Electrically Conductive Two-Dimensional Metal-Organic Frameworks

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The traditional 3D metal-organic frameworks (MOFs) are considered as insulators due to the insulating organic ligands, which are rarely applied in organic electronics. However, the recently rising π-conjugated two-dimensional metal-organic frameworks (2D MOFs) have emerged as highly appealing electronic materials for energy storage and conversion, and field-effect transistors, owing to their high electrical conductivity and tuned band gaps. In our work, we have demonstrated the “bottom-up” synthesis of a series of π-conjugated 2D MOFs based on metal-bis(dithiolene)/bis(diimino)/catecholate complexes via interfacial synthesis and one-pot solvothermal synthesis, which exhibit high DC conductivity. For instance, the achieved hexathiatriphenylene-fused iron-bis(dithiolene) 2D MOF film (Fe₂(HTT)_3) by interfacial synthesis firstly shows n-type semiconducting behavior with small band gap of 0.6 eV and electron mobility of ~2 cm²/Vs at 298 K via Hall effect measurements. Moreover, for the first time, we demonstrate the room temperature band-like transport in such 2D MOFs by time-resolved terahertz pulse spectroscopy. A Drude model is applied to perfectly fit the alternating current (AC) transport and presents that the charge mobility could reach as high as ~150 cm²/Vs. Given the promising conductivity as well as their intrinsic porous structures, we explored the functions of these π-conjugated 2D MOFs in Li-ion batteries, micro-supercapacitors, electrocatalysis, chemiresistive sensing and spintronics.

References


Figures

Figure 1: (a) Structural schematics of Fe₂(HTT)_3 2D MOF. (b) Room temperature photoconductivity of Fe₂(HTT)_3 measured by THz spectroscopy.