22)

$$\sigma = Rexp(\lambda^{\psi}) R^T$$

(23)

where $\lambda^{\sigma}$ and $\lambda^{\psi}$ are tensors made by eigenvalues of conformation and log-conformation tensors, respectively. The relations used to acquire the
eigenvalues and eigenvectors of the log-conformation tensor are shown in Appendix 1.

**B. Two-phase Flow Equations**

To capture the interface, the phase-field method is adopted, and the surface tension force is applied to every node near the interface as the body force. First, the phase-field parameter is defined as follows:

\[ \emptyset = \frac{m_1 - m_2}{m_1 + m_2} \]  \hspace{1cm} (24)

where \( m_1 \) and \( m_2 \) are the masses of each phase. Alternatively, Eq. (24) can be interpreted as indicating the differences in concentration between two phases, where the concentration for each phase has a value between 0 and 1, and consequently, the phase-field parameter can vary between -1 and 1.

The chemical energy density and the Helmholtz free energy for unit volume in a homogenous mixture are defined as follows:

\[ F(\emptyset) = \frac{1}{4} (\emptyset^2 - 1)^2 \]  \hspace{1cm} (25)

Additionally, the chemical potential is defined as follows:

\[ f(\emptyset) = F'(\emptyset) - \varepsilon^2 \Delta \emptyset \]  \hspace{1cm} (26)

where \( \varepsilon \) is the interface thickness. The Cahn–Hilliard equation is a conserved form for the phase-field model, and it is written as follows:

\[ \frac{\partial \emptyset}{\partial t} + u.\nabla \emptyset = \nabla \cdot (M(\emptyset) \nabla f(\emptyset)) \]  \hspace{1cm} (27)

where \( \lambda \) is the mixing energy density. \( M(\emptyset) \), denoting mobility, has been given various definitions in the literature. In the phase-field method, the surface tension force is obtained by the following:

\[ F_{ST} = \frac{\lambda \varepsilon}{2} f(\emptyset) \nabla \emptyset \]  \hspace{1cm} (28)

Then, the volume faction of each phase is defined by phase-field parameter, as follows:

\[ Vf_1 = \frac{1-\emptyset}{2} \quad \& \quad Vf_2 = \frac{1+\emptyset}{2} \]  \hspace{1cm} (29)

**C. Electrostatic Equations**

As previously noted, the leaky dielectric model is used to simulate the electric field. Neither perfectly dielectric nor perfectly conductive, poorly conductive fluids are typically modeled with the leaky dielectric model, where the effect of the accumulation of charge on a finitely thin interface is taken into account. As shown by Saville, magnetic effects in EHD problems can be omitted because the characteristic time for magnetic phenomena is several orders of magnitude smaller than the characteristic time for electric phenomena. Therefore, it is only necessary to deal with general electrostatic equations:

\[ \nabla \times E = 0 \]  \hspace{1cm} (30)

\[ E = -\nabla V \]  \hspace{1cm} (31)

The space charge density is related to the electric field (or potential difference) through the following relations:

\[ \nabla \cdot (\varepsilon_0 \varepsilon E) = -\varepsilon_0 \nabla \cdot (\varepsilon \nabla V) = \rho_e \]  \hspace{1cm} (32)

Furthermore, the following charge-conservation law should be satisfied at every node:

\[ \frac{\partial \rho_e}{\partial t} + \nabla J = 0 \]  \hspace{1cm} (33)

where \( J \) is the electric charge flux, defined as:

\[ J = \sigma E + \rho_e u \]  \hspace{1cm} (34)

The first term in Eq. (34) represents ohmic charge conduction, and the second term represents charge convection by velocity field. Before any further simplification, the charge relaxation time, viscous relaxation time, and capillary time scale are defined as follows:

\[ \tau_c = \frac{\varepsilon_0 \varepsilon}{\sigma} \]  \hspace{1.5cm} (35)

\[ \tau_{\mu} = \frac{\rho l^2}{\mu} \]  \hspace{1.5cm} (36)

\[ \tau_{cap} = \sqrt{\frac{\rho l^3}{\gamma}} \]  \hspace{1.5cm} (37)

