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Recent developments of metallic nanoparticle-graphene nanocatalysts

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Graphene-based nanomaterials are promising in a wide range of applications due to their unique structural, chemical and physical properties that have recently led to their investigations as co-catalysts and nanoparticle catalyst templates for organic, electrochemical and photochemical reactions. Herein, we present an overview of hybrid metal nanoparticle-graphene catalysts with specific attention to the most recent achievements since 2015. The state-of-the-art of their synthesis and characterization is also summarized, and the synergistic metallic nanoparticle-graphene interactions and their functions in catalysis are discussed. Finally, challenges and future outlooks for this hot area are envisaged.

Contents

Abbreviations: AB, ammonia borane; AA, ascorbic acid; AFM, atomic force microscopy; CTAB, cetyltrimethylammonium bromide; CCG, chemically converted graphene; $HAuCl_4$, chloroauric (or tetrachloroauric) acid; CB, conduction band; DFT, density functional theory; DMAB, dimethylamine-borane; DBFC, direct borohydride fuel cell; DEFCs, direct ethanol fuel cells; DSC, dye-sensitized solar cell; EA, elemental analysis; ECSA, electrochemical surface area; SWP, electrochemical squarewave potential; ORR, electrochemical reduction of oxygen; EDTA-2Na, ethylenediaminetetraacetic acid disodium salt; ED, ethylene diamine; EG, ethylene glycol; EOR, ethanol oxidation reaction; FAOR, formic acid oxidation reaction; FA, formic acid; GCE, glass carbon electrode; GO, graphene oxide; GONR, graphene oxide nanoribbons; HEH, Hantzsch 1,4-dihydropyridine; HER, hydrogen evolution reaction; ICP, inductively coupled plasma analysis; k_{app}, kinetic apparent rate constant; L-H, Langmuir-Hinshelwood; LSPR, localized surface plasmon resonance; MNP, metallic nanoparticle; MOR, methanol oxidation reaction; MeAB, methylamine borane; MDR, methanol decomposition reaction; MWCNTs, multiwall carbon nanotubes; NB, nanobelts; NC, nanocluster; NP, nanoparticle; NRs, nanorods; NT, nanotubes; NIR, near-infrared; NRGO, nitrogen-doped reduced graphene oxide; NG, nitrogen-doped graphene; ODA, octadecylamine; OER, oxygen evolution reaction; PIRET, plasmon-induced resonance energy transfer; PDA, polydopamine; PPI, polyethyleneimine; PEI, polyetheylenimine; PDDA, poly(diallyldimethylammoniumchloride); PEDOT, poly(3,4-ethylenedioxythio phene); PEDOT, Poly(3,4-ethylenedioxythiophene); rGO, reduced graphene oxide; RhB, rhodamine B; rt, room temperature; RRDE, rotating ring-disk electrode; STM, scanning tunneling microscopy; SEM, scanning electron microscopy; SDS, sodium dodecyl sulfate; RDE, thin-film rotating disk electrode; TOF, turnover frequencies; 2D, two-dimensional; TGA, thermogravimetric analysis; TEM, transmission electron microscopy; TETA, triethylene tetramine; VB, valence band; XRD, X-ray diffraction; XPS, X-ray photoelectron spectroscopy; ODE, 1-octadecene; HEPES, 2-[4-(2-hydroxyethyl)-1-piperazinyl]ethan esulfonic acid; 2-CP, 2-chlorophenol; 3DG, 3D graphene; 4-NP, 4-nitrophene.

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1. Introduction, scope and organization of the text

Since the first isolation of graphene [1], followed by the recognition of its significance by the Physics Noble Prize in 2010, the interest of graphene and its derivatives has extraordinarily increased in the fields of condensed-matter physics [2–4], materials science [5–8], and chemistry [9–14]. The unique and superior physical properties of graphene, such as high carrier mobility, high electrical and thermal conductivity are now frequently utilized with new applications. Chemists have taken advantage of its huge surface area (theoretical value of 2630 m^2/g), unique two-dimensional (2D) structure, high electron mobility and easy surface functionalization [13,15–17]. With such remarkable properties, the research on graphene and its nanocomposites, graphene oxide (GO) and reduced graphene oxide (rGO), have successfully led to the discovery of efficient nanocatalysts that have been reviewed in organic chemistry [18–20], photochemistry [19,21] and electrochemistry [11,22–25].

Yet, much prior to the exfoliation of graphene from graphite, seminal studies on the excellent catalytic activity of metal nanoparticles (MNPs) for a large variety of reactions, in particular very small ones (<5 nm), as exemplified by Haruta'seminal studies [26], had attracted attention [27]. From then on, this topic has been very active owing to the implications from molecular, theoretical and physical chemistry as well as to the improved understanding of how these nanocatalysts function [28–37]. The remarkable properties of NPs are related to their easy preparation, stabilization and handling through nanoengineering of their size, shape, surrounding environment and metal-support interactions.

When these two active topics of graphene nanomaterials and MNP catalysts met, the field of nanocatalysis was revolutionized, speeding up research and leading to the production of hundreds of research papers per year in the fields of graphene nanomaterial-supported NP catalysis of organic, electrochemical and photochemical reactions (Fig. 1). Given the fast growing achievements in this new area, comprehensive review of the state-of-the-art in the synthesis, characterization, and catalytic applications becomes timely. Reviews on MNP/graphene hybrid nanomaterial catalysts have covered electrochemistry [38–40], photocatalysis [41], and general heterogeneous catalysis using graphene as support [42,43], but a comprehensive review dealing with the MNPs/graphene hybrid nanomaterials for catalysts has not yet appeared, especially including recent years. Such a review is proposed here for the concepts and recent achievements covering research since 2015.

Thus in this Review we will systematically discuss the MNP/graphene hybrid nanomaterials in terms of their state-of-the-art synthesis (Section 2), characterization and theoretical understanding with emphasis on the synergistic interactions