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A HYBRID MODEL FOR THE SIMULATION OF ELECTROOSMOTIC FLOWS IN MICROCHANNELS, INDUCED BY INTERNAL ELECTRODES

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ABSTRACT

In general, the modeling of electroosmotic flows can be approached in two fundamentally-different ways. (i) The thickness of electrical double layer (EDL) is ignored and the effect of the electrical forces within the EDL is imaged into a modified kinematic boundary condition, the so-called Helmholtz-Smoluchowski slip condition. This approach is numerically simple and inexpensive, but implies several restrictions. (ii) The EDL is fully resolved, using a first-principle approach based on differential conservation equations for mass, momentum, and charge. This approach is enormously elaborate and numerically expensive, but appears to be applicable for a much wider range of problems. As an example, the treatment of internal electrodes, adjacent to insulating walls at defined zeta potential, appears difficult with the simple approach (i), since any non-continuous potential distribution at the wall leads to a singularity of the electrical field strength.

To avoid these difficulties, we develop a hybrid model which, on the one hand, electrically resolves the EDL to reveal a perfectly-continuous potential distribution in the complete microchannel. On the other hand, the flow equations are solved in the fluid bulk only, not comprising the EDL. Hence, the effect of the EDL is still incorporated by means of modified kinematic boundary conditions. The advantage of this hybrid model is, firstly, to avoid artificial singularities of the electrical field strength, where regions of different surface charge meet. These singularities are clearly artificial, since they result from neglect-

ing the extend of the EDL. Secondly, the hybrid model, at each time step, needs to solve only once for the potential distribution, which makes it numerically inexpensive and simple. Hence, systematic parameter studies are within reach.

We apply the hybrid model to investigate the influence of internal electrodes onto the flow field, driven by electroosmosis in a modular rectangular microchannel. As internal electrodes can be positioned at lower distances (if compared to external electrodes), they can be operated at lower voltages and still ensure strong electrical field strength. Systematic studies on the effect of different electrode positions and voltages are presented, leading to optimized settings for specific tasks as pumping or mixing. Further, a comparison to first-principle simulations using the approach (ii) is presented for selected cases. This demonstrates that the hybrid model perfectly captures the dominant physics.

NOMENCLATURE

Symbol	Unit	Description
C	[1]	dimensionless concentration field
d_0	[m]	channel width
$\mathbf{E}$	[V/m]	electrical field vector
l_D	[m]	Debye length
M	[1]	mixing quality
p	[Pa]	pressure
P	[1]	dimensionless pressure
q	[C/m ³]	charge density
Re	[1]	Reynolds number
t	[s]	time
u_0	[m/s]	electroosmotic velocity
$\mathbf{w}$	[m/s]	velocity vector
$\mathbf{W}$	[1]	dimensionless velocity vector
ϵ_0	[C/(V m)]	permittivity in vacuum
ϵ_r	[1]	relative permittivity
μ	[Pa s]	dynamic viscosity
ϕ	[V]	electrical potential
ϕ_0	[V]	applied potential difference
Φ	[1]	dimensionless electrical potential
ρ	[kg/m ³]	density
τ	[1]	dimensionless time
ζ	[V]	Zeta potential

1 INTRODUCTION

For the miniaturization of chemical or biochemical analytics, all steps usually performed in a conventional laboratory have to be integrated on a single chip (μ TAS). Therefore, devices are needed, which allow e.g. pumping, mixing, and separation. Given typical low Reynolds numbers ($Re < 1$) in such devices, no turbulence develops and e.g. mixing appears non-trivial. Further, movable parts, as e.g. in mechanical pumps, should be avoided. One solution to perform multiple steps within such μ TAS devices is the implementation of electrical fields. If a solid wall is in contact with e.g. an electrolyte, an electrical double layer (EDL) occurs near the channel wall. For microchannels of small width, e.g. in the range of 10 – 200 μ m, forces within the electrical double layer (EDL) in the direct vicinity of the walls may have a significant effect. In contrast, for channels of larger width the influence of forces within the EDL is negligible, due to its small thickness of typically 100 nm. The application of electrical fields is responsible for electrokinetic effects, such as electroosmosis or electrophoresis, which generally allow to control fluid flow and mass transport in microchannels. By these means, e.g. pumping of fluid without mechanical parts, mixing at low Reynolds number, or separation of different species can be achieved in microchannels [1].