In a leaky dielectric system, electric charges are accumulated near the interface, and it is assumed that the thickness of the electric double layer is...
very small relative to the scale of the problem length. For this case, the diffusion of electric charges is neglected, so the space charge density is assumed to be zero and the effects of surface charges are considered to be a boundary condition. The electric relaxation time is small relative to the viscous time scale; consequently, the charge-conservation equation is simplified by omitting the convection term and considering the quasi-static form of the following equation:
\[ \nabla \cdot (\sigma \varepsilon F) = 0 \]  \hspace{1cm} (38)
In the dripping mode, the capillary time scale has the same order of magnitude as the electric relaxation time, so Eq. (38) is only plausible for the simulation of the jet mode. In brief, the simulations presented in this report only include viscoelastic electrified jet modeling. Finally, the Maxwell stress tensor is defined as follows:
\[ \tau_e = \epsilon_0 \epsilon (\vec{E} \cdot \vec{E} - \frac{1}{2} \vec{E} \cdot \vec{E}) \]  \hspace{1cm} (39)
The divergence of the Maxwell stress tensor yields the force exerted on the ejecting fluid by the electric field:
\[ F_e = \nabla \cdot \tau_e = -\frac{1}{2} \vec{E} \cdot \vec{E} \nabla \epsilon_0 \epsilon + \rho_e \vec{E} \]  \hspace{1cm} (40)

D. Fluid Flow Equations

The fluid flow equations include incompressible continuity and the Navier–Stokes equation:
\[ \nabla \cdot u = 0 \]  \hspace{1cm} (41)
\[ \rho \frac{\partial u}{\partial t} + \rho (u \cdot \nabla) u = \nabla \cdot \beta + \rho g + F_{ST} + F_e + F_p \]  \hspace{1cm} (42)
Where \( F_p \), \( F_{ST} \) and \( F_e \) are previously defined by Eqs. (11), (28), and (40), respectively. Additionally, \( \beta \) relates the pressure and Newtonian viscous stress, as follows:
\[ \beta = -pI + \eta_s \dot{y} \]  \hspace{1cm} (43)
Inevitably, every physical property described in the above equations should be smeared across the interface through the volume fraction of fluids.

For instance, the following relation is written for the relaxation time:
\[ \lambda_r = \lambda_1 V_{f1} + \lambda_2 V_{f2} \]  \hspace{1cm} (44)
where \( V_f \) is the volume fraction of corresponding fluids. To avoid numerical complications, a very small value is considered for the air relaxation time. (Refer to Table II)

E. Dimensionless Numbers

In this section, the essential dimensionless numbers controlling the electrospray process are introduced. The Webber number, which describes the ratio of inertia forces to surface tension forces, is defined as follows:
\[ We = \frac{\rho u^2 l}{\gamma} \]  \hspace{1cm} (45)
The electric capillary number is the ratio of the electric field force to the surface tension forces, and is given by:
\[ Ca_e = \frac{\epsilon_0 \epsilon_r E^2 R_0}{\gamma} \]  \hspace{1cm} (46)
where \( R_0 \) is half of the characteristic length, which in our case is the outer radius of the nozzle. In this analytical study, the strength of the electric field at the tip of a positively charged cylinder with a semi-finite ground terminal positioned below the cylinder is calculated using a relationship originally proposed by Jones and Thong:
\[ E = \frac{\sqrt{2V_0}}{r_c \ln(4z_0/r_c)} \]  \hspace{1cm} (47)
where \( z_0 \) is the distance between cylinder and ground terminal, \( V_0 \) is applied voltage, and \( r_c \) is the outer radius of the cylinder (or in our case, the nozzle). Additionally, \( \epsilon_r \) in Eq. (46) is termed the characteristic relative permittivity and is derived using the Lorentz model for the interaction of electromagnetic waves in dielectric materials:
\[ \epsilon_r = 1 + \frac{\sigma}{\epsilon_0 \omega} \]  \hspace{1cm} (48)
where \( \omega \) is characteristic frequency:
\[ \omega = \frac{c}{L} \]  \hspace{1cm} (49)
In Eq. (49), \( c \) is the speed of light in air, and \( L \) is the distance between the center of the capillary tip and the inner edge of an annular disk taken as the substrate. Last but not least, the dimensionless droplet frequency is defined as follows:\(^\dagger\):

\[
 n^* = \left( \frac{nR_0^3}{Q} \right)^{\frac{1}{3}} \tag{50}
\]

where \( n \) is the droplet frequency, and \( Q \) is volumetric flowrate. The dimensionless numbers introduced in this subsection are used in Section IV to categorize and classify the results.

### III. Experimental Setup

A schematic representation of our experimental setup is given in Fig. 1. Every electrospray test was conducted under conditions of atmospheric pressure and 25°C temperature. A stainless steel nozzle with an inner diameter of 0.31 mm and an outer diameter of 0.63 mm was connected to a syringe pump to inject fluid between the nozzle and a round aluminum disk taken as the substrate. The nozzle was positioned vertically to prevent wetting effects, and the distance between the nozzle and the substrate was 30 mm. A high voltage source was used to exert the potential difference between the nozzle and the substrate. As the voltage was varied, different EHD modes were observed. The viscoelastic solutions used in the electrospray tests were aqueous PAA solutions in three different concentrations, including 50, 100, and 150 ppm, where the molecular weight of the PAA powder was \( 2 \times 10^7 \) gr/mole. The fluid was pumped using an SP-100s syringe pump and high-speed photography equipment included a PCO high-speed camera and a NIKON AF Micro-NIKKOR 200 mm f/4D IF-ED lens.

