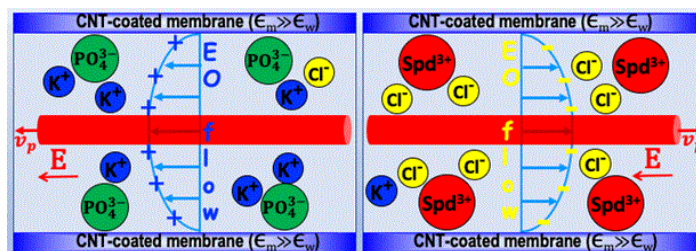


# Manipulating nanoconfined charge and macromolecule transport via electrostatic strong coupling interactions

**Sahin Buyukdagli**

*Bilkent University, Department Of Physics, Ankara, Turkey*

The accurate identification of the electrostatic correlation effects omnipresent in nanoconfined electrolytes is a critical task for the predictive design of nanotransport devices for energy conversion, water purification, and biosequencing purposes. The composite structure of the electrolyte mixtures involved in these processes requires the characterization of these systems via theoretical formalisms incorporating the distinct electrostatic coupling strengths associated with the constituent ion species with different valencies. I will present a unified theory of charge and polymer transport that includes on the same footing the weak- and strong-coupling interactions of mono- and multivalent ion species in confined electrolyte mixtures [1,2]. Within this formalism, I will first investigate the electrohydrodynamic mechanism driving the experimentally observed negative streaming currents in anionic nanochannels [3]. I will show that these like-charged currents triggered by multivalent cation addition into a monovalent KCl solution originate from the collective effect of the chloride attraction by the pore-adsorbed multivalent cations, and the hydrodynamic no-slip layer suppressing the contribution of these cations to the streaming current. The formalism indicates that the criterion for the emergence of these like-charge currents is the reversal of the electrostatic potential within the no-slip zone rather than the inversion of the pore surface charge. In the second part of the talk, the transport formalism will be applied to carbon-coated nanopores characterized by the giant membrane permittivity condition. In the corresponding electrostatic configuration, the theory predicts a novel molecular transport mechanism enabling the dielectric control of the polymer translocation dynamics in mixed electrolytes. Namely, in the giant permittivity regime of these engineered membranes, the emergence of attractive surface polarization forces leads to the surface adsorption of added multivalent ions with arbitrary sign. These interfacially adsorbed multivalent charges attract their monovalent counterions to the nanopore and exclude their monovalent coions from the pore into the ion reservoir. Under the effect of an externally applied voltage, the resulting monovalent ion separation induces an electroosmotic (EO) counterion flow. The shear force exerted by this EO current on the transported analyte is strong enough to cancel or invert the electrophoretic translocation velocity of the macromolecule, or equally to induce the mobility of neutral or non-uniformly charged proteins whose speed and transport direction can be tuned exclusively via the sign, charge, and amount of the added multivalent ions. Owing to the relevance of molecular transport speeds for the resolution of pore-based biosequencing techniques, this dielectric transport mechanism presents itself as a promising strategy for the extension of the mean dwell times in polyelectrolyte translocation, and also for the transport of non-uniformly charged proteins that cannot be controlled by an external voltage.



## References

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