Internal electrodes may have several advantages if compared to external electrodes [2]. Firstly, as internal electrodes can be arranged at lower distances, lower voltages are needed to achieve the desired electrical field strength. Secondly, internal electrodes can be arranged in arrays, which allow by means of different voltage pattern to perform multiple task within the same device. Of course, there are likewise problems, linked to internal electrodes. For reactive systems, redox reactions can be expected and, therefore, electrode reactions have to be considered. Such reactions can be observed even at low voltages and they change the composition of the fluid [3]. Small changes of the fluid composition may lead to false results within the analytical steps. Moreover, reaction products may be gaseous and perhaps cannot be kept in dissolution. This would cause another non-acceptable situation.

Numerical simulations of electrokinetic effects in microchannels are often based on the so-called Debye-Hückel approximation [4], i.e. on a linearized Poisson equation for the electrical charge distribution near the wall. Hence, in essence the EDL is assumed to be of constant thickness and charge distribution [5, 6]. For the simulation of internal electrodes, in contrast, an exact description of the electrical double layer is necessary. This is due to the effect of the electrode reactions and due to the heterogeneity of the wall (isolated walls/electrodes). As shown by Wälter & Ehrhard [7], engaging in an extension of the work of Yoon [8] or Olesen [9], the electroosmotic flow around the isolated walls appears closely linked to the flow around the electrodes.

The focus of this article is the simulation of the behavior of a modular microchannel with internal electrodes, particularly its potential to induce mixing and fluid flow. For this purpose a hybrid model is developed, which allows to treat non-continuous wall charge distributions, which arise for internal electrodes. With this model an efficient numerical simulation of such electroosmotic flows is possible. The fluid motion around the electrodes and electrically-isolated wall segments are analyzed in detail and validated by a comparison with the results of the exact description of the electrical double layer by Wälter & Ehrhard [7] for the identical physical problem. Moreover, parameter optimization to achieve optimal pumping and/or mixing in such modular microchannels can be performed.

1.1 ELECTRICAL FIELD AND ELECTROKINETICS

The origin for the electrical double layer are wall charges, resulting from defects within the solid structure or wall-liquid interactions. As consequence, counterions accumulate near the channel wall or even get adsorbed and ions of equal charge are rejected [3]. The resulting EDL in the vicinity of the wall is electrically non-neutral. It can be divided into a fixed and a diffuse layer, according to the Gouy-Chapman-Stern model [10]. The fixed layer is composed of the Stern layer, where counterions are adsorbed due to electrostatic forces between wall and fluid, and

the shear layer of almost immobile counterions. Unlike the fixed layer, ions in the diffuse layer are free to move due to their large distance to the wall, responsible for weak electrostatic forces. A schematic drawing of the EDL (for a negatively-charged wall) is shown in figure 1. Conceptually, any flow and ion transport will appear outside the shear layer. An EDL can be likewise found at the contact of an electrically-conducting solid and an electrolyte. This EDL at electrodes is additionally influenced by electrochemistry and electrode reactions [3, 7]. These effects are, however, not detailed in this article. As has been shown be-

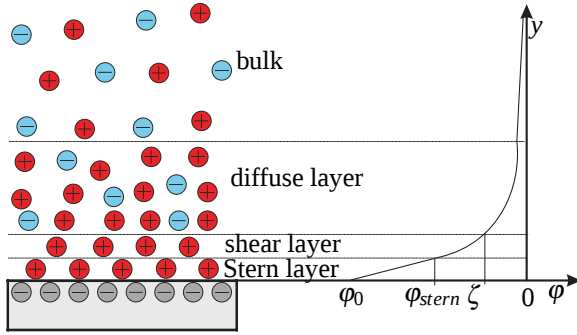


FIGURE 1. Schematic diagram of the electrical double layer and the potential distribution.

fore, e.g. by Barz [5], fluid flow and mass transport can be controlled by electrical fields using electroosmosis or electrophoresis. Both effects are based on the same principle, namely on a non-homogeneous ion distribution within the EDL. Given an applied electrical field, a directed streaming of the ions near the wall towards the electrode of opposite charge will result. These ions in excess, by viscous effects, drag the surrounding fluid until, for a small channel width, the whole fluid is in motion [1].

