

From polycyclic aromatic hydrocarbons to nanopore-nanogap devices

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Starting at the smallest scale, with polycyclic aromatic hydrocarbons, we investigate the conformational preference of propeller-shaped polycyclic aromatic hydrocarbons to design propellerene molecules as building blocks of millimeter to centimeter scale devices. We design and synthesize polyaromatic hydrocarbon based macromonomers capable of self-assembling into 2D materials, which we integrate in fuel cells, electrolyzers, electro-osmosis and desalination devices. Zooming out a little bit more, we use as-prepared membranes like graphene to tailor nanopores tiny and functional enough to – we hope – only allow the passage of a proton. We are currently looking into time-dependent diazonium solution treatment to obtain leak-tight graphene selective ion channels. At the device level, we use chemistry to tailor graphene devices. We designed graphene field-effect transistors that can electrically probe the molecular state of spin crossover crystals using chemo-electrical gating. As an electrode, we use graphene as a model carbon-based electrode and systematically introduce nitrogen and oxygen dopants, together with vacancy defects, to study ORR. The latter work not only indicates a non-negligible contribution of oxygen and especially oxygenated vacancy defects for the catalytic activity of nitrogen-doped graphene, but also provides important insights into the fundamental understanding of activity-structure correlations for tailoring the catalytic performance of carbon-based, most particularly graphene-based electrode materials. Importantly, graphene edges offer interesting chemical opportunities for sensing. The inability to systematically characterize covalently functionalized graphene edges however, could still potentially limit the use of graphene in nanogap constructs for single molecule detection and recognition. We develop now parallel routes for edge functionalization that will allow the in situ spectroscopic characterization of edge functionality. On the other side of the spectrum, our knowledge of graphene handling and graphene transfer allows now a high-yield graphene liquid cell fabrication, which will enable – we believe – the progression of the liquid phase electron microscopy field with 2D materials. Chemistry has a lot to bring to the research field of graphene and 2D materials in general, from the smallest molecular scale to the device level: graphene intrinsic's hydrophilicity in water, edge reactivity in devices, chemical routes for the alignment of tunneling junction in nanoporous nanofluidic devices for single molecule detection, graphene devices in water, on ice and on hydrogels, 2D functional membranes, to name a few.