Figure 1. Schematic representation of the experimental setup used in this study.

The measured physical properties of the PAA solutions can be found in Table I. An Anton–Paar modular compact rheometer (MCR) was used for rheometric tests, where the relaxation time of the solutions were determined with a small-amplitude oscillatory shear test (the \( G' \) test), and the shear thinning behavior of solutions was examined. Negligible shear thinning behavior was observed (the polymeric viscosity remained fairly constant for different shear rates), indicating that the Oldroyd-B model can predict viscoelastic behavior for PAA
solutions with acceptable accuracy. It should be added that glycerin was not used in the viscoelastic solution solvents, so the solvent viscosity remains low, and the fluid flows through the nozzle conveniently. Furthermore, the surface tension of the viscoelastic solutions was measured using a Dataphysics DCAT 11, following the ASTM D 1331-14 standard and using the Wilhelmy plate method, and the solution electric conductivity was measured with a JENWAY 3540 conductometer. The physical properties of the DI water and air, which are used in numerical simulations and to calculate dimensionless numbers, are listed in Table II. It is worth noting that the polymeric viscosity and relaxation time are zero in air; nevertheless, in numerical simulations, small quantities were allocated to these properties to avoid numerical difficulties.

| Concentration (ppm) | Surface Tension (mN/m) | Electrical Conductivity (μS/m) | Density (kg/m³) | Solvent Viscosity (Pa.s) | Polymeric Viscosity (Pa.s) | Relaxation Time (s) |
|---------------------|------------------------|-------------------------------|-----------------|--------------------------|--------------------------|-------------------|
| 50                  | 70.739                 | 20.3                          | 1000.05         | 0.001                    | 0.0047                   | 0.518             |
| 100                 | 71.751                 | 36.6                          | 1000.1          | 0.001                    | 0.035                    | 1.094             |
| 150                 | 72.169                 | 59.9                          | 1000.15         | 0.001                    | 0.068                    | 1.709             |

| Fluid               | Surface Tension (mN/m) | Electrical Conductivity (μS/m) | Density (kg/m³) | Solvent Viscosity (Pa.s) | Polymeric Viscosity (Pa.s) | Relaxation Time (s) |
|---------------------|------------------------|-------------------------------|-----------------|--------------------------|--------------------------|-------------------|
| DI Water            | 72                     | 5.5                           | 1000            | 0.001                    | -                        | -                 |
| Air                 | -                      | 8e-9                          | 1.225           | 1.18e-5                  | 1e-14                    | 1e-14             |

IV. Results
A. Experimental Results
At the beginning of every experiment, the electrospay of DI water was performed to verify the repeatability of the experimental results. For this purpose, the experimental data presented by Jaworek and Krupa10 was used to analyze and validate the results. This procedure reliably ensures that the experimental conditions will lead to the same results, and no new adjustments or calibrations are needed. Additionally, every test was repeated three times, and the obtained results were compared such that the authenticity of the results can be examined. High-speed photography was begun after each mode reached steady state. The main sources of error in the experimental tests are voltage measurement (0.1 kV), flowrate measurement (1 mL/h), and image-processing algorithms (one pixel size). In the analysis of the results, these sources of error are taken into account.
For the sake of comparison with the viscoelastic results, various EHD modes of DI water are shown in Fig. 2. As can be seen in the figure, these EHD modes include the dripping ($\text{Ca}_E = 0$–0.93), microdripping ($\text{Ca}_E = 1.37$), spindle ($\text{Ca}_E = 1.91$), cone-jet ($\text{Ca}_E = 2.57$–6.31), and precession ($\text{Ca}_E = 6.69$–7.08) modes.

From this point forward, we concentrate on the EHD modes of viscoelastic diluted solutions. The first distinguishable characteristic in the viscoelastic solutions can be seen in the dripping mode, where a droplet is followed by a thin filament of fluid.

![Figure 2. Observed DI water EHD modes for a 108 mL/hr flowrate, categorized according to electric capillary number.](image)

It is clear from Fig. 3 that the detachment of the drop from the tip of the nozzle varies significantly for viscoelastic solutions. Generally speaking, Newtonian droplets detach instantly from the nozzle, or, in some cases, they are followed by a small satellite droplet. Viscoelastic droplets, on the other hand, are known for depicting strong elastic behavior upon detachment. The thin filament formed in viscoelastic fluids is stretched, and its thickness plummets as the drop moves further away from the nozzle. Consequently, the filament becomes extremely thin, until it breaks.
In Fig. 4, every viscoelastic EHD mode is illustrated in an increasing sequence of electric capillary number for a 108 mL/h flowrate and a 100 ppm PAA aqueous solution. The first mode observed in viscoelastic solutions is the dripping mode, where the droplet size falls, while the frequency increases as the voltage increases \( (Ca = 0.79) \). The next identifiable mode is the beads on string structure \( (Ca = 1.05–1.6) \), which has been observed and reported in previous publications\(^{63,64} \). Additionally, a transition mode is observed between the dripping mode and the beads on string structure, where the two modes can be observed intermittently.

