Revisiting Graphene Oxide Structure via Spatially-Resolved Electron Energy Loss Spectroscopy

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Graphite Oxide is produced by chemical oxidation of graphite and liquid exfoliation yields Graphene Oxide (GO) monolayers. The oxygen content can be lowered to obtain Reduced Graphene Oxide (RGO). Among numerous applications, GO has lately attracted renewed interest as a precursor for low-cost large-scale production of graphene. However, about 150 years since its discovery, GO atomic structure is still greatly debated. Spectroscopic studies have evaluated the overall atomic oxygen content in GO as 20%-35%. Nevertheless GO non-stoichiometric nature requires local analytic techniques: in this work we investigate GO and RGO by core Electron Energy Loss Spectroscopy (EELS) in a STEM microscope. A new experimental set up combining a liquid nitrogen system at the sample stage and a low-noise liquid nitrogen cooled CCD camera has allowed us to overcome the extreme sensitivity of these materials to illumination. We have determined a maximal electron dose of order 10^3 $e^-Å^{-2}$ and a 3 nm spatial resolution. Chemical maps show that the oxidation level varies spatially in the range of 15-50% in GO and 5-20% in RGO. In GO, typical Energy-Loss Near-Edge Structures (ELNES) profiles at carbon K-edge appear associated to low and high oxidation levels, while RGO presents intense graphitic peaks. We suggest that GO 50% oxygen-rich regions are almost fully functionalized by hydroxyls and we interpret ELNES peaks on this basis.