Chemically functionalized graphene materials for selective ion separation and sensing

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Pristine graphene, known for its exceptional strength and ultrathin structure, serves as an ideal separation barrier that we explore for templating graphene liquid cells to contain biological materials for liquid phase electron microscopy. By introducing nitrogen into the graphene lattice, nitrogen-doped graphene fulfills dual functions: acting as a containment barrier for the electrolyte and as electrodes for the oxygen reduction reaction. These advanced devices allow real-time x-ray photoelectron spectroscopy (XPS) imaging under high vacuum conditions with the synchrotron, pushing the boundaries at which we can increase our fundamental understanding of surface catalysis. By functionalizing the graphene basal plane, either covalently with diazonium radical reactions or noncovalently by layer-by-layer deposition, we develop proton-selective membranes aimed at improving the performance of direct methanol fuel cells. Additionally, graphene's large surface area and strong interaction with lipids are being explored for precise detection of chemical and biological molecules, such as insulin, using quartz crystal microbalance with dissipation monitoring (QCM-D). Beyond the basal plane, graphene edges hold great promise for next-generation sensing and sequencing technologies. We are developing high-density graphene edge devices and subsequent functionalization methods with single-stranded DNA for use in graphene field-effect transistors (GFETs) to allow specific DNA detection. In parallel, we are exploring facile and reproducible graphene nanopore and graphene nanogap device fabrication by controlled dielectric breakdown and photolithography, respectively. A key challenge in achieving graphene-based sequencing is specific functionalization of the graphene edges. To deepen our understanding of graphene and 2D materials in general, our research also extends into organic chemistry. We are investigating the interactions between molecular tweezers and nanographenes, as well as bottom-up fabrication methods such as the Langmuir-Blodgett technique to create chemically defined nanometer films with functional groups like borazines. Overall, graphene and other 2D materials are becoming increasingly significant as research progresses: their excellent separation properties enable real-time imaging of biological and electrochemical processes; versatile functionalization techniques of the basal plane offer new materials and devices for improved energy storage and sensing applications; and edge functionalization holds promise for tunneling junctions and biomolecular sequencing.