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In our work, we have employed interfacial chemistry toward the precision synthesis of 2D framework materials (2DFMs) with diverse structures and functions. For instance, we demonstrated the synthesis of 2D conjugated metal-organic framework (2D c-MOF) at the air-water or liquid-liquid interfaces. The 2D c-MOFs feature with stacked layers and possess unique electronic properties, such as full  $\pi$  delocalization, narrowed band gaps and high conductivity, which render 2D c-MOFs as advanced electronic materials. One representative iron-bis(dithiolene) 2D MOF exhibited as a p-type semiconductor with a band-like transport and high mobility of ~220 cm<sup>2</sup>/Vs.<sup>[1]</sup> Owing to their conductivity, the 2D c-MOFs have shown potential for transistors, photodetectors, sensing, magnetics, and energy storage and conversion.<sup>[2]</sup> In addition, we have also synthesized highly crystalline 2D polymers on the water surface. For instance, we have employed a surfactant-monolayerassisted interfacial synthesis (SMAIS) method to prepare 2D polymers, like 2D polyimides, 2D polyimines and boronate ester 2D polymers, which exhibit few-layers and micrometer-sized single-crystalline domains, which have been utilized as active layers for optoelectronics and memory devices.<sup>[3,4]</sup> In our latest work, we have developed charged 2D polymer single crystals through an irreversible Katritzky reaction under pH control, which could act as an anion-selective membrane for osmotic energy generation, offering a high chloride ion selectivity.<sup>[5]</sup>

[1] Nat. Mater. 17 (2018) 1027-1032.

- [2] Chem. Soc. Rev. 50 (2021) 2764-2793.
- [3] Angew. Chem. Int. Ed. 59 (2020) 8218-8224.
- [4] Angew. Chem. Int. Ed. 60 (2021) 13859-13864.
- [5] Nat. Synth. 1 (2022) 69-76.

Figures