Interfacial Synthesis of 2D Conjugated Polymers: the Rise of Organic 2D Materials

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The discovery of graphene one decade ago has triggered enormous interest in developing two-dimensional materials (2DMs). At present, various synthetic strategies have been devoted to produce 2DMs, such as top-down exfoliation and bottom-up chemical vapor deposition and solution synthesis.[1] In our work, we have employed the interfacial chemistry toward the controlled synthesis of organic 2D materials with varied structural features and diverse functions. For example, we demonstrated the synthesis of 2D conjugated metal-organic framework (2D *c*-MOF)—crystalline porous coordination polymer—at the air-water or liquid-liquid interfaces via metal-bis(dithiolene)/-bis(diimino)/-catecholate complexes. The resultant MOFs feature with large-area single layer or van der Waals stacked multi-layers and possess unique electronic properties, such as full delocalization of π-electrons, narrowed band gaps, largely improved conductivity and high charge mobility, which render 2D *c*-MOFs as advanced electroactive materials. One representative triphenylene-based iron-bis(dithiolene) MOF is a *p*-type semiconductor with a band-like transport and high charge mobility of ~220 cm^2/Vs.[2] Currently, the 2D *c*-MOFs have been applied in transistors, sensing, magnetics, and energy storage and conversion.[3-5] In addition, we have also synthesized conjugated 2D covalent polymers, such as polyimines, polyimides and polypyrazines, at the air-water or liquid-liquid interfaces. For example, we synthesized a single-layer polycrystalline 2D polyimine at the air-water interface in a Langmuir-Blodget trough. The resultant 2D polyimine displayed an outstanding Young's modulus (267±30 GPa) comparable to that of graphene and multi-functions in thin film field effect transistor (FET) and electrocatalytic water splitting.[6] In our latest work, by employing surfactant monolayer as a soft-template at the air-water interface, we were able to synthesize imide-based 2D polymers with highly ordered square units up to remarkable ~4 μm^2 for a single crystalline domain. This preliminary result clearly highlights the feasibility to synthesize highly crystalline 2D polymers via soft-template-assisted interfacial synthesis strategy. In short, we expect to develop interfacial chemistry toward the synthetic 2DMs and achieve delineation of reliable chemistry-structure-property relationships and superior physical and chemical performances.

References

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