ABSTRACT

Electrically driven flows are widely used in various microfluidic applications where gradients in electrical properties of fluids such as conductivity are present. The variation in electrical conductivity influences these flows in two ways firstly, it creates a non-uniform electroosmotic flow and secondly, it can lead to generate electrokinetic instability. In the present work, we investigate the dynamics of such flows in two configurations: (i) where the electric field is applied parallel to the conductivity gradient known as collinear configuration, and (ii) when the electric field and conductivity gradients are orthogonal to each other. The collinear configuration is of particular interest in applications related to electrophoretic techniques such as field-amplified sample stacking. On the other hand, orthogonal configuration finds its applications where rapid mixing is desired in low Reynolds number flows.

At first, an experimental investigation is carried out to underline the effect of conductivity gradients in on-chip electrophoresis, a technique used for analyte separation. Experiments are performed in a cross-shaped microfluidic chip at various values of electric fields. Two fluorescent dyes rhodamine-B and rhodamine-6G are used as analytes to visualize the separation. The fluorescence intensity data of separated analytes is recorded as an electropherogram at a fixed axial location in the channel. We compare the electropherograms for the experiments performed with and without considering a conductivity gradient, which reveals that the conductivity gradient has an adverse effect on the separation performance. The experimental data shows that rapid dispersion occurs in the presence of conductivity gradients at high electric fields which lowers the peak intensity and increases the peak width. We analyse the data for various performance parameters such as peak separation, peak width, and peak resolution. The results show that although the conductivity gradient improves the signal intensity, non-uniform EOF and instabilities deteriorate the separation performance.

Next, we investigated the dynamics of field-amplified sample stacking that uses differential electrophoretic velocity of analyte ions in the high-conductivity background electrolyte zone and low conductivity sample zone for increasing the analyte concentration.

The stacking rate of analyte ions in FASS is limited by molecular diffusion and convective dispersion due to non-uniform electroosmotic flow (EOF). We present a theoretical scaling analysis of stacking dynamics in FASS and its validation with a large set of on-chip sample stacking experiments and numerical simulations. Through scaling analysis, two stacking regimes have been identified that are relevant for on-chip FASS, depending upon whether the broadening of the stacked peak is dominated by axial diffusion or convective dispersion. These two regimes are characterized by distinct length and time scales, based on which we obtain simplified nondimensional relations for the temporal growth of peak concentration and width in FASS. We first verify the theoretical scaling behavior in diffusion- and convectiondominated regimes using numerical simulations. Thereafter, we show that the experimental data of temporal growth of peak concentration and width at varying electric fields, conductivity gradients, and EOF exhibit the theoretically predicted scaling behavior. The scaling behavior described in this work provides insights into the effect of varying experimental parameters, such as electric field, conductivity gradient, electroosmotic mobility, and electrophoretic mobility of the analyte on the dynamics of on-chip FASS.

Further, we present an experimental and numerical investigation of electrokinetic instability (EKI) in a microchannel flow with streamwise conductivity gradients such as those in sample stacking during capillary electrophoresis. The base state consists of a plug of low-conductivity electrolyte solution between two high-conductivity zones in a microchannel, subjected to an external electric field applied parallel to the conductivity variation. The experiments of EKI were performed at varying values of the electric field and the flow was visualized using a passive fluorescent tracer. By sequentially varying the electric field, we show that the instability occurs beyond a threshold electric field. The experimental data was analyzed using proper orthogonal decomposition (POD) technique which provides, for the first time, an insight into instability modes and its dominant flow structures. The coherent structures help in identifying the onset of instability condition at which instability sets in the flow. In a frame of reference that moves with the mean

EOF velocity, temporal growth rate is governed by electroviscous time in high electric field limits. To explain the physical mechanism of the instability, we performed numerical simulations based on the Ohmic model for electrolyte solutions combined with the Navier-Stokes equations with additional electric body force term. Simulation results reveal that the nonuniform electroosmotic flow due to axially varying conductivity field causes a recirculatory flow within the low-conductivity zone. The role of electroviscous effects on EKI is demonstrated by analysing the results using appropriate velocity and time scales.

Lastly, we investigated EKI with orthogonal conductivity-gradient and electric field. In such configuration EKI can be characterized by transverse and longitudinal coherent structures which depend strongly on electric Rayleigh number (Ra_e). By sequentially increasing the electric field in experiments, we have obtained the coherent structures for the onset of instability, appearance of higher harmonics, period-doubling bifurcations, and chaotic flow regime. At the onset of instability, the electrokinetic flow is characterized by a transverse mode of instability. A secondary longitudinal mode appears in the nonlinear regime at higher electric field. Further increase in electric field leads to period-doubling bifurcations and eventually chaotic flow. The coherent structures of different flow regimes provide valuable insight into the dynamics, spatio-temporal scales, and the physical mechanism of electrokinetic instability.