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Highly selective covalent organic functionalization of epitaxial graphene: a platform nano-bio-hybrid composites

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Graphene functionalization is an important step for the development of graphene-based materials with tailored electronic properties. However, its high chemical inertness makes difficult a selective covalent functionalization, and most of the works performed up to the date report electrostatic molecular adsorption or unruly functionalization. We show a new mechanism for promoting highly specific covalent bonding of any amino-terminated molecule and a description of the operating processes. We show, by different experimental techniques and theoretical methods, that the excess of charge at carbon dangling-bonds formed on single-atomic vacancies at the graphene surface induces enhanced reactivity towards a selective oxidation of the amino group and subsequent integration of the N within the graphene network. Functionalized surfaces retain the electronic properties of pristine graphene as unaltered.

Thus, we use this strategy to link to graphene a molecule with a robust thiol-terminated moiety derived from a p-aminothiophenol (p-ATP) molecules. We use this facile strategy to covalently link gold nanoparticles, which remain firmly anchored to the surface after many washing and annealing cycles. In parallel, we also couple in vitro a thiol-modified ssDNA aptamer to the p-ATP-functionalized graphene surface, and successfully obtain atomic force microscopy images both in air and in a liquid environment, and demonstrate that the anchored aptamer retains the functionality required to recognize its target protein, PCBP-2. This work opens the door to the integration of high-quality graphene layers in technological platforms for plasmonics, biosensing or advanced field-effect transistor devices.
Ref. Bueno et al. Nat. Comm. 2017; 8: 15306.

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