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Injection induced instabilities and chaos in electrohydrodynamics

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Abstract. The Coulomb force exerted by an electric field on any charge present in a dielectric liquid may induce fluid motion. At high applied electric fields in an industrial grade insulating liquid, charge carriers are created at metallic/liquid interfaces, a process referred to as ion injection, and result from electrochemical reactions. Recently it has become possible to reproduce these injection processes and carefully designed experiments may be related to analytical models. In this review we shall focus attention on the electrohydrodynamic instabilities and chaos induced by unipolar charge injection.

1. Introduction

Electrohydrodynamics (EHD) is a mature science, which can be regarded as a branch of electrodynamics of moving media concerned with the interaction of dielectric fluids with electric fields, and its applications form the basis of major industries [1]. Recently fundamental advances in the electrodics of metal/liquid interfaces, together with new techniques to control the injection of charge and novel methods of purification of insulating liquids, have permitted us to relate analytical models to a set of carefully controlled experiments. The motive force in these experiments is the Coulomb force acting upon the charge carriers that have been injected at the electrode/liquid interface by means of electrochemical reactions. The aim of this paper is to consider the EHD instabilities and chaos produced by Coulomb forces in these insulating liquids when subjected to stationary DC voltages. The EHD phenomena produced when a step voltage (or an AC voltage) is applied will not be considered (see [2] for a review on these transient regimes). Also the case of non-isothermal conditions will not be examined, as a recent review has been given in [3].

2. Definition of the problem

2.1. Basic equations

Since the hydrodynamics of a dielectric fluid is quasi-electrostatical, the Maxwell equations reduce to the irrotational nature of the electric field $\boldsymbol{E}$, the Gauss law and the

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charge conservation equation. The equations of motion are those of a viscous incompressible liquid, i.e. the Navier–Stokes equation and the continuity equation. The only modification is a body force of electrical origin, $\boldsymbol{f}_e$ given by [1]

$$\boldsymbol{f}_e = q\boldsymbol{E} - \tfrac{1}{2}E^2\nabla\epsilon + \tfrac{1}{2}\nabla\left[\rho\left(\frac{\mathrm{d}\epsilon}{\mathrm{d}\rho}\right)_\Theta E^2\right]$$

where q is the free charge density, ϵ the permittivity, ρ the mass density and Θ the absolute temperature. The first term, called the Coulomb force, is the force per unit volume on a medium containing free electric charge. It is the strongest EHD force term and usually dominates when DC electric fields are present. The second term, called the dielectric force, is due to the force exerted on a dielectric liquid by a non-uniform electric field. It is usually weaker than the Coulomb force and only dominates when an AC electric field of high enough frequency is imposed. The third term, called the electrostrictive term, being the gradient of an scalar is treated as a modification to the fluid pressure.

2.2. Constitutive law for the current density

In dielectric liquids of high enough resistivity Ohm's law and electroneutrality often fail to be valid. Conduction in these insulating liquids when subjected to intense electric fields is mainly controlled by electrode/liquid interface phenomena that create new charge carriers of the same polarity as the electrode that are injected into the liquid. The constitutive law for the current density that results from the passage of the injected ions is

$$\boldsymbol{J} = qK\boldsymbol{E} - D\nabla q + q\boldsymbol{v}. \tag{1}$$

The first term gives the contribution of the injected ions that under the influence of the electric field move relative to the fluid with a velocity $K\boldsymbol{E}$ with K the ionic mobility. The second term accounts for molecular diffusion with D the charge diffusion coefficient. The third term is due to convection of charge density by the velocity field.

