chem2Dmat
September 03-06, 2019 ● Dresden, Germany
European Conference on Chemistry of Two-Dimensional Materials

Enriching and quantifying porous single layer 2D polymers by exfoliation of chemically modified van der Waals crystals

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Porous 2D polymer sheets[1] with six positively charged pyrylium groups at each pore edge in a stacked single crystal can be transformed into sheets of a 2D polymer with six neutral pyridine units per pore by exposure to gaseous ammonia. This reaction converts approximately 1'400'000 pyrylium ions per sheet µm^2 into the corresponding pyridines and is amongst the most complex post-polymerizations with molecular definition. Liquid exfoliation based on ultrasonic treatment of the product in the aqueous surfactant sodium cholate followed by an ultracentrifugation cascade[2] provides mg fractions with up to 29% of monolayers of this novel 2D polymer with an average length of ~130 nm. By the application of secondary cascades, this content can be increased further to 33% with a length of ~140 nm. This the first time the amount of monolayers of an exfoliated 2D material has been quantified and shown to be equally efficient as graphite exfoliation. The solution processability allows to cast thin films, making the material potentially accessible as separation membrane. We expect that recent advances in exfoliation of graphite or inorganic crystals (e.g. scale-up, printing etc.) can be directly applied not only to this 2D polymer, but also to other organic sheet stacks such as covalent organic frameworks.

References

[1] Servalli, M.; Öttinger, H. C.; Schlüter, A. D., Physics Today 71, (2018) 41-47.
[2] Backes, C.; Szydłowska, B. M.; Harvey, A.; Yuan, S.; Vega-Mayoral, V.; Davies, B. R.; Zhao, P.-l.; Hanlon, D.; Santos, E. J. G.; Katsnelson, M. I.; Blau, W. J.; Gadermaier, C.; Coleman, J. N., ACS Nano, 10 (2016), 1589-1601.

Figures

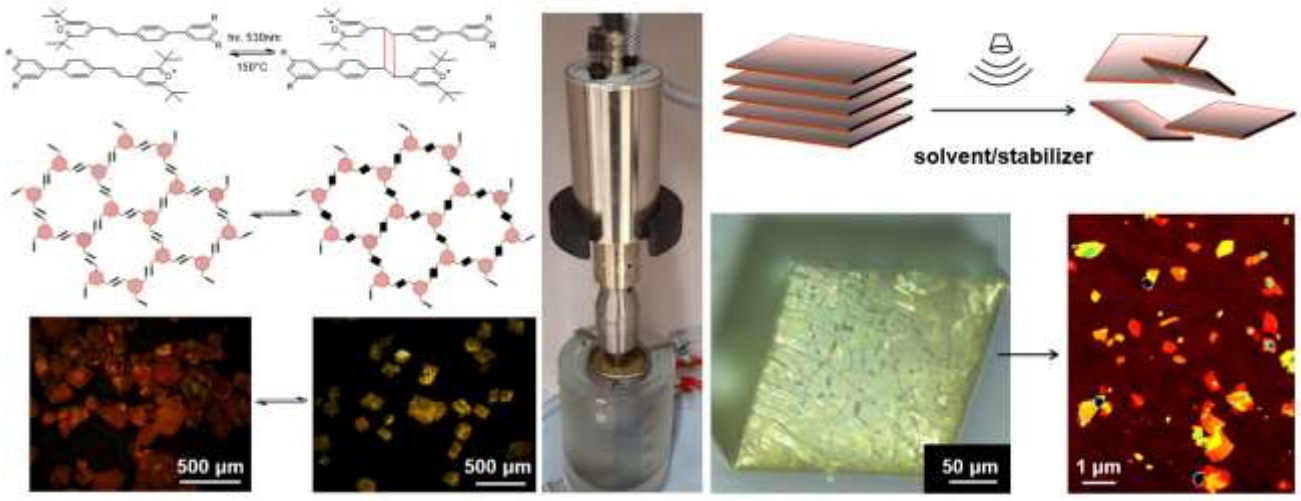

Figure 1: Simplified illustration of the synthesis and liquid-phase exfoliation of the porous 2D-Polymer