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On-Water Surface Synthesis of Two-Dimensional Polymer Membranes toward Effective Ion Transport

Zhiyong Wang, Xinliang Feng

^aMax Planck Institute for Microstructure Physics, Halle (Saale), Germany ^bCenter for Advancing Electronics Dresden (cfaed) & Faculty of Chemistry and Food Chemistry, Technische Universität Dresden, Germany

wang.zhiyong@tu-dresden.de

Synthetic two-dimensional polymers (2DPs) are an emerging class of structurally-defined crystalline materials that comprise covalent networks with topologically planar repeat units. Yet, synthesizing 2DP single crystals via irreversible reactions remains challenging. Herein, utilizing the surfactant-monolayerassisted interfacial synthesis (SMAIS) method, few-layer, large-area, skeleton-charged 2DP (C2DP) single crystals were successfully synthesized through irreversible Katritzky reaction, under pH control. The resultant periodically ordered 2DPs comprise aromatic pyridinium cations and counter BF_4^- anions. The representative C2DP-Por crystals display a tunable thickness of 2-30 nm and a lateral size of up to 120 μm². Using imaging and diffraction methods, a highly uniform square-patterned structure with the in-plane lattice of a = b = 30.5 Å was resolved with near-atomic precision. Significantly, the C2DP-Por crystals with cationic polymer skeleton and columnar-like pore arrays offer a high chloride ion selectivity with a coefficient up to 0.9, thus ensuring the integration as the anion-selective membrane for the osmotic energy generation. In addition, as the graphite electrode skin, we demonstrate that C2DP enables to prevent the cation/solvent co-intercalation into the graphite electrode and suppress the consequent structure collapse, leading to enhanced durability of Li battery. Our studies reveal a route to synthesize 2DP single crystals using a kinetically controlled irreversible reaction and will propel the development of membrane-based energy-conversion and storage technologies.

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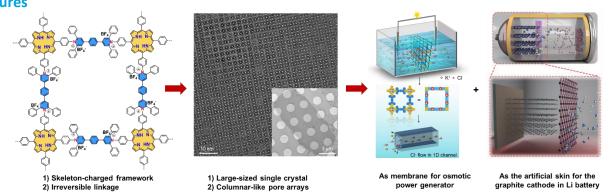


Figure 1: On-water surface synthesis of charged 2DP single crystal as the membrane for osmotic power generation, and electrode skin for Li battery.

Figures