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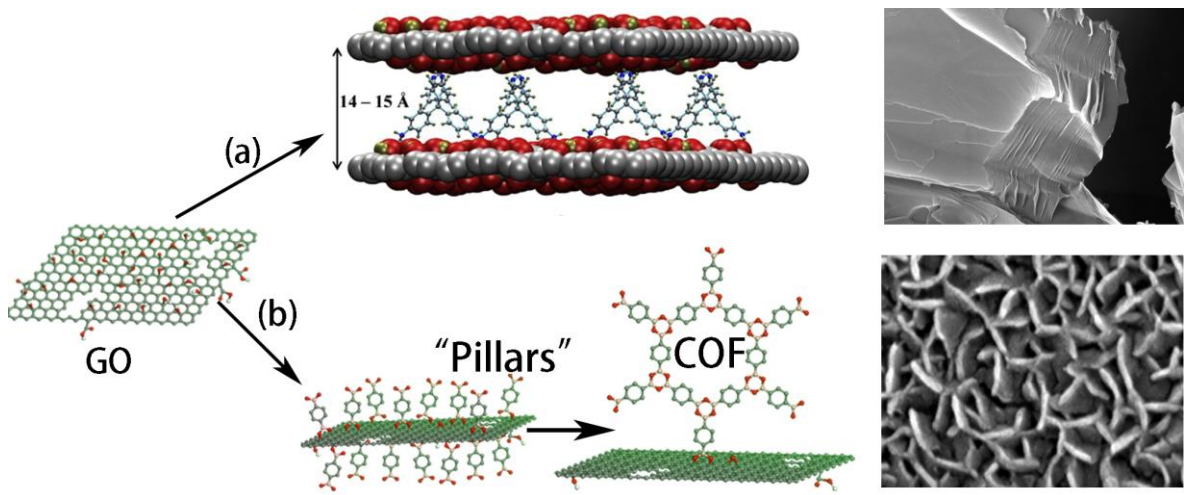
## Molecular pillar approach to construct functionalized graphene for energy storage

The rich and numerous oxygen-containing functional groups decorated on graphene oxide (GO) provide a possibility to chemically modify GO for various applications. In my talk, I will first introduce our research progress in tuning the interlayer distance of GO in the precision of angstrom by swelling in different solvents and using intercalation of differently sized organic molecular pillars, with the focus on using rigid molecule (Tetrakis(4-aminophenyl)methane (TKAm)) to make inter-linking of GO planes (Figure 1a).[1] The 3D shape of this rigid molecule could be of advantage for stability of pillared structures. The synthesized GO pillared with TKAm shows an expansion of inter-layer distance of GO from  $\sim 7.5\text{\AA}$  to  $13\text{-}14\text{\AA}$ . The intercalated TKAm as molecular pillars expand the GO structure resulting in high surface area up to  $660\text{ m}^2\text{ g}^{-1}$ . Besides 3D molecules, a simple molecule 4-benzene diboronic acid (DBA) as an idealized linker was reported to be efficient for linking GO.[2] However, our previous studies have demonstrated that the DBA molecules are likely to attach only on one side of inter-layer. Instead of using it to prepare graphene pillared structure, our alternative idea is to develop a molecular pillar approach to construct 2D-2D hybrid materials by using DBA pillars on GO. I will introduce how we utilize the chemically attached DBA as molecular pillars to directing grow ultrathin covalent organic frameworks (COF) nanosheets (v-COF-GO) (Figure 1b).[3] The hybrid material shows forest of COF-1 nanosheets with thickness of  $\sim 3$  to  $15\text{ nm}$  vertically grafting on the surface of GO via DBA linker. To emphasize the important role of DBA pillars on vertical growth of COF-1, the same reaction in absence of molecular pillars was performed and resulted in uncontrollable growth of thick COF-1 platelets parallel to the surface of GO. The v-COF-GO was converted into conductive carbon material with ultrathin porous carbon nanosheets grafted to graphene in edge-on orientation. It was demonstrated as a high-performance electrode material for supercapacitors. Controlling nucleation of 2D COF sheets and their growth direction by using molecular pillars covalently attached to graphene as structure-directing agent provides very promising route for preparation of new family hybrid materials.

### References

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- [2] J. W. Burrell, S. Gadipelli, J. Ford, et al., *Angew. Chem. Int. Ed.* 49 (2010) 8902-8904.
- [3] J. H. Sun, A. Klechikov, A. V. Talyzin, et al., *Angew. Chem. Int. Ed.* 57 (2018) 1034-1038.

### Figures



**Figure 1:** (a) Intercalation of rigid 3D molecules into interlayers of graphene for the synthesis of pillared graphene. (b) Growth of vertical COF-1 nanosheets using DBA as molecular nucleation sites grafted on GO.