By increasing the electric capillary number, beads gradually diminish in size, and the string is transformed into a cone shaped jet \( (Ca = 2.03–4.16) \). This behavior can be attributed to the action of tangential electric stresses on the surface of the string. For viscoelastic solutions, the cone-jet mode is stable across a wider range of potential differences than DI water. Furthermore, as the electric field is augmented, the cone angle increases, and the asymptotic thickness of the jet falls. The cone angle surges until the jet sticks to the annular section of the nozzle \( (Ca = 7.84–10.35) \), which is why this mode was termed the stick jet mode\(^3 \). In DI water, the jet usually ramifies in the downstream of the flow. By intensifying the strength of the electric field, this ramification moves upstream until the multijet mode is reached, while in the viscoelastic
solutions, no ramification is observed downstream of the flow. Finally, if the electric capillary number reaches an instability threshold, the thin jet produced begins to oscillate and shows erratic behavior ($Ca_E = 11.03$). The crucial result that can be derived from this part is that viscoelasticity stabilizes the jet and escalates the stability limit. To avoid redundancy, in Fig. 5, the EHD modes of other concentrations and flowrates are classified according to their dimensionless numbers, where the observed range for every mode is specified.

![Figure 4. Snapshots of the viscoelastic EHD modes for a 100 ppm PAA solution and a 108 mL/h flowrate.](image)

The extent to which each EHD mode is produced in a nondimensionalized operating map is depicted in Fig. 5. This graph clearly shows that by increasing the concentration of the viscoelastic solution, regions pertinent to the dripping and transition modes dwindle, so the beads on string structure is obtained for lower electric capillary numbers. At higher $Wi$, the beads on string structure and the cone-jet mode are observed in a narrower range of electric capillary number, while the stick jet mode shows a reverse trend. As the solution concentration increases, the stick jet mode stabilizes in a wider range of electric capillary numbers. This behavior can be attributed to two simultaneous reasons, namely, the onset voltage for stick jet formation falls, while the stability threshold surges. This is because electric repulsion is neutralized by viscoelastic stresses; therefore, the threshold of thin jet stability is increased, and the stick jet mode, a mode initially
formed through the action of forces of tangential viscoelastic volume on the interface, is observed at a lower electric field strength.

To determine the influence of different parameters on droplet size, images of droplets acquired by high-speed photography were processed using image-processing codes. First, noise on the surface of the droplets and its circumference were eliminated. Using the outer diameter of the nozzle, the size of each pixel was obtained, and the mean diameter of the droplets was calculated using the Riemann sum method. The formation of a thin filament of fluid in the breakup led to the calculation of the diameter of every droplet at the onset of neck formation. The calculated diameters are plotted versus the potential difference for each flow rate and solution concentration in Fig. 6. Possible error in voltage measurement and the standard deviation for the diameters of the droplets are plotted in Fig. 6. It is inferred from the figure that when the flowrate or solution concentration is increased, the drop size also grows, while increased voltage reduces the diameter of the drops. A similar trend has also been observed for DI water and nanosuspension droplets in previous studies, although these were obtained with different nozzle diameters.

Figure 5. Operating range of various viscoelastic EHD modes, plotted according to the electric capillary and Webber numbers for PAA solutions at three concentrations.

Figure 6. Droplet size versus applied voltage for every tested case.

In Fig. 7, the same results are depicted in terms of nondimensional quantities. The droplet diameter in the figure is non-dimensionalized by
the diameter of the same case in the absence of the electric field. The plotting of the results to a single curve shows that the non-dimensionalized droplet diameter only depends on the electric capillary number, and it is independent from the concentration and flowrate of the solution. In addition, the trend given in Fig. 7 is a curve fitted to a line that has the general form of \( \frac{r}{r_0} = a \frac{Ca_E + b}{\text{Ca}_E} \), where \( a \) and \( b \) are -0.408 and 1.02, respectively.

Figure 7. Non-dimensionalized droplet size versus electric capillary number for every tested case.

To investigate the uncertainty further, the distribution of droplet sizes are plotted as normalized histograms for two different cases in Fig. 8. Additionally, the log-normal distribution is plotted for each histogram to produce a better view of the standard deviation and the symmetry of the distribution of droplet sizes. In Fig. 8, it can be explicitly seen that, although the average droplet size falls with increasing electric field strength, the size distribution markedly broadens. This result stems from the fact that when the voltage is increased, the repulsion between the polarized droplets produces minor deviations of droplet ejection from the vertex of the cone. This phenomenon causes increased irregular behavior during the breakup. The results shown in Figs. 7 and 8 have also been found for nanosuspension droplets.

