

Organic 2D Materials: Grand Challenges and Opportunities

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In the past decade, as inspired by the discovery of graphene, two-dimensional (2D) materials which possess a periodic network structure and with a topographical thickness of atomic/molecular level, have emerged as the new paradigm of materials with enormous potentials, ranging from electronics and optoelectronics to energy technology, membrane, sensing and biomedical applications. Various fabrication strategies have been developed to attain high quality 2D materials. Among of them, mechanical exfoliation remains the most popular protocol to isolate single-layer high quality 2D materials for fundamental physical studies. In contrast to the tremendous exploration of graphene and inorganic 2D crystals such as metal dichalcogenides, boron nitride, black phosphorus, metal oxides and nitrides, the study on organic 2D crystal systems including the bottom-up organic synthesis of graphene, 2D metal-organic frameworks, 2D polymers/supramolecular polymers as well as supramolecular approach to 2D organic nanostructures remains under development.

In this lecture, we will present our recent efforts on the synthetic approaches towards novel 2D organic materials with structural control at the atomic/molecular-level or at the meso-scale. First, we will present the solution synthesis of 2D sp²-carbon based conjugated polymer frameworks as the new generation covalent-organic frameworks. Second, we will introduce the latest development on the synthetic 2D conjugated polymers including 2D Schiff-base type covalent

polymers and 2D metal-dithienene/diamine coordination supramolecular polymers at the air-water or liquid-liquid interfaces. The resulting 2D conjugated polymers exhibit single-layer feature, good local structural ordering and with a size of cm². The functional exploration of such 2D single-layer conjugated polymers for the electrical and mechanical properties, as well as serving as efficient electrocatalytic water splitting catalysts will be demonstrated. Third, we will introduce the self-assembly of a host-guest enhanced donor-acceptor interaction, consisting of a tris(methoxynaphthyl)-substituted truxene spacer, and a naphthalene diimide substituted with N-methyl viologenyl moieties as donor and acceptor monomers, respectively, in combination with cucurbit[8]uril as host monomer toward monolayers of an unprecedented 2D supramolecular polymers at liquid-liquid interface. Finally, we will present the supramolecular approaches to synergetic control the multi-component assembly, which results into 2D conducting polymers, such as polypyrrole and polyaniline nanosheets featuring 2D structures and with adjustable mesopores with/without on various functional free-standing surfaces. The unique structure with adjustable pore sizes (5–20 nm) and thickness (35–45 nm), enlarged specific surface area as well as high electrical conductivity make 2D conducting polymers promising for a number of applications. The future perspective and outlook regarding the goal towards highly crystalline organic 2D materials will be provided.