Effect of the mobility parameter on the oscillatory electro-convection of dielectric liquids subject to strong unipolar charge injection

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Abstract -- Two-dimensional numerical calculations were carried out for the isothermal electro-convection in a dielectric liquid induced by the unipolar charge injection. In the dimensionless form, the system is mainly governed by the injection strength number \( C \), the electric Rayleigh number \( T \) and a mobility number \( M \). A strong injection case with \( C = 10 \) was considered. Along with the increase of the driving parameter \( T \), oscillatory electro-convection occurs. Critical values of \( T \) corresponding to the onset of such oscillatory convection were shown to be highly dependent on \( M \), and were accurately determined. Some precautions including the algorithm for the charge density equation were taken to reduce the numerical diffusion and to ensure the reliability of our findings.

Keywords--Electrohydrodynamics; unipolar injection; numerical simulation; oscillatory convection; non-dimensional mobility number

I. INTRODUCTION

Electro-convection induced by a unipolar charge injection from an electrode into a perfectly insulating liquid between two parallel plates is a fundamental problem in Electro-Hydro-Dynamics (EHD) [1], [2]. This problem has been widely investigated theoretically, experimentally and numerically to understand the transport process of electric charges and the interaction between the electric field and flow fields. When subjected to an external direct-current (DC) field, the electro-chemical reaction at the interface between dielectric liquid and electrode leads to free ions injecting into the bulk liquid [3]. When the electric field is not strong, ion injection usually takes place at one electrode only (unipolar injection) [4]. Once free ions enter into the bulk liquid, the resulting Coulomb force tends to destabilize the flow and to induce electro-convection.

Similar to the Rayleigh-Bénard convection, electro-convection in highly symmetrical configurations (e.g., parallel plates, coaxial cylinders and concentric spheres) attracts a lot of attention for two main reasons. Firstly, the linear stability phenomenon with these symmetrical configurations serves as an important starting point for the theoretical analysis. Secondly, experimental conditions are relatively easily controlled. For example, the space charge limited (SCL) injection can be achieved by covering electrodes with perm-selective membranes [5]. Stability analysis revealed that the electro-convection in the parallel plate configuration was characterized by a linear and a nonlinear stability criterion, which correspond to the infinitely small and finite amplitude perturbations, respectively [6], [7]. Stability criteria are usually expressed through the non-dimensional electric Rayleigh number \( \nu \), which is proportional to the applied voltage. Both the linear and nonlinear bifurcations were qualitatively confirmed by physical experiments [5], [8]. However, some discrepancies between theoretical and experimental results remain unexplained. For example, for SCL injection, analytical value for the linear criterion is 161 [6] while experimental finding is about 100 [7]. In addition, at the onset of convection, an unsteady flow with fluctuating around some averaged level was experimentally observed, which has never been explained reasonably.

Since 1987, the direct numerical analysis approach has been used to investigate the electro-convection between parallel plates [9], [10]. In numerical studies, the core difficulty lies in the algorithm for the conservation equation of charge density. Some ‘special’ schemes, such as the particle in cell (PIC) [11]-[13], flux corrected transport (FCT) [12], [14] and total variation diminishing (TVD) [15]-[17] have been used to compute the charge density distribution. However, most numerical studies focused on the finite amplitude regime, which means the driving parameter is close to the linear stability criterion.

Along with the driving parameter increasing, more nonlinear instabilities generally set in and oscillatory or even chaotic flows appear. Experimental results showed dramatically chaotic flow patterns with \( T \) parameter much higher than the stability criterion [8]. Some effects have also been made to distinguish the viscosity and inertial effects dominated convection [5]. Recently, numerical simulations have been performed for strong injection case with high \( T \) values. In [14], Vázquez et al., presented results for \( T = 500 \) and a new structure of two convective cells was reported.

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[16], Traoré et al., varied the $T$ parameter in a wide range. The two cell structure was reproduced with $T = 300$. In addition, a regular oscillation flow pattern was observed with $T = 600$, and a preliminary route to the final chaotic (including electro-plumes) state was defined. Kourmatzis and Shrimpton [18] presented three dimensional results of $T = 500$ in a large cavity with the aspect ratio of 5 or 6. An unsteady flow owning a large number of eddies with different scales was shown.