Figure 8. Normalized histograms together with their corresponding log-normal distributions for a 100 ppm PAA solution and a 108 mL/h flowrate: (a) \( \text{Ca}_E = 0.55 \) and (b) \( \text{Ca}_E = 1.08 \).

Next, in Fig. 9, the frequency of droplet generation is plotted against the voltage for every tested case. It is observed that droplet frequency shows an upsurge when the applied voltage or flowrate is increased; however, the effect of rising solution concentration on droplet frequency is nonsignificant. This is because surface tension, the main parameter determining the time of droplet generation, is fairly constant for every solution concentration, while polymeric viscosity alters the severity of elastic behavior during breakup process. The general trend of the data plotted in Fig. 9 is similar to the corresponding
trends depicted for DI water\textsuperscript{13} and nanosuspensions\textsuperscript{4}; nevertheless, the frequency of droplet generation in viscoelastic solutions is substantially lower than in Newtonian fluids because of the increased total viscosity and delayed breakup. For every flowrate, the data plotted in Fig. 9 is curve fitted with a third-order polynomial, and the corresponding equations of curve-fitted diagrams can be found in the figure.

![Figure 9](image.png)

Figure 9. Droplet-generation frequency plotted against the applied voltage for every tested case, together with the third-order polynomial curve-fitted diagrams and their corresponding equations.

When the data in Fig. 9 are rearranged in terms of the electric capillary number and dimensionless droplet frequency, as seen in Fig. 10, we recognize that the dimensionless droplet frequency increases almost linearly with the electric capillary number, but it is barely affected by the solution concentration or flowrate.

![Figure 10](image.png)

Figure 10. Dimensionless droplet frequency, plotted against the electric capillary number for every tested case, along with the curve-fitted line and its corresponding equation.

As in the algorithms used for droplet image processing, the images pertinent to the cone-jet and stick jet modes were processed, and the noise from the captured pictures was eliminated. Afterwards, every jet profile was curve fitted with a power-law relationship of an arbitrary power. The effects of the operating parameters on the stable jet profile are shown in Figs. 11–13. First, we examine the effects of electric field strength on the final diameter of the jet in Fig. 11. It is clear from the figure that the final thickness of the jet plummets when the electric field strength is increased. Because the jet mode is initially formed by the action of tangential electric stresses on the interface, it is expected that the increased electric forces diminish the asymptotic thickness of the jet. On the contrary, Fig. 12 indicates that when the flowrate is increased, a monotonic increase in the diameter of the jet appears for a constant strength of the electric field and solution concentration. By contrast with electrostatic effects, the increasing flowrate magnifies the inertial forces, resulting in a thicker jet. As shown in Fig. 13, although the behavior of the stick jet becomes more evident, the parameters of viscoelasticity have little
influence on the asymptotic jet thickness. The alterations in jet profile are caused by enhanced stretching at the beginning, which is harnessed by tangential electrostatic forces further downstream.

Figure 11. Effects of the electric capillary number on the viscoelastic jet profile for a 100 ppm PAA solution and a 108 mL/h flowrate.

Figure 12. Effects of the Webber number on the viscoelastic jet profile for a 100 ppm PAA solution and a 2.883 electric capillary number.

In Fig. 14, the cone angles, calculated from the slope of the tangential lines on each jet profile, are plotted against the electric capillary number. It is obvious from the figure that the cone angle alterations within each mode are rather small, while a jump in the cone angle is seen in the transition from one EHD mode to another. These alterations are curve fitted with a third-order polynomial, and the corresponding equation for this diagram can be found in the figure.

Figure 13. Effects of the Weissenberg number on the viscoelastic jet profile for a 108 mL/h flowrate and a 2.883 electric capillary number.

Figure 14. The calculated cone angles plotted against the electric capillary number along with the third-order polynomial curve-fitted diagram and its corresponding equation for a 100 ppm PAA solution and a 108 mL/h flowrate.
B. Numerical Results

The governing equations introduced in Section II are discretized with the Petrov–Galerkin finite-element method in axisymmetric coordinates. The coupling between the electric and viscoelastic stresses and flow equations is accomplished using an iterative segregated approach, and the non-linear system of equations is solved using the Newtonian method. For every iteration in a new timestep, first, the electric-potential equation is solved. Then, using the electric stresses obtained from the previous step and the viscoelastic stresses obtained from the previous iteration, the flow equations and the phase-field equation are solved to update the velocity components and the interface position. Finally, the LCM equations are solved using the updated velocity components and the interface position. Iterations continue until a simultaneous convergence is met for all equations. In the following, the proposed algorithm is first validated with consideration of the benchmark problem of the sedimenting sphere, and the acquired results are compared to the previously reported results in the literature. Subsequently, this method is used to simulate an electrified viscoelastic jet and an in-depth analysis is given of the results.

