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Electrokinetic effects in the breakup of electrified jets: A Volume-Of-Fluid numerical study

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ABSTRACT

The breakup of a charged liquid column is studied numerically using Volume-OF-Fluid (VOF) for a range of timescales where electrokinetic phenomena may become significant, i.e when the time to breakup becomes comparable or shorter than the diffusion and the electroosmotic migration times of charged species. Here we propose a conservative method to deal with the diffusion of a tracer in VOF schemes when the diffusion is limited to one of the phases. The method consists in weighing the diffusivity with the value of the volume fraction computed from the analytically reconstructed interface. In this way, the interface is made impermeable to the tracer, which is conservatively kept within one of the phases. The performance of this method is first tested by comparing simple configurations with existing analytical solutions. In the cases when the diffusion, electroosmotic motion and hydrodynamic singularities compete, the results indicate that, after breakup, charges distribute between droplets differently from models assuming homogeneous and constant electrical conductivities (i.e. no electrokinetic effects). However, such departure does not alter the main hydrodynamic balances leading to well-established scaling laws of breakup.

Introduction

Electrokinetic effects in liquids determine bulk charge distributions when existing ionic species in solution respond to applied electric fields, but may also contribute to the global mechanical behavior of the system when free surfaces and interfaces are present. For ordinary values of surface tension, these effects naturally take place when the length scales of the system are below the millimetric scale, and particularly at the micro- and nano-scale in general microfluidic systems.

Among these systems, electrospray is probably the most studied and exploited natural example of global hydrodynamic consequences of electrokinetic effects in the presence of free surfaces. The extensive literature on the physics and biochemical analysis applications of electrospray amounts to more than 10^5 papers and a vast, complex network of citations. However, as puzzling as it may be, this network does not necessarily reflect a complete or sufficient scientific understanding. This may be a consequence of the fast growth rate of this publication network, compared to the average rate of general scientific knowledge assimilation.

In particular, while the basics of electrokinetics were established early, a detailed analysis of relevant publications up to the present day reveals striking understanding gaps in the real sequence of electro-physical processes taking place at the smallest scales: not only in cone–jet electrospray but also in many other phenomena such as the breakup of charged capillary liquid jets. These sequences determine the macroscopic outcome of electrospray in terms of issued charges per unit time and characteristic length scales of liquid emissions (droplets or particles). Interestingly, one may also observe how basic inconsistencies and customary assumptions become fossilized in the foundations of an ample literature.

de la Mora and Loscertales (1994) postulated a set of scaling laws for the electric current and characteristic scales of liquid emission in the form of a jet issued from Taylor cones. These scaling laws were derived from the assumption that electrokinetic migration, or free charge relaxation towards the surface, was halted (or "frozen") at the apex of the cone as the jet scale was reached. A more relaxed version assuming that the jet scale emerged where the relaxation times of free charges became comparable to hydrodynamic residence times led to identical results. Even earlier, Gañán-Calvo et al. (1993) suggested the

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opposite assumption (i.e. short electrical relaxation times compared to hydrodynamic ones) to reach alternative scaling laws, subsequently revisited in Gañán-Calvo (1997, 1999, 2004) and Gañán-Calvo and Montanero (2009) and numerically confirmed in Hartman et al. (1999). Both extreme assumptions and corresponding models recognize electrokinetic effects as the ultimate reason for the appearance of driving forces deriving from Maxwell bulk stresses in the presence of interfaces. However, their extreme nature, the striking relative proximity of their results, and the inextricably indirect way to experimentally verify their validity have hardly been of help to build true and deep scientific knowledge in the community. In particular, the early introduction in the former model (de la Mora and Loscertales, 1994; de la Mora, 2007) of a fitting function $f(\varepsilon)$, where ε is the electric permittivity of the liquid relative to vacuum, helped many subsequent authors to fit this model to their experimental results (e.g. Chen et al., 1995; Chen and Pui, 1997 among hundreds of works).

The rapid evolution of computational power and the increase in efficiency and precision of numerical schemes and methods have paved the way to the widespread but bold idea of tackling scientific conundrums like the one above via numerical simulation. In reality, the physics of fluid motions at the microscopic scale exhibits many features making their study particularly appealing to numerical modeling and simulation. In general, the small scale generally characterizing fluid motions in microfluidic systems limits the relative weight of convective effects compared to diffusion. This feature, reflected in moderate to small Reynolds number values, provides the adequate traits for full numerical simulation, where a high predictive power and accuracy has already been demonstrated. In general, the Lab-On-Chip (LOC) research community has a background in biology-related issues and are less familiar with engineering aspects such as numerical simulation and its advantages. Indeed, Boy et al. (2008) wrote that their focus article on available computational methods for LOC systems, could serve "...to convince the LOC community that computation is a valuable tool and should be increasingly used over the next decade ...". In particular, numerical simulations allow researchers to determine in a rapid step how a design decision can affect the performance of a particular device. This way the development cost would drastically drop by reducing the number of prototyping iterations (Boy et al., 2008).