2 MATHEMATICAL FORMULATION

To describe the electrical potential in a dielectric medium, the Poisson equation

$$\nabla(\epsilon_r \nabla \phi) = -\frac{q}{\epsilon_0} \quad (1)$$

is used, where ϕ is the electrical potential, q the free-charge density, and ϵ_r and ϵ_0 are the relative permittivity and the permittivity in vacuum [1, 11]. The boundary conditions for the Poisson equation can be formulated according to the above given ideas. On the isolated walls, the Zeta potential defines the electrical potential; on the electrodes the applied potential defines the potential. The flow field, in general, is governed by the continuity equation, and

the time-dependent Navier-Stokes and species transport equations, i.e.

$$\nabla \cdot \mathbf{w} = 0, \quad (2)$$

$$\rho \left(\frac{\partial \mathbf{w}}{\partial t} + (\mathbf{w} \cdot \nabla) \mathbf{w} \right) = -\nabla p + \mu \Delta \mathbf{w} - q \nabla \phi, \quad (3)$$

$$\left(\frac{\partial c}{\partial t} + (\mathbf{w} \cdot \nabla) c \right) = D \Delta c, \quad (4)$$

augmented by an electrical force term $\mathbf{F}_{el} = -q \nabla \phi$. In equations (2-4) the variables are velocity $\mathbf{w}$, time t , pressure p , density ρ , dynamic viscosity μ , concentration c , and diffusivity D . In the above formulation (2-4), the kinematic boundary conditions, in general, would comprise no-slip conditions at all walls. The con-

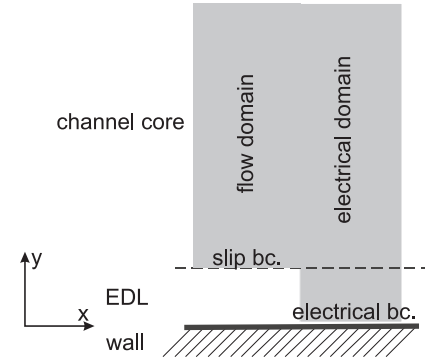


FIGURE 2. Electrical domain and flow domain for the simulations.

cept of our hybrid model involves the treatment of the electrical problem and of the flow problem on two different computational domains. In detail, (i) the solution to the electrical problem is sought on a periodic module with two internal electrodes, comprising the EDL. Hence, at the module walls (cf. figure 2), the zeta potential or the electrode potential are prescribed, while periodic boundary conditions are posed at the module ends. (ii) The solution to the flow problem is sought on a periodic module, not comprising the EDL (cf. figure 2). Hence, the boundary conditions are periodic at the module ends, and involve the Helmholtz-Smoluchowski slip condition at the wall-sided boundary of the core region. This boundary condition is given by

$$u = u_{slip} = \frac{\epsilon_0 \epsilon_r}{\mu} \zeta E_x, \quad (5)$$

$$v = 0. \quad (6)$$

Given the flow domain without the EDL, it is obvious that the entire domain is electrically neutral. Hence, it is consistent to

use the flow equations (3,2) without electrical forces, i.e.

$$\nabla \cdot \mathbf{w} = 0, \quad (7)$$

$$\rho \left(\frac{\partial \mathbf{w}}{\partial t} + (\mathbf{w} \cdot \nabla) \mathbf{w} \right) = -\nabla p + \mu \Delta \mathbf{w}. \quad (8)$$

2.1 DIMENSIONLESS EQUATIONS

To obtain more general results and to ensure a stable numerics, all simulations are based on dimensionless equations. All variables are scaled by characteristic quantities, which are in detail the channel width d_0 for all coordinates, the electroosmotic velocity u_0 for all velocities, the viscous pressure scale, a transport time scale, and the applied potential difference ϕ_0 between the electrodes. The dimensionless variables are noted in upper-case letters. For constant permittivities, we arrive at a dimensionless Poisson equation for the applied electrical field, namely at

$$\Delta \Phi = 0. \quad (9)$$

Equation (9) governs the applied part of the electrical potential in the entire domain, including bulk and EDL. The self-induced part of the electrical field within the EDL could be superimposed, though, if needed. The dimensionless continuity, Navier–Stokes, and species transport equations are

$$\nabla \cdot \mathbf{W} = 0, \quad (10)$$

$$Re \left(\frac{\partial \mathbf{W}}{\partial \tau} + (\mathbf{W} \cdot \nabla) \mathbf{W} \right) = -\nabla P + \Delta \mathbf{W}, \quad (11)$$

$$Re Sc \left(\frac{\partial C}{\partial \tau} + (\mathbf{W} \cdot \nabla) C \right) = \Delta C. \quad (12)$$

