## On-Water Surface Synthesis of Charged Two-Dimensional Polymers Enabling Effective Ion Transport

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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-monolayer-assisted 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 BF4<sup>-</sup> anions. The representative C2DP-Por crystals display a tunable thickness of 2-30 nm and a lateral size up to 120  $\mu$ m<sup>2</sup>. 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. 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 technologies.

## References

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## Figures

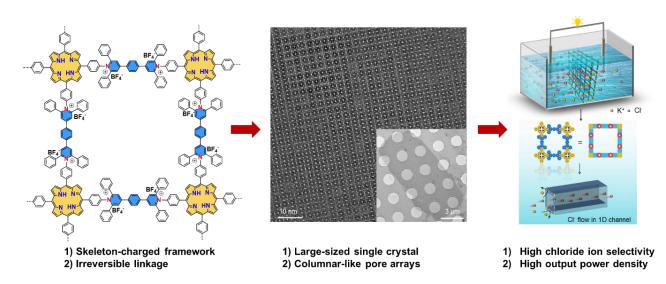


Figure 1: On-water surface synthesis of charged 2DP single crystal via irreversible reactions towards osmotic energy generation.