More recently, Wörner (2012) exhaustively classified and described the foundations of the diverse numerical methods for two-phase flows and performed a complete review on the state of the art of numerical procedures to deal with challenging problems such as moving boundaries, Marangoni effects and surfactants, or heat and mass transfer across the interfaces. In spite of the strong predictive capabilities developed in this field, two-phase micro-flow problems involving the behavior of ionic species under the action of electric fields require an even deeper degree of physical insight. Contrary to what might initially appear, these problems are ubiquitous in fields handling several fluid phases at microscopic scales in predictive and consistent ways, which range from modern chemical engineering, biophysics, pharmaceutical research, to modern food processing, to name a few. Several instruments based on electrokinetic phenomena like the ζ-potential have even been developed (Kirby, 2010). However, those problems have traditionally been tackled following drastic electrokinetic simplifications that either assume (i) complete relaxation of all free charges at free surfaces, where bulks are neutral with homogeneous electrical conductivities (i. e., the leaky dielectric model of Melcher and Taylor (1969), Saville (1997), which entails having hydrodynamic times long compared to electrical relaxation), or (ii) the other extreme case where the liquids are assumed dielectric (O'Konski and Thacher, 1953; Allan and Mason, 1962). However, following the rationale of basic electrokinetics, the liquid electrical conductivity (Saville, 1997) can no longer be considered a homogeneously distributed value in the liquid bulk when charge relaxation is compromised by hydrodynamic motion. Therefore, the customary assumption of a constant liquid conductivity and liquid bulk electric neutrality would be inconsistent if one aims to compare numerical results based on that assumption with scaling laws such as the one in de la Mora and Loscertales (1994) and de la Mora (2007).

Moreover, the electrokinetic phenomena appearing in many electrohydrodynamic problems in the microfluidics field are strongly related to the presence of the electric double layers (EDL) that appear on interfaces when they are brought into contact with electrolytes (Kirby, 2010; Zhao and Yang, 2012). The thickness of the EDL, λ_D, is in the nanometric scale and can be very different from the characteristic length L_o of the system being investigated. For example, in electroosmotic pumping the characteristic width of the impulsion channel is typically of the order of dozens of microns. In contrast, the EDLs present at the channel walls are nanometric. The ionic channels present in biological membranes are also of nanometric size (Schoch et al., 2008; Zheng et al., 2011). Consequently, very different numerical approaches have been developed depending on the particular electrokinetic system considered. In the case where the EDL can be assumed thin, $\lambda_D \ll L_o$, a detailed EDL resolution can be avoided by either using the Helmholtz–Smoluchowski slip velocity or by using techniques like matched asymptotic expansions (Squires and Bazant, 2004). At the other extreme, for $\lambda_D \geqslant L_o$, the electrokinetic systems are best described by considering all the individual atomic interactions, for example using molecular dynamics (MD) simulations (Eijkel and van den Berg, 2005). Between the aforementioned limits, the continuum approach in which ions are not treated as microscopic discrete entities but as continuous charged species densities (Zheng et al., 2011) is the proper choice.

Some of the ways used to manipulate droplets and bubbles in fluidic microsystems have their origin in electrokinetic phenomena like electrosmosis or electrophoresis (Stone et al., 2004). For instance, a dielectric fluid of negligible conductivity like an oil can be continuously pumped through a channel by electroosmotic means by adding a suitable layer on an electrolyte (Lee et al., 2006; Gao et al., 2005). Hence, investigations on the physics of electrokinetic effects on fluid–fluid interfaces such as the recent work of Pascall and Squires (2011) are of great interest. This work explains the physical reasons by which electrokinetics effects are enhanced at liquid/liquid interfaces. In this context, the work of Zholkovskij et al. (2002) sheds light on the deformation of suspended droplets under an imposed axial electric field. The model of Zholkovskij et al. keeps the ionic nature of the charge and provides an analytical solution for the deformation. Interestingly, they show how the classic expressions obtained by Allan and Mason (1962) for pure dielectric fluids and by Taylor (1966) using the leaky-dielectric model are limit cases of the more general electrokinetic model. In the same manner, the work of Zholkovskij et al. can be used as a very complete benchmark for testing numerical models of two-phase electrokinetic problems (Berry et al., 2013).

In the present paper, we will focus on the numerical treatment of two-phase problems involving physical phenomena of electrokinetic nature using a Volume-Of-Fluid (VOF) method. In particular, our contribution here aims at the development of an accurate, efficient tool to deal with these general electrokinetic problems where the characteristic times associated to either the electrokinetics or the hydrodynamics can be comparable, with the ultimate objective to tackle the existing conundrum in electrospray physics and the electrohydrodynamic emission of extremely small charged droplets and particles from Taylor cone–jets. To this end, we use as foundation the open source code Gerris (Popinet). Gerris, originally conceived as an incompressible Navier–Stokes equations solver