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Micro/Nanoscale Electrochemistry: Advancing Detection and Molecular Dynamics Studies

Advances in nanofabrication techniques have enabled the construction of sophisticated nanoscale architectures, allowing electrochemical measurements in low dimensional nanostructures that can reveal unique electrochemical phenomena. In particular, nanopore electrode arrays (NEAs) serve as valuable platforms for investigating a variety of interesting nanoelectrochemical characteristics in confined volumes, typ. ~10-18 L. This small confined volume enables the exploration of chemical reaction dynamics involving a single molecule or just a few molecules. The first topic focuses on demonstrating biomimetic ion gating in block copolymer (BCP)-coated NEAs in response to external stimuli, such as pH and ionic charge state. The BCP membrane acts as a pH-gate, controlling ion transfer into the nanopores due to pH dependent structural transitions. Redox species are selectively transported through the BCP membrane into the NEAs, where they can then be sensitively detected through electrochemical signal amplification via redox cycling. Extending this concept, potential-induced wetting and dewetting behavior is characterized in BCP membranes on NEAs. Interestingly, mass transport across the hydrophobic BCP nanochannels is switched on by sufficiently negative potentials applied across the BCP membrane, resulting in the introduction and isolation of the electrolyte solution within the nanopores. Moreover, NEAs are employed in a bifunctional zero mode waveguide (ZMW) configuration that combines electrochemistry with spectroscopy for single enzyme molecule studies. The potential-dependent fluorescence dynamics are investigated in an electrochemical ZMW, where a single Au ring electrode embedded in each nanopore simultaneously controls the electrochemical potential and confines electromagnetic radiation to a zeptoliter-scale observation volume within the nanopores. The final topic addresses microscale CO2-sensitive ion-selective electrodes, combining scanning electrochemical microscopy to investigate the local activities of surface-bound enzymes.

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