The art of synthesizing pure hydroxylated graphene

Fabian Grote, Siegfried Eigler

Freie Universität Berlin
Institute for Chemistry and Biochemistry
Takustraße 3, Berlin, Germany

fabian.grote@stud-mail.uni-wuerzburg.de

Wet-chemically synthesized hydroxylated graphene acts as a precursor for high-quality graphene. Graphene sheets with defect densities as low as 0.02% on average have been obtained (Figure 1).[1]
Thereby, hydroxylated graphene is the key intermediate in the production process. It is characterized by a low degree of functionalization (4-6 %) with hydroxyl-groups (hydroxyl$_{4\%}$-G$_1$). Unlike graphene oxide, hydroxyl$_{4\%}$-G$_1$ is not dispersible in water but only in water/methanol mixtures, preferably in equal volumes. This behaviour of dispersibility is attributed to the low degree of functionalization. Achieving the dispersion of monolayers requires highly purified solvents. Chemical reduction yields graphene, however, impurities are introduced. Undesired contaminants are avoided by thermal processing; however, the carbon framework is commonly ruptured in the process due to CO_2 formation. We report that thermal processing of hydroxyl$_{4\%}$-G$_1$ leads to larger hole defects in the lattice, respectively hole formation, next to intact graphene areas and statistically distributed point defects. The defect concentration in the product depends on the number of defects already present in the hydroxylated graphene. We could quantify that high-quality hydroxylated graphene can be thermally processed and yield graphene with defect densities as low as 0.8%, as a consequence of the disproportionation reaction, on average. The quantification was confirmed by Raman spectroscopy and transmission electron microscopy at atomic resolution (HRTEM, Figure 2, in collaboration with Felix Börrnert and Ute Kaiser, Ulm).[1][2]

References

[1] Grote, F.; Gruber, C.; Borrnert, F.; Kaiser, U.; Eigler, S., *Angew. Chem. Int. Ed. Engl.* 2017, *56* (31), 9222-9225.
[2] Seiler, S.; Halbig, C. E.; Grote, F.; Rietsch, P.; Borrnert, F.; Kaiser, U.; Meyer, B.; Eigler, S., *Nat Commun* 2018, *9* (1), 836.

Figures

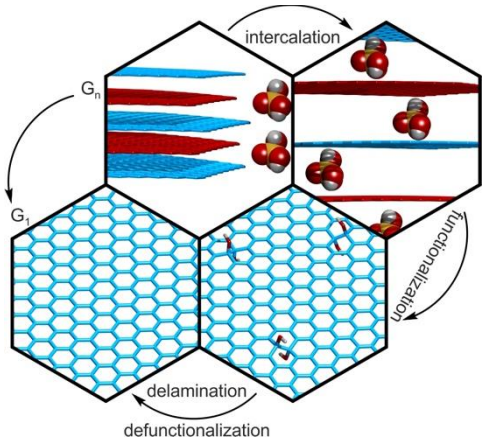

Figure 1: Graphene formation by oxidative delamination of graphite via hydroxylated graphene [2]

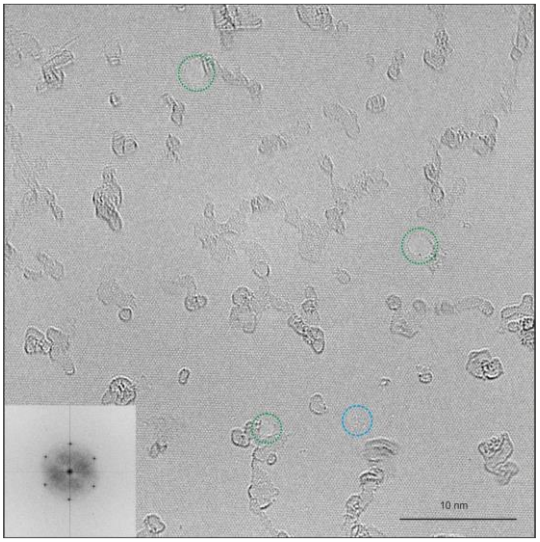

Figure 2: HRTEM image showing the intact carbon lattice of hydroxylated graphene with amorphous carbon impurities and few lattice defects [2]