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Abstract

Graphene Nanoflakes Incorporating Natural Phytochemicals Containing Catechols as Functional Material for Sensors †

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Abstract: Phytochemical products start to be employed to assist 2D nanomaterials exfoliation. However, a lack of studies regarding the molecules involved and their capacity to give rise to functional materials is evident. In this work, a novel green liquid-phase exfoliation strategy (LPE) is proposed, wherein a flavonoid namely catechin (CT) exclusively assists the exfoliation of bulk graphite in conductive water-soluble graphene nanoflakes (GF). Physicochemical and electrochemical methods have been employed to characterize the morphological, structural, and electrochemical features of the GF-CT. Surprisingly, the obtained GF-CT integrates well-defined electroactive quinoid adducts. The resulting few-layers graphene flakes intercalated with CT aromatic skeleton ensure strict electrical contact among graphene sheets, whereas the fully reversible quinoid electrochemistry (ΔE = 28 mV, Ip, a/Ip, c = ~1) is attributed to the residual catechol moieties, which work as an electrochemical mediator. The GF-CT intimate electrochemistry is generated directly during the LPE of graphite, not requiring any modification or electro-polymerization steps, resulting in stable (8 months) and reproducible material. The electrocatalytic activity has been proven towards hydrazine (HY) and β-nicotinamide adenine dinucleotide (NADH), a pollutant and a coenzyme, respectively. High sensitivity in extended linear ranges (HY: LOD = 0.1 μM, L.R. 0.5–150 μM; NADH: LOD = 0.6 μM, L.R. 2.5–200 μM) at low overpotential (+0.15 V) was obtained using amperometry, avoiding electrode-fouling. Improved performances, compared with graphite commercial electrodes and graphene exfoliated with a conventional surfactant, were obtained. The GF-CT was successfully used to perform the detection of HY and NADH (recoveries 94–107%, RSD ≤ 8%) in environmental and biological matrices, proving the material exploitability even in challenging analytical applications. On course studies aim to combine the intrinsic conductivity of the GF-CT with flexible substrates, in order to construct flexible electrodes/devices able to house GF-CT-exclusively composed conductive films. In our opinion, the proposed GF-CT elects itself as a cost-effective and sustainable material, particularly captivating in the (bio)sensoristics scenario.

Keywords: nanostructured-functional-material; grapheme; 2D-materials; mediator; phytochemicals; catechol-moieties; liquid-phase-exfoliation

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