

Advances in Graphene Preparation through Chemical Routes Studied by XPS, XRD, STEM and Electrical Transport

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Abstract. Graphene oxide (GO) is a material made of atomically thin graphitic sheets prepared by the oxidation of graphite. It is widely considered as a precursor for the large-scale production of graphene upon reduction. This is why GO is a subject of research for many potential applications including supercapacitors, solar cells, memory devices, bio-inspired systems and importantly conductive coatings. There are several chemical routes to GO, all based on using strongly oxidizing agents. One of the properties sensitively reflecting the structural disorder in graphene is charge transport. As a consequence of covalent functionalization, charge localization eliminates conductance in GO. Removing the functional groups by chemical or thermal methods to some extent restores charge delocalization and restores the conductance in the reduced GO lattice.

Here we present experiments on graphite oxidation by a well-established Hummers' method¹ spanning a time scale from minutes to several weeks. We trace a gradual change of the material properties and characterize the samples along the oxidation process. Our aim was to identify the stage when the conversion of graphite to GO is completed. To study the time evolution of structural and chemical properties of oxidized graphite, samples at different temporal stages of oxidation were selected and characterized through a number of techniques: X-ray photoelectron spectroscopy for the content and bonding of oxygen, X-ray diffraction for the level of intercalation, Raman spectroscopy for detection of structural changes, electrical resistivity measurements for probing charge localization on the macroscopic scale, and scanning transmission electron microscopy for the atomic structure of the graphene oxide flakes. At the final time, the interlayer distance expanded to more than twice the value of graphite and the electrical resistivity increased by 7 orders of magnitude².

In contrast to the observations on the product of the Hummer's methods, the newly developed an oxidation process of graphite leads only to very minor structural degradation of graphene structure. We will show that the significant improvement of the chemical and physical properties offers a very promising alternative to a waste majority of graphene on the market.

References

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