There remain two dimensionless groups, namely the Reynolds number Re and the Schmidt number Sc , defined by

$$Re = \frac{u_0 d_0 \rho}{\mu}, \quad (13)$$

$$Sc = \frac{\mu}{\rho D}. \quad (14)$$

The dimensionless boundary conditions for the electrical potential are $\Phi = \pm 0.5$ at the electrodes, and $\Phi = \zeta/\phi_0 = -0.1$ at the insulating walls. Kinematically, we have at all wall-sided boundaries

$$U = U_{slip} = -\frac{\epsilon_0 \epsilon_r \zeta \phi_0}{\mu u_0 d_0} \frac{\partial \Phi}{\partial X}, \quad (15)$$

$$V = 0. \quad (16)$$

2.2 NUMERICS

All simulations are done by the software *OpenFOAM* (Open Field Operation and Manipulation) [12]. The simulations are time-dependent and two-dimensional (plane) in nature. The spatial discretisation is based on the finite-volume method (FVM), we engage second-order square elements. Volume integration is e.g. done by the standard Gaussian scheme, temporal discretisation is done by the Euler implicit scheme. Figure 3 gives the standard grids for the simulation of both the flow problem (left) and the electrical problem (right). Refinement of both grids has been performed, to ensure that both electrical field and flow field from the simulations are not dependent on the grid.

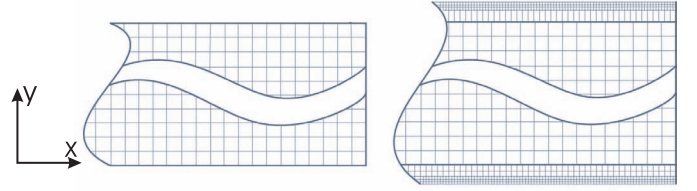


FIGURE 3. Grids for the simulation of (a) the flow problem and (b) the electrical problem.

3 RESULTS

The results section is divided into two parts. In the first part, we compare for a selected case the present results from the hybrid model with results of a detailed model by Walter & Ehrhard [7]. In the second part, we systematically demonstrate the influence of the electrode positions in the modular microchannel onto net fluid flow and mixing.

3.1 VALIDATION OF THE HYBRID MODEL

In this section, we validate the results from the hybrid model against results from the fully-resolved model by Walter & Ehrhard [7]. This detailed model spatially resolves the EDL and relies on the transport of all charged species. Figure 4 shows streamlines around the edge of the anode from both simulations. The anode is at the dimensionless potential $\Phi = +0.5$, the isolated wall is at $\Phi = -0.1$, due to a negative zeta potential. Figure 4a, from the detailed simulations, shows a large clockwise vortex, rising at the electrode center, set up by the electrical field between both electrodes. Additionally, a small counter-clockwise vortex is visible, induced by the different electrical double layers, adjacent to the insulating wall and adjacent to the anode. The comparison with figure 4b clarifies, that the flow results from the hybrid model resemble the detailed results with regard to the large clockwise vortex. However, the small vortex due to the

insulated wall is not captured. Given the hybrid model assumptions, particularly an identical slip condition for both electrical double layers, this appears not surprising.

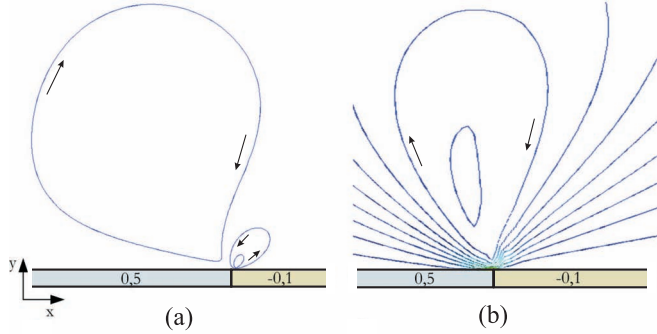


FIGURE 4. Results from (a) detailed model, (b) hybrid model. Both images show details at one electrode edge; the simulation geometries and parameters are identical.

3.2 VARIED ELECTRODE POSITION

In figure 5 and figure 6 we present simulation results for the flow field at varied electrode positions, obtained from the hybrid model. In figure 5 the electrodes are positioned face to face and from the streamlines four vortices can be inferred. The situation appears symmetric both with respect to the horizontal and vertical center line. The flow of all four vortices points outward from the electrode centers. A closer inspection of figure 5 reveals, however, that the top/bottom symmetry is not really given due to the non-symmetric electrical boundary conditions: at the electrodes we have $\Theta = \pm 0.5$, while at all isolated walls we have $\Theta = \zeta/\varphi_0 = -0.1$. Hence, for increasing applied potential difference φ_0 , a top/bottom symmetry is approached, and the effect of the zeta potential at isolated walls becomes negligible. In contrast, for small φ_0 , the top/bottom symmetry is broken.

