Structural elucidation of shape-assisted self-assembled nanosheets from π-saddles

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Questioning how molecular topography can govern supramolecular ordering is a relatively unexplored avenue of thought and one which poses great synthetic challenge that could lead towards new functional materials. “Carpyridines” (Fig. 1a) — saddle-shaped macrocycles fused from carbazoles and pyridines — are an underdeveloped example of a simple system bearing negative Gaussian curvature that could hold such promise.[1]

Derivatisation of these non-planar systems through peripheral functionalisation of the aromatic core has yielded supramolecular assemblies (Fig. 1b, c) in the form of 2D sheets on surface with the thickness of a single molecule and fantastic edge definition.[1] Varying the length of alkyl chain in the macrocycle has then resulted in different types of nanosheet,[2] which hints at the subtle interplay between weak forces alongside the assisting role of shape. Using a combination of diffraction techniques, including micro-electron diffraction, the composition of these nanosheets can be elucidated to examine what effect shape has upon supramolecular ordering within the assembly.[2]

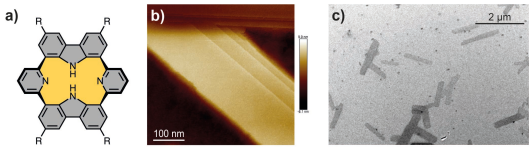


Fig. 1. a) Structure of Carpyridine (2H-Car-R) functionalised with R groups, and when R = *n*-C_6H_{13}, assemblies were visualized under b) AFM and c) TEM as 2D sheets.

[1] Woods, J. F.; Gallego, L.; Pfister, P.; Maaloum, M.; Vargas Jentzsch, A.; Rickhaus, M. *Nat. Comm.*, **2022**, 13, 3681.
[2] Woods, J. F.; Gallego, L.; Maisch, A.; Renggli, D.; Cuocci, C.; Blacque, O.; Steinfeld, G.; Kaech, A.; Spingler, B.; Vargas Jentzsch, A.; Rickhaus, M. *Chemrxiv*, **2023**, DOI: 10.26434/chemrxiv-2023-whjkw