1. Benchmark Problem of the Sedimenting Sphere

To validate the viscoelastic constitutive equations and their implementation, the benchmark problem of a sedimenting sphere is modeled. Many researchers have considered this problem\(^{38, 39}\), nevertheless, the Knechtges study\(^{39}\) is chosen as our main reference due to the similar viscoelastic constitutive equations and the LCM reformulation used in this work. This benchmark problem can easily demonstrate the ability of the LCM to solve sophisticated viscoelastic problems because viscoelastic stresses are resolved in a purely extensional flow in the wake of the sphere while the flow is subjected to a contraction–expansion cross section. The normal component of the conformation tensor in the flow direction is chosen as the main validation factor. The computational domain, shown in Fig. 15, and the physical properties of the fluid, listed in Table III, are identical to the values used in the previous study\(^{39}\).

Figure 15. The computational domain utilized in the benchmark problem of the sedimenting sphere. All dimensions are given in millimeters.

Table III. Physical properties of the fluid used in the benchmark problem of the sedimenting sphere.

| Density (\(kg/m^3\)) | Solvent Viscosity (\(Pa.s\)) | Polymeric Viscosity (\(Pa.s\)) |
|----------------------|-------------------------------|-------------------------------|
| 1000                 | 0.5                           | 0.5                           |

In the current simulation, inertia terms are neglected and the Stokes flow equations are solved. Moreover, the no-slip boundary condition is applied to the wall of the sphere, and the gravity effect is neglected. Uniform velocity and the constant-pressure boundary conditions are applied at the inlet and outlet of the domain, respectively. It should be noted that, due to the long inlet length, flow in the channel becomes fully developed before reaching the sphere. In addition, viscoelastic stresses are assumed to be
zero at the inlet, and their flux is set to zero at the symmetry axis. With the exception of those indicating convection, the terms in LCM equations are treated explicitly using the previous iteration values. A grid study is performed using three different triangular meshes with properties listed in Table IV.

Table IV. Different properties of the triangular meshes utilized in the benchmark problem of the sedimenting sphere.

| Mesh | Number of Domain Elements | Number of Boundary Elements |
|------|--------------------------|----------------------------|
| M1   | 6644                     | 455                        |
| M2   | 11791                    | 537                        |
| M3   | 32206                    | 701                        |

The results are reported for when the solution reaches a steady state. The characteristic length and velocity used in the definition of the Wi are the radius of the sphere and the mean value of the fully developed velocity profile, respectively. It is notable that the computational time required to attain a steady-state solution rapidly increases as the Wi increases, as the wake requires a considerably longer time to develop. Fig. 16 shows the normal component of the conformation tensor on the symmetry axis in the wake area of the sphere for the three mesh sizes listed in Table IV and the three different Wis. The results depicted in the figure show good agreement with the results provided by Knechtges, which supports our implementation of the LCM. As reported previously, when the Wi is increased beyond 1, mesh convergence is gradually lost due to the large growth of stress in the wake of the sphere.

![Figure 16](image-url)
mesh size of M2, where the elongation of the viscoelastic stresses along the flow direction is visible. The elongated stresses are initially formed in the wake of the sphere and grow gradually as the wake grows. These contours follow the pattern of pressure modifications behind the sphere due to the pulling of the wake and the action of drag forces in the shear layer\(^{38}\).

Finally, to find the stability threshold of this benchmark problem, several numerical simulations are conducted for an increasing sequence of \(W_i\)s (Fig. 18). This limit was previously reported by Knechtges\(^ {39}\) to be \(W_i = 1.4\); nevertheless, our results show that the \(W_i\) can be increased up to 2.6 without the appearance of any oscillations in the solution. This increase in the stability threshold is a vital factor for the success of electrospray simulation, where the \(W_i\) is inherently large due to the small characteristic length.

### 2. DI Water Jet Simulation

First, we examine our model by simulating an electrified DI water jet, meaning that viscoelastic effects are temporarily neglected. The domain geometry consists of a nozzle positioned on top of the computational domain, as shown in Fig. 19. The boundary conditions used for solving the electrostatic equations are as follows: a constant potential of 11 kV is applied to the walls of the nozzle, while the lower boundary is set as the substrate. For other boundaries, normal electric displacement is set to zero. The DI water physical properties used in the computation are listed in Table II. To reduce the computational cost, a very fine mesh is used in a narrow region close to the symmetry axis where the jet is developing, and the remainder of the domain is covered by a coarser mesh, with a smooth growth factor. Detailed information concerning the grids are listed in Table V. The electrostatic equations are discretized with the Galerkin finite element method using quadratic shape functions. Similarly, the coupling between the electrostatic and other equations is accomplished using an iterative, segregated method.
Figure 19. Domain geometry of DI water and viscoelastic solution jet simulations. All dimensions are given in millimeters.