Besides the electric Rayleigh number, there are two other non-dimensional parameters governing the system. One is linked to the injection strength at the emitter (labeled as $C$) and the other one represents the mobility property of the injected ions (labeled as $M$). It should be noted that all above mentioned studies always fixed $M$. In a very recent paper [19], the flow structure of the weak injection at the threshold of linear stability has been shown to be highly dependent on $M$. In this study, we will show later that this mobility number also greatly affects the critical values of $T$ separating the steady and oscillatory convections.

Similar to the oscillatory thermal convection in the Rayleigh-Bénard system [20]-[22], the occurrence of oscillatory electro-convective is also highly sensitive to the numerical diffusion introduced during the computation. Thus some precautions are required to minimize numerical diffusion. For unsteady flows, numerical diffusion is determined together by the spatial and temporal schemes, grid size and time step.

The present study can be viewed as the first step for accurately determining the route to chaos for electro-convection between two parallel plates. In this study, we will first highlight that numerical diffusion greatly affects the numerical estimation of critical $T$ separating the steady and oscillatory convections. Secondly, reference values of $T$ for several $M$ will be provided. The reminder of this paper is organized as follows: the physical problem and governing equations are stated in Section II. Section III briefly explains of the numerical method. In Section IV, the numerical results are presented. The paper is concluded in the last section.

II. PROBLEM FORMULATION

We consider a perfectly insulating liquid layer of thickness $H$ enclosed between two parallel electrodes of length $L$. The liquid is assumed to be incompressible, Newtonian and isothermal. Fig. 1 shows a schematic of the two-dimensional system. A potential difference $\Delta V = V_1 - V_0$ is applied between the two electrodes. Free charges are assumed to be injected only from the electrode with higher electric potential (unipolar injection). To simply the problem, the injected charge density at the emitter ($q_0$) is assumed to be constant and uniform (autonomous and homogeneous).

Electro-convection is modeled by the following equations of continuity, momentum, charge density, and electric potential in dimensionless form [11]:

$$\nabla \cdot \vec{U} = 0$$

$$\frac{\partial \vec{U}}{\partial t} + (\vec{U} \cdot \nabla) \vec{U} = -\nabla \tilde{p} + \frac{M^2}{T} \Delta \vec{U} + M^2 q \tilde{E}$$

$$\frac{\partial \tilde{q}}{\partial t} + \nabla \cdot (q \tilde{U} + \tilde{E}) = \alpha \Delta \tilde{q}$$

$$\Delta \tilde{V} = -q$$

$$\tilde{E} = -\nabla \tilde{V}$$

where $\vec{U} = [u, v]$ and $\vec{E}$ are the velocity and electric fields, respectively. $\tilde{p}$ is the modified pressure including the electrostriction force’s contribution [2]. $\tilde{q}$ denotes the charge density. The last term in Eqn. (2) stands for the driving Coulomb force.

![Fig. 1. Schematic of the system.](image)

The dimensionless variables are defined as follows: $H$, $\Delta V$, $\varepsilon \Delta V / H^2$ and $K \Delta V / H$ for, in order, length, electric potential, charge density and velocity. $\varepsilon$ and $K$ represent the dielectric permittivity and ionic mobility. The system is then governed by the following four non-dimensional parameters,

$$T = \frac{\varepsilon \Delta V}{\rho v K}, \quad C = \frac{q_0 H^2}{\varepsilon \Delta V}, \quad M = \frac{1}{K} \left( \frac{\varepsilon}{\rho} \right)^{1/2}, \quad \alpha = \frac{D}{K \Delta V},$$

where $\rho$ and $v$ are the density and viscosity of the fluid, and $D$ is the charge-diffusion coefficient. $T$ is the electric Rayleigh number, which is the ratio between the Coulomb force and the viscous force. $C$ represents the injection strength. $M$ is the non-dimensional mobility parameter, which is the ratio between the so-called hydrodynamic mobility to the actual ionic mobility. From the definition equation, we know that $M$ depends only on fluid and ion properties. For most ions in liquids, its typical value is higher than 3 [2], [22]. $\alpha$ is the non-dimensional charge-diffusion number with its typical value in the range between $10^3$ and $10^4$ [23]. Because of the small value, the diffusion mechanism for the charge transport is always neglected for the stability analysis in the finite amplitude regime.