2.3. Charge relaxation

Using Gauss's law and equation (1) in the charge conservation equation, neglecting diffusion, we have

$$\frac{\mathrm{d}q}{\mathrm{d}t} \equiv \frac{\partial q}{\partial t} + (K\boldsymbol{E} + \boldsymbol{v})\cdot\nabla\boldsymbol{v} = -\frac{K}{\epsilon}q^2$$

whose solution is $q = q_0(1 + t/\tau)^{-1}$ with $\tau = \epsilon/Kq_0$ the algebraic bulk relaxation time. This solution is valid on $\mathrm{d}\boldsymbol{r}/\mathrm{d}t = K\boldsymbol{E} + \boldsymbol{v}$, so that unless a given element of liquid can be traced via a particle line to a source of charge, it will support no bulk charge density.

2.4. Boundary conditions

For the mechanical problem the boundary conditions are the usual no-slip conditions at the electrodes, $\boldsymbol{v} = 0$. For the Poisson equation they become $\phi = \phi_0$ on the injector and $\phi = 0$ on the collector. For the charge density equation we need to specify

the way in which charge is injected into the liquid at one boundary and removed at the other. An injection law relating charge density to electric field has been deduced and experimentally verified [4] for bare electrodes. It turns out that for an ample range of values of field strength ($10^3 \leq E \leq 5 \times 10^4$ V cm^{-1}) the charge density is approximately constant at the injector. On the other hand electrodes covered with appropriate electrodyalitical membranes or varnishes behave as strong injectors [5], being quite possible to reach the space-charge-limited-current (SCLC) regime, characterized by $q_0 = \infty$ or equivalently $E_0 = 0$ at the injector. For simplicity the hypothesis of autonomous injection is usually adopted, i.e., $q = q_0$ at the emitter independently of E. Also we assume that the collector is a completely open electrode. Anyway the effect of giving a more realistic ejection law would be to produce, at most, charge-diffusive boundary layers at the collector, without affecting the mainstream solution of our problem. It should be noted that this latter condition has to be applied only when diffusion cannot be neglected and this will only happens in some limiting situations (see [6] for a detailed discussion).

3. Instabilities and chaos

3.1. Linear instability

For isothermal dielectric liquid layers, under DC conditions, the dominant electric force is the Coulomb force, $q\boldsymbol{E}$. In general geometries it is not possible to have a motionless state for the liquid because of the rotational nature of the electric forces. For simple boundary conditions this is possible only in planar, cylindrical and spherical geometries. For these situations the charge density decreases away from the injector. This is a potentially unstable situation, and it is quite similar to the Rayleigh–Benard problem. In effect, the heuristic argument of Rayleigh may be translated 'literally' with the only difference that the relaxation time for the displaced parcel, of size d (d being the distance between the electrodes) is now due to Coulomb repulsion instead of molecular diffusion (see [7] for a detailed discussion of Rayleigh's argument)

$$\underbrace{\delta q\, \boldsymbol{E} d^3}_{\text{differential Coulomb force}} \sim |\nabla q| \tau w \boldsymbol{E} d^3 \sim \frac{q}{d}\frac{\epsilon}{Kq} w \frac{\phi_0}{d} d^3 \geq \underbrace{\eta \frac{w}{d^2} d^3}_{\text{viscous drag}}$$

where w is the perturbed velocity, $w\tau$ the distance travelled by the particle before the charge relaxes and η the dynamic viscosity. Thus at the unstable equilibrium point $T \geq \mathrm{O}(1)$ with $T = \epsilon\phi_0/K\eta$ the electrical stability parameter. This rough estimate does not give any quantitative value of the critical stability parameter, but merely singles out the appropriate combination of parameters. As $K\eta \sim Cte$ according to Walden's law the physical magnitude characterising stability is the electrical potential. The numerical values of T_c depends on the non-dimensional injection strength C given by $C = q_0 d^2/\epsilon\phi_0$. See [8–10] for quantitative values in planar, spherical and cylindrical geometries. For SCLC regime in planar layers is $T_c \simeq 161$ whereas the experiments give a value of $T_c \simeq 100$. This discrepancy, due probably to the simplicity of the model, remains to be explained.