Table V. Various properties of the triangular meshes utilized in DI water and viscoelastic solution jet simulations.

| Mesh  | Maximum Element Size (mm) | Number of Domain Elements |
|-------|---------------------------|---------------------------|
| Fine  | 0.004                     | 46971                     |
| Coarse| 0.428                     | 3956                      |

The simulated jet profile for a 108 mL/h flowrate is shown in Fig. 20. In the simulation results, it is demonstrated that miniscule droplets are detached from the tip of DI water jet, in agreement with the results acquired by Narvaez Munoz\textsuperscript{69} and the ramifying behavior observed at the tip of DI water jet in our experimental tests (Fig. 2, \( C_{ae} = 6.31 \)). However, the breakup process is not precisely the same as the experimental results, due to the axisymmetric limitations imposed on the problem. In addition, the distance at which drop detachment occurs in the simulation is approximately eight outer diameters of the nozzle, while the ramifying behavior in experimental tests begins after about 12 diameters.

In Fig. 21, a comparison is made between the simulated jet profile and its corresponding experimental profile obtained from image-processing data. As mentioned earlier, the image processing error equals one pixel, which is also taken into account in the experimental data shown in the figure. It is clear from the figure that the simulation results predict a higher cone angle, and the tip of the nozzle is more wetted in the experiment. On the other hand, the asymptotic thickness of the jet is reached at a smaller length from the nozzle in the numerical results. The observed discrepancy between the two jet profiles may be rooted in several sources, including the deviation of the electric field in the experiment from the ideal field used in the computation, due to the presence of a supporting frame and other measurement devices near the nozzle.
3. Viscoelastic Solution Jet Simulation

The computational domain and mesh size utilized for a viscoelastic electrified-jet simulation are the same as those used in the previous subsection for DI water. The viscoelastic stress equations are discretized using the Petrov–Galerkin finite element method and coupled to the previous equations with an iterative segregated approach, as described earlier. The jet profile for a 100 ppm PAA solution, a 108 mL/h flowrate, and an 11.4 kV applied voltage is demonstrated in Fig. 22. Major differences are observed between DI water and the viscoelastic solution jet profiles. By contrast to DI water, the viscoelastic jet is stable, and no breakup is seen in the results. Similar behavior can be observed in the experiment (Fig. 4, \( \text{Ca}_E = 2.03-7.84 \)). In agreement with the experimental results, for the same operating parameters, the viscoelastic jet is markedly thinner than the DI water jet. These changes in jet behavior can be attributed to the extension of viscoelastic stresses in the main body of the jet when it is deformed against tangential electric stresses.

In Fig. 23, the space charge density contours are shown in the body of the jet. It can be deduced from the accumulation of the space charge density contours at the interface that our leaky dielectric model successfully retains the electric charges close to the interface. Fig. 24 depicts the effects of Wi on a simulated viscoelastic jet profile. As can be seen in the figure, when the Wi is increased, the asymptotic profile of the jet is reached at a smaller length from the nozzle, while the final thickness of the jet is slightly reduced. As noted previously in the experimental results (Fig. 13), the observed alterations in jet profile can be attributed to the vigorous stretching of viscoelastic stresses near the nozzle, which is suppressed by the electrostatic forces further downstream. This suppression is amplified as the jet moves away from the nozzle, and the asymptotic thickness of different jets converge, as is evident in Figs. 13 and 24.
Figure 23. Space charge density contours plotted in the body of the simulated viscoelastic-solution jet for a 100 ppm PAA solution, a 108 mL/h flowrate, and an 11.4 kV applied voltage.

Figure 24. Effects of Weissenberg number on simulated viscoelastic jet profiles for a 108 mL/h flowrate and an 11.4 kV applied voltage.

Fig. 25 compares the simulated results to the experimental data obtained from image processing for viscoelastic jet with operating parameters of a 100 ppm PAA solution, a 108 mL/h flowrate, and an 11.4 kV applied voltage. The discrepancy observed between the two jet profiles is similar to the results for DI water, and the respective reasons are elucidated above.