The boundary conditions are as follows: the no-slip conditions are applied to horizontal electrodes for velocity and symmetry conditions are applied to the two vertical sides aiming at keeping consistence with the previous studies [14],
the half wave-length of the most unstable mode of the charge density equation, please refer to [17], [32].

For the purpose of comparison, the classic first order upwind (FUD) scheme and the solution physically bounded [29], [30]. For the purpose of temporal derivatives in Eqn. (3). Both SMART and RK3TVD Runge-Kutta scheme (RK3TVD) [28] for the convective and boundary conditions, were discretized using a finite volume interpolation, respectively.

Special attention is required for solving the charge density equation. As we already mentioned in the introduction section that the method for this equation significantly affected the global performance of the numerical simulation with EHD free convection. In this study, we applied the third order SMART algorithm [27] and the third order optimal TVD scheme (RK3TVD) [28] for the convective and temporal derivatives in Eqn. (3). Both SMART and RK3TVD are specially designed for strong convection dominated problems, aiming at reducing numerical diffusion and keeping the solution physically bounded [29], [30]. For the purpose of comparison, the classic first order upwind (FUD) scheme and explicit Euler (EE) were also used. Both the SMART algorithm and FUD scheme were converted into the upwind based unified formulation [31] for easy implementation. For more details about the application of TVD scheme in solving the charge density equation, please refer to [17], [32].

III. NUMERICAL METHODS

The governing equations, Eqns.(1)-(4), together with the boundary conditions, were discretized using a finite volume approach on a collocated grid. All equations are solved in an iterative segregated way. For Navier-Stokes equations, spatial derivatives (convection and diffusion terms) were discretized with the second order central differencing (CD) scheme, while the time derivative was discretized with the second order semi-implicit three time levels scheme [24]. The SIMPLE algorithm [25] and Rhie-Chow algorithm [26] were undertaken for the velocity-pressure coupling and momentum interpolation, respectively.

In this study, we numerically proved that both \( T_c \) and \( T_f \) were independent of \( M \). For \( C = 10 \), typical values for \( T_c \) and \( T_f \) are 164.1 and 111.7, respectively.

A basic requirement for the numerical analysis is the accurate reproduction of these criteria. In Fig. 2a and 2b, we plotted time histories of the electric current through the collector and the angular momentum in the domain with the case of \( T = 200 \). Electric current is defined as

\[
I = \int_{\text{domain}} \left[ \frac{\partial E_y}{\partial t} \right] + q(U_y + E_y) dx,
\]

where \( U_y \) and \( E_y \) are the vertical components of the velocity and electric fields; while the angular momentum is defined as

\[
AM = \int_{\text{domain}} (\vec{r} - \vec{r}_0) \times \vec{U} dS,
\]

where \( \vec{r}_0 \) is the position vector of a given point (generally the central point). Both electric current and angular momentum experienced an exponentially growing stage before reaching the steady convective state. The exponential growth rates were extracted to determine the linear stability criterion [10], [16], and our numerical results were presented in Table 1. We observed that values of \( I \) and \( AM \) at steady convective state were not sensitive to \( M \), which was well explained in [19]. The flow structure with one convective cell was highlighted in Fig. 3, in which we presented iso-contours of charge density and stream function. The representative region avoid of charges was vividly seen in the central area of Fig. 3a.

Starting with the convective state and gradually decreasing \( T \), we observed that the strength of motion also gradually decreased until a critical value, at which the motion suddenly stopped. This critical value corresponds to the nonlinear stability criteria, \( T_c \), and our numerical results were summarized in Table 1. For all \( M \) values, our numerical predictions of \( T_c \) and \( T_f \) are in good agreement with analytical values, which validate the accuracy of our solver. In this study, we numerically proved that both \( T_c \) and \( T_f \) were independent of \( M \) for the first time. As demonstrated in our
previous study [19] that numerical $T_e$ was very sensitive to numerical diffusion, we used a grid with 100x200 control volumes (CVs) to obtain results in Table 1.

### Table 1

<table>
<thead>
<tr>
<th>$M$ parameter</th>
<th>$T_e$</th>
<th>$T_f$</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>163.9</td>
<td>[108, 109]</td>
</tr>
<tr>
<td>10</td>
<td>163.1</td>
<td>[108, 109]</td>
</tr>
<tr>
<td>50</td>
<td>163.2</td>
<td>[109, 110]</td>
</tr>
<tr>
<td>100</td>
<td>163.5</td>
<td>[108, 109]</td>
</tr>
<tr>
<td>200</td>
<td>163.5</td>
<td>[108, 109]</td>
</tr>
</tbody>
</table>

Note: Theoretical values for $T_e$ and $T_f$ are 164.1 [6], and 111.7 [7], respectively.

