

New Insights into Graphene Oxide Chemistry

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Abstract (Arial 10)

Graphene oxide is regarded as a major precursor for graphene-based materials. The development of graphene oxide based derivatives with new functionalities requires a thorough understanding of its chemical reactivity, especially for canonical synthetic methods such as the Diels–Alder cycloaddition. For the first part of the talk, we will show that the Diels–Alder reaction has been successfully extended with graphene oxide as a source of dienes by using maleic anhydride as a dienophile, thereby outlining the presence of the cis diene present in the graphene oxide framework. We found that this reaction could provide fundamental information for understanding the exact structure and chemical nature of graphene oxide. On the basis of high-resolution ^{13}C -SS NMR spectra, we will show evidence for the formation of new sp^3 carbon centers covalently bonded to graphene oxide following hydrolysis of the reaction product. We used DFT calculations to show that the presence of a cis dihydroxyl and C vacancy on the surface of graphene oxide are promoting the reaction with significant negative reaction enthalpies [1]. For the second part of the talk, a gram-scale synthesis of graphene oxide-based metal oxide nanocomposites with tunable fraction of reduced carbon will be discussed. Graphene oxide-based metal oxide nanoparticles hybrid materials with tunable fraction of reduced carbon were synthesized via a simple one-step reaction in aqueous solution at room temperature using water soluble metal halides such as stannous chloride, ferrous chloride, zinc chloride, aluminum chloride and magnesium chloride as a cheap, abundant, non-toxic and green source of reductants. The obtained graphene oxide-based materials have a more important fraction of reduced carbon when metallic ions with higher standard reduction potential such as Sn^{2+} ($E^0 = -0.14\text{ V}$) and Fe^{2+} ($E^0 = -0.44\text{ V}$) were used for the reduction. According to our observations, spontaneous formation of metal oxide NPs occurs on the surface of GO without damaging the integrity of its framework. NMR integration showed that GO is only slightly reduced during the transformation and the method allowed GO-MO composites preparation with a tunable fraction of reduced carbon ranging from 26-75%.

References

- [1] P. Brisebois, C. Kuss, Steen B. Schougaard, R. Izquierdo and M. Sijj, *Chemistry A-European Journal*, **22**, (2016), 5849–5852. DOI: 10.1002/chem.201504984.