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Pulicaria glutinosa plant extract: a green and eco-friendly reducing agent for the preparation of highly reduced graphene oxide†

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The environmentally friendly synthesis of nanomaterials using green chemistry has attracted tremendous attention in recent years due to its easy handling, low cost, and biocompatibility. Here we demonstrate a facile and efficient route for the synthesis of highly reduced graphene oxide (PE-HRG) by the green reduction of graphene oxide (GRO) using the *Pulicaria glutinosa* plant extract (PE). The phytomolecules present in the *P. glutinosa* extract are not only responsible for the reduction of GRO, but also for the functionalization of the surface of the PE-HRG nanosheets and stabilize them in various solvents, thereby limiting the use of any other external and harmful chemical reductants and surfactants. The effect of PE on the dispersibility of PE-HRG in various solvents was investigated by preparing PE-HRG with different amounts of PE, and the dispersibility of PE-HRG was compared with that of chemically reduced graphene oxide (CRG). The reduction of GRO was confirmed by ultraviolet-visible (UV-vis), Fourier-transform infrared (FT-IR), Raman and X-ray photoelectron (XPS) spectroscopies, thermogravimetric analysis (TGA), X-ray powder diffraction (XRD) and transmission electron microscopy (TEM).

Introduction

Graphene-based materials have been a target for nanotechnologists owing to their exceptionally high crystal and electronic qualities. They have emerged as promising new nanomaterials for a variety of exciting applications despite their short history.[1,2] Due to the stable 2D structure and superior thermal, mechanical and electrical properties,[3–5] graphene is widely used in various fields, including electronics,[6] energy storage,[7] catalysis,[8–11] biosensors[12] and biomedicine.[13,14] Although graphene has been used as a theoretical model to describe the electronic structure of graphitic species for over half a century,[15] researchers have had difficulties in obtaining experimentally relevant amounts of this material until the very recent development of mechanical[16] and chemical synthetic methods.[17–19] Tremendous attention has been paid to the low-cost bulk production of graphene and graphene-based materials,[20–22] for which several methods have been reported. Chemical exfoliation strategies have been applied extensively,[23–25] and sequential oxidation-reduction of graphite and chemical exfoliation of graphite, followed by chemical reduction typically yields flakes of graphene.[26,27] These techniques yield bulk amounts of graphene-like sheets that are best described as highly reduced graphene oxide (HRG).[28,29] This material is not defect free, but highly processable and can be incorporated with a variety of materials, including various graphene-based bio- and nano-composites.[30–32]

The reduction of graphite oxide (GO) or graphene oxide (GRO) to HRG can be performed by chemical,[33–35] thermal,[36] electrochemical[37] and photochemical methods.[38,39] The chemical reduction of GRO is considered the most promising for a large-scale preparation of HRG at a low cost.[40] To date, strong reducing agents such as hydrazine, sodium borohydride, hydroquinone, hydrohalic acid or formamidinesulfinic acid have been applied for the reduction of GRO to HRG.[41–43] However, these chemical reductants, particularly the commonly applied hydrazine, are hazardous and harmful to both human life and the environment. Moreover, the chemical methods may result in "doping" with other elements (nitrogen in the case of hydrazine), which leads to an alteration of the electronic

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