In figure 6, the flow field is given for an electrode offset of 50 %. (An electrode offset of 100 % corresponds, via the periodicity of the module, again to a face to face arrangement.) We recognize again four vortices, which now are point-symmetric with regard to the center point of the microchannel module. Still the flow is outward from the electrode centers. Figure 6 indicates that wall-tangential flow contributions are present across the (periodic) ends of the microchannel module. Hence, such an electrode arrangement induces pumping through the module. This is in contrast to the situation in figure 5, where no net flow can be observed across the module ends.

The effect of both electrode arrangements on mixing can be inspected in figure 7 and figure 8. Here, we give simulation re-

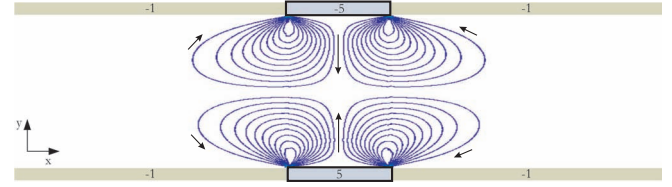


FIGURE 5. Flow field with no electrode offset.

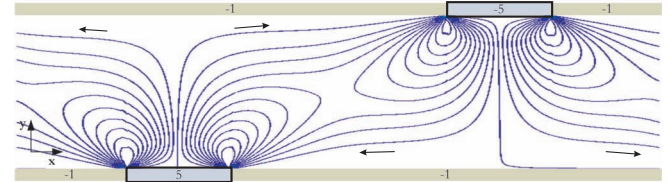


FIGURE 6. Flow field at 50 % electrode offset.

sults for the concentration fields at time $\tau = 0.025$. As initial condition ($\tau = 0$) we use a layered situation with $C = 1$ (red) in the lower half, and $C = 0$ (blue) in the upper half of the microchannel module. From figure 7 we recognize that for the face to face electrode position, mixing occurs in the upper and lower half separately, with no convective mixing across the horizontal center line, where $C = 0.5$ can be observed. For an electrode offset of 50 % in figure 8 though, mixing occurs convectively across the complete module height. This is caused by vortices, which extend across the entire module height (cf. figure 6).

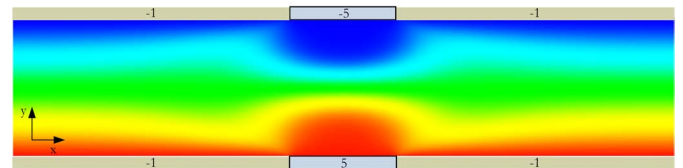


FIGURE 7. Concentration field with no electrode offset.

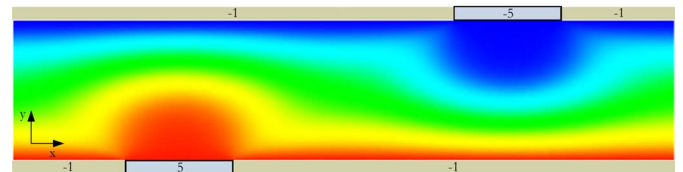


FIGURE 8. Concentration field at 50 % electrode offset.

We can systematically evaluate both the pumping and the mixing performance of the module, by analyzing the flow and concentration fields for various electrode offsets. In detail, to evaluate the pumping performance, we determine the horizontal mean velocity $\bar{U}_X$ within the module from integration along the module ends. The results of this procedure are summarized in figure 9. As expected, for an electrode offset of zero (or 100 %), we find $\bar{U}_X \simeq 0$. Hence, for a face to face arrangement of the electrodes no pumping occurs. For electrode offsets of 50 % or 75 %, though, considerable horizontal flow across the module ends is found. For these cases, horizontal mean velocities of up to $\bar{U}_X \simeq 0.35$ are computed. Consistently, integration along the left and right module ends, at very good accuracy, gives identical results.