V. Conclusion

In this study, the influence of viscoelasticity on EHD modes was examined in detail. The change in the mechanical behavior of fluid caused by viscoelastic stresses leads to the observation of new EHD modes. The results of every electrospray test were classified in dimensionless operating maps, where the effects of different parameters on operating range of each mode were examined. By increasing the flowrate or solution concentration, the region of the stable jet, especially the region of the stick jet mode, enlarges, and the fluid stability threshold grows considerably. On the other hand, the regions pertinent to other EHD modes diminish when the flowrate or solution concentration increases. It can also be deduced that the breakup process in viscoelastic solutions is entirely different from that of Newtonian fluids. Additionally, it is noted that the dimensionless droplet diameter and the dimensionless droplet frequency vary almost linearly with the electric capillary number. Although increasing the electric capillary number reduces the average droplet diameter, the distribution of droplet size broadens significantly. If the electric capillary number or solution
concentration increases, the asymptotic thickness of the jet diminishes, while increasing the flowrate thickens the jet. Changing marginally in every mode, the cone angle steeply increases during mode transitions. From numerical simulations, it can be concluded that the stabilization limit for the benchmark problem of the sedimenting sphere increases even though the solution lost mesh convergence when the Wi rose beyond 1. Next, the jet profiles of the DI water and viscoelastic solution were compared with their corresponding experimental profiles. The simulation of viscoelastic jets in an increasing sequence of WIs indicated that the asymptotic profile of the jet is reached at a smaller length from the nozzle, and the final thickness of the jet is slightly reduced. Overall, the effects of viscoelasticity on the simulated jet profiles closely resembled the experimental results.

Appendix A

Here, the equations associated with the decomposition of the velocity gradient transpose, together with the relationships used for the computation of eigenvectors and eigenvalues of the log-conformation tensor, are discussed in detail. If $R$ and its transpose are applied to all the terms of Eq. (13), and Eqs. (17) to (20) are substituted into Eq. (13), the following relationships are obtained:

$$\omega_{ij} = \begin{cases} 0 & \text{if } i = j \\ \frac{\lambda_i m_{ij} + \lambda_j m_{ji}}{\lambda_j - \lambda_i} & \text{if } i < j \\ -\omega_{ji} & \text{if } j < i \end{cases} \quad (A.1)$$

$$n_{ij} = \begin{cases} 0 & \text{if } i = j \\ \frac{m_{ij} + m_{ji}}{\lambda_j - \lambda_i} & \text{if } i < j \\ -n_{ji} & \text{if } j < i \end{cases} \quad (A.2)$$

$$b_{ij} = \begin{cases} m_{ij} & \text{if } i = j \\ 0 & \text{if } i \neq j \end{cases} \quad (A.3)$$

Using these equations, every term in the decomposition of the velocity gradient transpose can be determined. Additionally, the eigenvalues of the log-conformation tensor are computed with the following equations:

$$\lambda_1 = \begin{cases} \frac{\psi_{11} + \psi_{33} + \sqrt{(\psi_{11} - \psi_{33})^2 + (2\psi_{13})^2}}{2} & \text{if } \psi_{13} \neq 0 \\ \psi_{11} & \text{if } |\psi_{13}| \leq \varepsilon \end{cases} \quad (A.4)$$

$$\lambda_2 = \psi_{22} \quad (A.5)$$

$$\lambda_3 = \begin{cases} \frac{\psi_{11} + \psi_{33} - \sqrt{(\psi_{11} - \psi_{33})^2 + (2\psi_{13})^2}}{2} & \text{if } \psi_{13} \neq 0 \\ \psi_{33} & \text{if } |\psi_{13}| \leq \varepsilon \end{cases} \quad (A.6)$$

The eigenvectors of the conformation tensor are computed with:

$$R = \left( \begin{array}{ccc} \frac{1}{A_1} & 0 & \frac{1}{A_3} \\ 0 & 1 & 0 \\ (\psi_{33} - \lambda_3)A_1 & 0 & (\psi_{33} - \lambda_3)A_3 \end{array} \right) \quad \text{if } \psi_{13} \neq 0 \quad (A.7)$$

where $A_1$ and $A_3$ are defined by the following equations:

$$A_1 = \sqrt{1 + \left( \frac{\psi_{13}}{\psi_{33} - \lambda_1} \right)^2} \quad (A.8)$$

$$A_3 = \sqrt{1 + \left( \frac{\psi_{13}}{\psi_{33} - \lambda_3} \right)^2} \quad (A.9)$$

In Eqs. (A.4)–(A.7), $1 \times 10^{-12}$ is considered for $\varepsilon$ to avoid division by zero. The following equations are the expanded form of Eq. (11) in axisymmetric coordinates:

$$F_{p,r} = \frac{\partial \tau_{11}}{\partial r} + \frac{\partial \tau_{13}}{\partial z} + \frac{\tau_{11}}{r} - \frac{\tau_{22}}{r} \quad (A.10)$$

$$F_{p,z} = \frac{\partial \tau_{13}}{\partial r} + \frac{\partial \tau_{33}}{\partial z} + \frac{\tau_{13}}{r} \quad (A.11)$$

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