

Graphene oxide in 2D and 3D: structures, defects and applications.

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Abstract

Graphene oxide (GO) is a material studied for over 150 years and still providing many intriguing questions. In fact, GO is not one material but a family of materials with properties strongly depending on methods of synthesis. GO synthesized using Hummers and Brodie methods demonstrate major differences in swelling, thermal exfoliation, mechanical strength of individual flakes, sorption of polar solvents and heavy metals. Dispersing GO and re-depositing individual sheets into multilayered 3D structures (e.g. membranes) provide new set of materials distinctly different from precursor graphite oxides. Moreover, the defects (holes and vacancies) are an essential part of the GO structure affecting e.g. permeation of GO membranes and providing additional sorption sites for heavy metals. High abundance of hole defects in Hummers GO was demonstrated by many methods and can be compared semi-quantitatively using analysis of XPS data. In other hand, Brodie graphene oxide is preferable in applications, which require less defects. The difference in defect state of Brodie GO and Hummers GO was recently demonstrated using direct AFM imaging of bi-layered flakes exposed to water and ethanol vapors.¹ Recent studies demonstrated also that an increased number of defects and carboxylic groups in GO is correlated with the improvement of its sorption capacity for several radionuclides.² This trend led us to the synthesis of extremely defect-rich GO (dGO), which demonstrated up to a 15-fold increase in sorption capacity towards U(VI) compared to standard GO.³ The material was also demonstrated as promising for the sorption of organic pollutants.⁴ However, both GO and dGO need to be dispersed in water in order to make their high surface area accessible for sorption. Therefore, we designed an alternative approach to prepare carbon sorbents with similar to GO oxidation state but rigid 3D structure using high surface area porous carbon materials as precursors. Porous carbons demonstrate very high surface area (up to ~3000 m²/g for "activated graphene") which is available for oxidation directly without the need for lattice expansion. The surface oxidation methods are sufficient to convert porous carbons into GO-like 3D architectures. Surface oxidation of "activated graphene" is demonstrated to result in a materials with oxidation degree similar to that of graphene oxide while preserving relatively high BET surface area. "Super-oxidized activated graphene" showed extremely high sorption capacity for U(VI) removal. Notably, the bulk "3D GO" can be used as a sorbent directly without dispersing, the step required for GO to make its surface area accessible for removal of pollutants.⁵

References

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