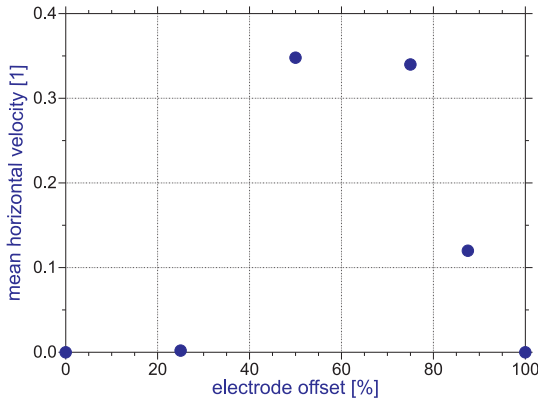


FIGURE 9. Horizontal mean velocity for varied electrode offset.

To evaluate the mixing performance, we determine the mixing quality M within the entire microchannel module. This mixing quality is defined by

$$M = 1 - \frac{\int |C - C_\infty| dV}{\int |C(\tau = 0) - C_\infty| dV} . \quad (17)$$

Here, C represents the actual concentration field (at $\tau = 0.025$), $C(\tau = 0)$ the initial (layered) concentration field, and $C_\infty = 0.5 = \text{constant}$ the perfectly-mixed concentration field. Hence, $M = 0$ means that the (initial) unmixed concentration field is present, while $M \rightarrow 1$ means that a perfectly-mixed situation has been achieved. Of course, $\tau = 0.025$ is an arbitrary choice, while $\tau \rightarrow \infty$ would in all cases result in $M \rightarrow 1$. Despite this arbitrary choice, a comparison of the effect of different electrode

positions appears valid without reservation. Figure 10 gives the corresponding results for the mixing quality for various electrode offsets. We recognize, that maxima of the mixing quality occur at electrode offsets of 25 % and 75 %, while at 0 %, 50 %, and 100 % lower mixing qualities are achieved. The data appear to be symmetric around the center of the horizontal axis at 50 % offset.

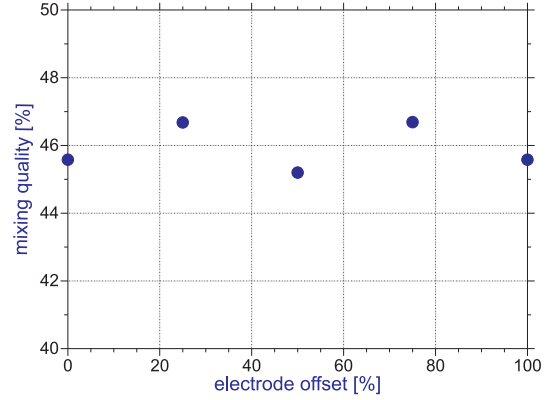


FIGURE 10. Mixing quality for varied electrode offset.

4 CONCLUSIONS

Following Walter & Ehrhard [7], the electroosmotic flow, caused by internal electrodes, is controlled by the applied voltage between the electrodes. Three regimes could be identified: (i) the applied voltage is very low, (ii) the applied voltage is higher, but no electrode reactions occur, (iii) the applied voltage is high enough for electrode reactions to occur. In regime (i), fluid flow is composed of four vortices around the electrode edges, which are influenced by the EDL at both the electrode and the isolated walls. In regime (ii), the electrical potential at the electrodes is high enough, so that effects due to the EDL at the isolated walls may be neglected. In regime (iii), electrode reactions have an appreciable effect onto the flow. While we ignore electrode reactions within the present article, and hence regime (iii), the present simulations with a hybrid model clearly confirm the existence of regimes (i) and (ii). This statement likewise involves a first validation of our hybrid model.

While Walter & Ehrhard [7] engage in a detailed model, which fully resolves all charged species concentration in the bulk and in the EDL, we infer a hybrid model, which is supposed to allow for systematic parameter studies of the problem. Our hy-

brid model captures the applied electrical field in the bulk and in the EDL, and, assuming constant properties of all electrical double layers, engages the Helmholtz–Smoluchowski slip condition to introduce electroosmosis into the flow problem. By this measure, a non–continuous potential distribution at walls, as is present e.g. at internal electrodes, can be treated in good approximation. Of course, the hybrid model is not able to capture the effects of electrode reactions and gas release, as is done in [7]. We have validated the hybrid model against the detailed model of Wälter & Ehrhard [7] and perform first systematic studies on the effect of the electrode positions onto pumping and mixing within a periodic microchannel module.

In the future, modular systems of internal electrodes and their potential for transport and mixing will be the focus. Systems of more than two electrodes, with changing electrical potentials in space and time, including the effect of the EDL at isolated walls, will be candidates of our investigations. Further, corresponding experimental investigations using the μ PIV and the μ LIF measuring techniques are already in preparation.

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