The analogy with the Rayleigh–Benard problem extends also to other situations where we have rotation [11], Taylor–Couette flow [12] or forced parallel flows [13, 14].

3.2. Non-linear behaviour

The analogy to the Rayleigh–Benard problem is no longer valid for the liquid in motion [15]. The bifurcation around the linear criterion is subcritical and there exists a non-linear criterion. Associated with these two criteria there is a hysteresis loop, at which discontinuities occur in the steady-state current and velocity amplitude. This behaviour may be easily understood, at least qualitatively, from the averaged Navier–Stokes equation. Near the instability thresholds, inertial effects may be neglected and an approximation for the velocity field, which is good within a few percent, is given by a self-similar convective roll. Taking as scales d for distance, ϕ_0 for potential, q_0 for charge and $K\phi_0/d$ for velocity, we have from the balance of driving force and viscous resistance [16]

$$A\int_{\text{cell}} \boldsymbol{v}_0 \cdot \nabla^2 \boldsymbol{v}_0 \, \mathrm{d}s + T\int_{\text{cell}} q\boldsymbol{E} \cdot \boldsymbol{v}_0 \, \mathrm{d}s = 0$$

where the dimensionless velocity is $\boldsymbol{v} = A\boldsymbol{v}_0$, with $\max|\boldsymbol{v}_0| = 1$. The solution of this equation gives T as a function of A, showing the existence of the two criteria and the hysteresis loop. For steady finite amplitude electroconvection is found always to be $A \geq 1.5$ and there exits an inner surface, called the separatrix, separating a region free of charge from a charged one. Finite-difference methods as well as modal analysis give precise quantitative values for these criteria. In particular modal analysis [15] has shown that, in accord with experiments in the SCLC regime, the motion organizes itself in the form of hexagonal convective cells, where the liquid flows toward the injecting electrode in each cell center with a velocity greater than the ionic drift velocity. The computed theoretical value $T_f \simeq 110$ compares favourably to the experimental one, $T_f \simeq 90$.

3.3. EHD chaos

Subsequent experimental studies in the case of the SCLC regime have revealed that small fluctuations of the electrical current around its mean value are always present [17]. Unlike the case of the Rayleigh–Benard problem the liquid always passes directly from rest to time-dependent motion. For small $\Gamma = R/d$ (R being the radius of the electrodes) the frequency power spectra of current fluctuations are discrete; they consist of one fundamental peak f_1, its harmonics and its subharmonics. By slightly increasing ϕ, one obtains biperiodic motion and then continuous spectra. For large Γ the spectra are always continuous but exhibit an enlarged peak corresponding to the same fundamental oscillation as for small Γ. The f_1 variations with ϕ have been studied and it appears to vary in proportion to the main velocity. An experimental determination of the EHD strange attractor, following the Grassberger–Procaccia method, indicates that its dimension increases without limit with the embedding space following a trend similar to that of grid turbulence [18]. This is in striking opposition to the Rayleigh–Benard case where it has been established that the dimension of the corresponding strange attractor is of order 3.

No theoretical explanation exists for this chaotic behaviour, with the exception of a proposed mechanism that suggests that the main frequency peak, measured in the fluctuations power spectra, may originate from an oscillatory instability of the finite amplitude steady convection in each cell. Examination of this instability is a hard question due to the intricate coupling between charge and velocity fields. The much

simpler problem of determining the ion trajectories when a periodic velocity field is superimposed on the steady finite amplitude convection reveals that ion trajectories in EHD convection become chaotic [19]. The separatrix gives place to a heteroclinic tangle and a resonance chaotic layer, the latter comprising a finite region of both injector and collector electrodes. We conjecture that this chaotic behaviour is at the basis of the chaotic behaviour of EHD flows near the instability thresholds. Numerical simulations using particle-type methods [20] and the method of characteristics give encouraging results but more studies are needed to understand this EHD chaos.

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