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Gas-Phase Nondestructive and Environmentally Friendly Covalent Functionalization of Graphene Oxide Paper with Amines

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Abstract : Direct covalent functionalization of prefabricated free-standing graphene oxide paper (GOP) is considered as the only approach suitable for systematic tuning of thermal, mechanical and electronic characteristics of this important class of carbon nanomaterials. At the same time, the traditional liquid-phase functionalization protocols can compromise physical integrity of the paper-like material up to its total disintegration. To avoid such undesirable effects, we explored the possibility of employing an alternative, solvent-free strategy for facile and nondestructive functionalization of GOP with two representative aliphatic amines, 1-octadecylamine (ODA) and 1,12-diaminododecane (DAD), as well as with two aromatic amines, 1-aminopyrene (AP) and 1,5-diaminonaphthalene (DAN). The functionalization was performed under moderate heating at 150-180 °C in vacuum. Under such conditions, it proceeds through both amidation and epoxy ring opening reactions. Comparative characterization of pristine and amine-functionalized GOP mats was carried out by using Fourier-transform infrared, Raman, and X-ray photoelectron spectroscopy (XPS), thermogravimetric (TGA) and differential thermal analysis, scanning electron and atomic force microscopy (SEM and AFM, respectively). Besides that, we compared the stability in water, wettability, electrical conductivity and elastic (Young's) modulus of GOP mats before and after amine functionalization. The highest content of organic species was obtained in the case of GOP-ODA, followed by GOP-DAD, GOP-AP and GOP-DAN samples. The covalent functionalization increased mechanical and thermal stability of GOP, as well as its electrical conductivity. The magnitude of each effect depends on the particular chemical structure of amine employed, which allows for tuning a given GOP property. Morphological characterization by using SEM showed that, compared to pristine graphene oxide paper, amine-modified GOP mats become relatively ordered layered assemblies, in which individual GO sheets are organized in a near-parallel pattern. Financial support from the National Autonomous University of Mexico (grants DGAPA-IN101118 and IN200516) and from the National Council of Science and Technology of Mexico (CONACYT, grant 250655) is greatly appreciated. The authors also thank David A. Domínguez (CNyN of UNAM) for XPS measurements and Dr. Edgar Alvarez-Zauco (Faculty of Science of UNAM) for the opportunity to use TGA equipment.

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