### B. Nonlinear bifurcations with higher electric Rayleigh number

A steady two-cell structure has been observed in [16] with $T = 300$ and $M = 10$, in [18] with $T = 500$ and $M = 22.4$, an unsteady two-cell structure in [14] with $T = 400, 500$ and $M = 20$. The unsteady behaviors in [18] is attributed to the PIC method used for the charge density equation, since even with $T = 178$ an unstable convection was always obtained [11]. In this study, we also observed the steady two-cell structure in a wide range of $T$ (see Fig. 4 and 5). Actually, there exists a critical $T$ value which separates the one- and two-cell structure (labeled as $T_{c2}$). This critical value is physically related to $M$ and also numerically affected by the numerical diffusion. For $M = 10$, the two-cell structure was obtained even with $T = 290$ when we carefully controlled the numerical diffusion by using a fine grid of 100 x 200 CVs and time step of 10$^{-5}$. For $M = 50$, with the same grid and time step, the two-cell structure was only observed with $T$ higher than 340. Other results of $T_{c2}$ were presented in Table 2.

Fig. 3. Numerical results at the steady convective state with $C = 10, T = 200$ and $M = 100$.

(a) Electric current

(b) Angular momentum

Fig. 4. Time histories of (a) electric current with various $M$ and (b) angular momentum with $M = 100$. Parameters: $C = 10$ and $T = 460$. In Fig. 4a, we presented the time history of the electric current with $T = 460$. We saw that all $M$ values led to a final steady convective state of two-cell structure after an irregular transition process from the one-cell structure. Again values of electric current at the final state for $M = 50, 100$ and 200 are almost indistinguishable. The transition process was more easily seen in Fig. 4b where we presented the angular
momentums of the left and right half domain, and of the whole domain. For time \( t < 20s \), the evolution route of \( AM \) with the central point being the reference point was similar to Fig. 2b and only one convective cell was formed. Then the one cell was separated into a pair of cells with the same intensity but opposite directions, which was expressed through the same absolute values of \( AM \) with the left and right central points as the reference points. The highly symmetrical flow structure was also clearly shown in iso-contours of charge density and stream function (Fig. 5).

Above another critical \( T \) value, the two convective cells started to slowly interact with each other and the flow exhibited a periodic oscillation behavior in both space and time. Such a transition from steady to unsteady convection is due to Holf bifurcations, which have been widely discovered in nonlinear dynamic systems. In the viewpoint of stability theory, the nonlinear system has evolved from the fixed critical point to the periodic attractor [33]. We name this critical value as the criteria for the onset of oscillatory convection. From these results we concluded that the SMART scheme combined with the RK3TVD scheme appeared to the most appropriate choice for the present oscillatory convection.

![Fig. 6. Transient responses of the maximum vertical velocity component for \( C = 10, T = 485 \) and \( M = 10 \).](image)

![Fig. 7. Power spectrum for the regular periodic signal of \( f_{osc} \) in Fig. 6.](image)

Only with the SMART and RK3TVD schemes on the fine grid, a regular oscillation was observed. With a spectra analysis with the periodic signal of Fig. 6, a basic frequency and several ultra-harmonic frequencies were observed (see Fig. 7). Such an unsteady flow was further confirmed with the same schemes with a finer grid of \( 150 \times 200 \) CVs and a smaller time step of \( 10^{-4} \). It is easy to understand the discrepancies in solutions of Fig. 6 since numerical diffusion always tends to damp out small oscillations and stabilize the system. Numerical diffusion introduced by the low order spatial and temporal schemes or the coarse grid in other three cases prevented the appearance of regular oscillations. Even with the fine grid, the FUD scheme showed the worst result with a steady one-cell structure, indicating that the FUD scheme is inappropriate for the simulation of free electro-convection. From these results we concluded that the SMART scheme combined with the RK3TVD scheme appeared to the most appropriate choice for the present oscillatory convection.

