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Electrokinetic flow in polyelectrolyte-modified nanopores

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We consider two fluid reservoirs separated by an electrically insulating membrane equipped with a single nanopore and polyelectrolyte (PE) chains are end-grafted to the nanopore wall of the membrane. A potential bias is externally imposed between the two fluid reservoirs. We investigated the electric-field induced electrokinetic flows in the absence and presence of charged DNA nanoparticles in the cathode fluid reservoir.

In the absence of DNA nanoparticles, the electric-field-induced ion transport and the resulting conductance in a PE-modified nanopore are theoretically studied using a continuum-based model, comprised of coupled Poisson-Nernst-Planck (PNP) equations for the ionic mass transport, and Stokes and Brinkman equations for the hydrodynamic fields in the exterior and interior of the PE layer, respectively. In contrast to a solid-state nanopore where ion concentration polarization (CP) occurs at the cathode side of the nanopore, two types of CP phenomena, occurring at either the cathode or anode side of the PE-modified nanopore, have been found. If the bulk ionic concentration is relatively low the counterions-rich CP occurs at the cathode side of the PE-modified nanopore. However, the counterions-rich CP occurs at the anode side of the nanopore when the bulk ionic concentration is sufficiently high and the imposed electric field is relatively weak. The induced CP phenomenon significantly affects the nanopore conductance. The counterions-rich CP phenomena in the PE-modified and solid-state nanopores are compared. The results clearly show that the CP phenomenon in the PE-modified nanopore is more significant than that in the solid-state nanopore regardless of the levels of site density of the molecular chains, the strength of the applied electric field, and the bulk ionic concentration. The results provided valuable information for utilizing polyelectrolyte brushes as a bridge to design the next-generation nanofluidic devices.

In the presence of DNA nanoparticles in the cathode fluid reservoir, DNA nanoparticles translocate electrophoretically from the cathode reservoir to the anode fluid reservoir and affect the ionic current flowing through the nanopore. Through the particles' effect on the ionic current, one hopes to detect the presence of the particles inside the pore as well as obtain information on the particles' characteristics such as DNA sequences in the next generation nanopore-based DNA sequencing technique. To increase the throughput in the nanopore-based DNA sequencing technique, one has to impose a high electric field across the nanopore, resulting in high-speed DNA translocation through the nanopore and consequently low read-out accuracy on the DNA sequences. Slowing down DNA translocation through the nanopore thus is one of the challenges in the nanopore-based DNA sequencing technique. DNA translocation through a PE-modified nanopore is theoretically investigated. In addition to the enhanced concentration polarization occurring near the entrance of the nanopore, the PE brushes increases electroosmotic flow inside the nanopore, the direction of which is opposite to the DNA electrophoretic motion. These effects simultaneously yield an increase in the DNA capture velocity at the nanopore mouth and a decrease in its translocation velocity within the nanopore. In general, if the salt concentration is high and the nanopore is short, regardless of the levels of the fixed charge density and the softness degree of the polyelectrolyte brushes, current blockade always occurs. This implies that the PE-modified nanopore can be applied to regulate the DNA translocation behavior without changing the ionic current signature. Considering recent advances in nanofabrication techniques and continuous growing of the widespread interests in chemically modified nanopores, the present work provides both necessary theoretical background and reasonable interpretations for the experimental observations on DNA translocation through a nanopore.

Keywords: Concentration Polarization, Nanopore, Polyelectrolyte Brush, Translocation