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Electrochemical sensors and biosensors based on less aggregated graphene



Xiangjie Bo, Ming Zhou*, Liping Guo*

Key Laboratory of Nanobiosensing and Nanobioanalysis at Universities of Jilin Province, Department of Chemistry, Northeast Normal University, 5268 Renmin Street, Changchun, Jilin Province 130024, PR China

A R T I C L E I N F O

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ABSTRACT

As a novel single-atom-thick sheet of sp² hybridized carbon atoms, graphene (GR) has attracted extensive attention in recent years because of its unique and remarkable properties, such as excellent electrical conductivity, large theoretical specific surface area, and strong mechanical strength. However, due to the π - π interaction, GR sheets are inclined to stack together, which may seriously degrade the performance of GR with the unique single-atom layer. In recent years, an increasing number of GR-based electrochemical sensors and biosensors are reported, which may reflect that GR has been considered as a kind of hot and promising electrode material for electrochemical sensor and biosensor construction. However, the active sites on GR surface induced by the irreversible GR aggregations would be deeply secluded inside the stacked GR sheets and therefore are not available for the electrocatalysis. So the alleviation or the minimization of the aggregation level for GR sheets would facilitate the exposure of active sites on GR and effectively upgrade the performance of GR-based electrochemical sensors and biosensors. Less aggregated GR with low aggregation and high dispersed structure can be used in improving the electrochemical activity of GR-based electrochemical sensors or biosensors. In this review, we summarize recent advances and new progress for the development of electrochemical sensors based on less aggregated GR. To achieve such goal, many strategies (such as the intercalation of carbon materials, surface modification, and structural engineering) have been applied to alleviate the aggregation level of GR in order to enhance the performance of GR-based electrochemical sensors and biosensors. Finally, the challenges associated with less aggregated GR-based electrochemical sensors and biosensors as well as related future research directions are discussed.

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* Corresponding authors.

E-mail addresses: zhoum739@nenu.edu.cn (M. Zhou), guolp078@nenu.edu.cn (L. Guo).

Abbreviations: APAP, acetaminophen; AA, ascorbic acid; AFM, atomic force microscopy; CNTs, carbon nanotubes; CEA, carcinoembryonic antigen; CVD, chemical vapor deposition; Chox, cholesterol oxidase; Co3O4, cobalt oxide; CV, cyclic voltammogram; CD, cyclodextrin; DA, dopamine; ERGO, electrochemically reduced GO; ECL, electrochemiluminescence; ET, electron-transfer; FET, field-effect transistor; GC, glassy carbon; Gox, glucose oxidase; GONW, GO nanowall; GR, grapheme; GO, graphene oxide; HRP, horseradish peroxidase; H₂O₂, hydrogen peroxide; HP-β-CD, hydroxypropyl-β-CD; IL, ionic liquid; LBL, layer-by-layer; LOD, limit of detection; LUMO, lowest unoccupied molecular orbital; HOMO, highest occupied molecular orbital; MG, methylene green; MIP, molecularly imprinted polymer; NPs, nanoparticles; NWA, nanowire arrays; NADH, nicotinamide adenine dinucleotide; NO, nitric oxide; PTCA, perylene tetracarboxylic acid; POM, polyoxometalate; PDDA, poly (diallyldimethylammonium chloride); PAA, poly(acrylic acid); PSS, poly(styrene sulfonate); PQ11, poly[(2-ethyldimethylammonioethyl methacrylate ethyl sulfate)-*co*-(1-vinylpyrrolidone)]; PFIL, poly-ethyleinmine-functionalized IL; PVP, polyvinylpyrrolidone; QDs, quantum dots; RGNW, reduced GR nanowall; RGNS, reduced GR nanosheet; SEM, scanning electron microscopy; 2D, two-dimensional; UA, uric acid; VACNTs, vertically aligned CNTs

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1. Introduction

Two-dimensional (2D) materials with atomic thickness are considered as a kind of ideal building blocks for the design of various novel functional materials because of their novel electronic, optical, and mechanical properties (Guo et al., 2015). The atomically thick 2D materials not only inherit the properties from their corresponding bulk samples but also exhibit many totally different properties (Dubertret et al., 2015). Because of their unique physical and chemical properties, such 2D materials have been attracted much attention for both of the fundamental scientific research and practical applications (Chhowalla et al., 2015; Chia et al., 2015; Dubertret et al., 2015; Yang et al., 2015b). Graphene (GR), a monolayer of sp²-bonded carbon atoms arranged in honeycomb lattice, is a typical 2D material for carbon materials (Mas-Balleste et al., 2011). In 2004, Geim and co-workers at the University of Manchester developed a very simple methodology (the so called scotch-tape technique) to isolate the GR, which allowed them to first produce and characterize few-layers GR on silicon wafers (Novoselov et al., 2004). Following this pioneering work, several physical and chemical methods have been developed for the production of GR, including the intercalation and chemical exfoliation of graphite, unrolling of carbon nanotubes (CNTs), chemical vapor deposition (CVD) or epitaxial growth, reduction of graphene oxide (GO) and other organic synthetic methods (Avouris and Dimitrakopoulos, 2012; Brownson et al., 2012; Cai et al., 2012; Cao et al., 2014; Chen et al., 2012a; Edwards and Coleman, 2013; Guo and Dong, 2011; Hummers Jr and



Fig. 1. Electrochemical sensors or biosensors based on NPs/GR (A), heteroatom-doped GR (B), redox mediators/GR (C), biomolecules/GR (D), recognition system/GR (E), MIP/ GR (F), polymer/GR (G), and GR QDs (H).

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