In Fig. 6, we took the case of \( T = 485 \) and \( M = 10 \) as a representative example to show the transient responses of the maximum vertical velocity component in the domain. Two spatial (FUD and SMART) and two temporal (EE and RK3TVD) schemes were used. The time step was set to be a small value, \( 2 \times 10^{-4} \), and two grids (\( 50 \times 100 \) and \( 100 \times 150 \) CVs) were used to represent the fine and coarse mesh sizes.

Above discussion with the numerical diffusion reminds us to reconsider the consequence of neglecting the diffusion mechanism for charge transport. For numerical analysis, solutions do not differentiate numerical and physical diffusion. In other words, the ‘inaccurate’ result of ‘SMART (50×100) & RK3TVD’ for \( \alpha = 0 \) in Fig. 6 may be the ‘accurate’ phenomena with a small value of \( \alpha \). As the non-dimensional coefficient (\( \alpha \)) is small, the diffusion term in Eqn. (3) is often neglected when performing both the theoretical and numerical
analysis. For $T$ close to $T_c$, we numerically illustrated that the stability criteria and flow structure with the strong injection regime were nearly not affected by this small value [32]. However, when $T$ is much higher than $T_c$, more nonlinear stability bifurcations appear, and these nonlinear behaviors are sensitive to diffusion which is either from the numerical aspect or the physical diffusion mechanism. Let us take $M = 10$ and $\alpha = 5 \times 10^{-4}$ (a typical value in the range of $[10^{-4}, 10^{-3}]$) as the example, our numerical values for $T_{c2}$ is in the range of $[420, 430]$, which is much higher than $[285, 290]$ for $\alpha = 0$. Since the value of $T$ in experimental studies is generally much higher than $T_c$, it may be more desirable to take into account the small diffusion term when trying to numerically reproduce experimental phenomena.

In Table 2, we summarized our numerical predictions of $T_{c2}$ and $T_{c3}$. It is interesting to note that both $T_{c2}$ and $T_{c3}$ increase with $M$ parameter. Now, we further make an explanation of the stability behaviors. In the convective problem, as simulated in this paper, the viscosity tends to stabilize the system by dissipating the fluid energy. When $T$ increases, the viscous effect decreases. Therefore, the unstable region of the system expands and the stable region shrinks. The parameter $M$ has dual effects on the stability nature of this system: stabilizing and destabilizing effects. First, when $M$ increases, the viscous term is enhanced and the system is stabilized. Such stabilizing effect accounts for the higher $T$ requirement for the onset of oscillatory motion with higher $M$. On the other hand, when $M$ increases, the electric driving force increases and destabilizes the system. Actually, we already observed that, once the flow was in the oscillatory pattern, the amplitude of oscillation increased with $M$ [33]. Based on our numerical, we found that in the stable region, the destabilizing effect was more significant than the destabilizing effect, while in the unstable region, the destabilizing effect is dominant. The destabilizing role of $M$ may also explain the higher amplitude of oscillation shown with PIC method in [11], [14].

<table>
<thead>
<tr>
<th>$T_{c2}$</th>
<th>$\alpha = 0$</th>
</tr>
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<tbody>
<tr>
<td>$[285, 290]$</td>
<td>$[335, 340]$</td>
</tr>
<tr>
<td>$T_{c3}$</td>
<td>$[480, 485]$</td>
</tr>
</tbody>
</table>

Note: results are obtained with SMART and RK3TVD schemes, grid of $100 \times 150$ CVs and time step of $2 \times 10^{-7}$.

V. CONCLUSION

In this paper, we performed a numerical investigation of electro-convection induced by strong unipolar charge injection in a plane layer of dielectric liquid. Along with the increasing of the driving electric Rayleigh number ($\gamma$), the system firstly experienced the transition from a single cell structure to a two cell structure, and then a regular oscillation showed. Unlike the well-known linear and nonlinear stability criteria in the finite amplitude regime, we numerically demonstrated that the criteria separating different flow patterns depended on the mobility number ($M$). High resolution spatial and temporal schemes were shown to be necessary for the simulation of unsteady electro-convection since numerical diffusion greatly affected oscillatory convections. Some reference values of $T$ criteria for three $M$ values (10, 50 and 100) were accurately determined. The route to chaos for electro-convection is an important issue to understand the observed experimental phenomena. It will be very interesting, in a future work, to accurately determine such a route and to investigate the role of $M$ parameter played.

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