



Graphene-based materials with tailored nanostructures for energy conversion and storage



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ABSTRACT

Intensive interest in graphene has centered on its unique 2D crystal lattice and remarkable properties that offer unique opportunities to address ever-increasing global energy demands. The past years have witnessed considerable advances in the fabrication of graphene-based materials and significant breakthroughs in advanced energy applications. In this Review, two methodologies for graphene production, namely, the bottom-up growth from hydrocarbon precursors and the top-down exfoliation of graphite (to graphene) and graphite oxide (to graphene oxide followed by reduction) are first summarized. The advantages and disadvantages of these methods regarding their accessibility, scalability, graphene quality, and inherent properties are compared. Particular attention is concentrated on tailored nanostructures, electronic properties, and surface activities of these intriguing materials. The preparation of graphene-based composites containing a wide range of active constituents (e.g., transition metals, metal oxides, and conducting polymers) by *in-situ* hybridization and *ex-situ* recombination is also discussed with an emphasis on their microstructures and hybrid architectures. This Review is devoted largely to current developments of graphene and its derivatives and composites in energy conversion (i.e., polymer solar cells, dye-sensitized solar cells, perovskite solar cells, and fuel cells) and energy storage (i.e., lithium-ion batteries and supercapacitors) on the basis of their intrinsic attributes in improving photovoltaic and electrochemical performance. By critically evaluating the relationship between the nanostructures and the device performance, we intend to provide general guidelines for the design of advanced graphene-based materials with structure-to-property tailored toward specific requirements for targeted energy applications. Lastly, the potential issues and the perspective for future research in graphene-based materials for energy applications are also presented.

Abbreviations: AFC, alkaline fuel cell; AFM, atomic force microscopy; BET, Brunauer–Emmett–Teller; BHJ, bulk-heterojunction; CNT, carbon nanotube; CTAB, cetyltrimethyl ammonium bromide; CVD, chemical vapor deposition; CV, cyclic voltammetry; DMAC, N, N'-dimethylacetamide; DMF, N, N'-dimethylformamide; DMFC, direct-methanol fuel cells; DMSO, dimethyl sulfoxide; DSSC, dye-sensitized solar cell; ECSA, electrochemically-active surface area; EDLC, electric double-layer capacitors; EIS, electrochemical impedance spectroscopy; ETL, electron transport layer; FOM, figure of merit; FTO, fluorine doped tin oxide (FTO); GBM, graphene-based material; GIC, graphite intercalated compound; GO, graphene oxide; QGD, graphene quantum dot; HF, hydrofluoric acid; HOMO, highest occupied molecular orbital; HOPG, highly oriented pyrolytic graphite; HRTEM, high resolution transmission electron microscopy; HTL, hole transport layer; IPA, Isopropanol; IPCE, incident photon-to-current conversion efficiency; ITO, indium tin oxide; LBL, layer-by-layer; LED, light-emitting diode; LIB, lithium-ion battery; LUMO, lowest unoccupied molecular orbital; MCFC, molten-carbonate fuel cell; MOR, methanol oxidation reaction; MPECVD, microwave plasma-enhanced CVD; MPN, 3-methoxypropionitrile; NMP, N-methylpyrrolidone; ORR, oxygen reduction reaction; PAA, poly(acrylic acid); PAFC, phosphoric-acid fuel cell; PANI, polyaniline; PC, propylene carbonate; PCBM, [6,6]-phenyl C₆₁-butyric acid methyl ester; PC71BM, [6,6]-phenyl C₇₁-butyric acid methyl ester; PCE, power conversion efficiency; PDDA, poly(diallyldimethylammonium chloride); PEDOT, poly(3,4-ethylenedioxythiophene); PEG, poly(ethylene glycol); PEM, polymer electrolyte membrane; PEMFC, polymer-electrolyte membrane fuel cell; PEO, polyethylene oxide; PET, poly(ethylene terephthalate); P3HT, poly(3-hexylthiophene); PMII, 1-propyl-3-methyl imidazolium iodide; P3OT, poly(3-octylthiophene); PPy, polypyrrole; PS-*b*-PMMA, poly(styrene-*b*-methyl methacrylate); PSC, polymer solar cell; PSS, polystyrene sulfonate; PVSC, perovskite solar cell; QD, quantum dot; QSSE, quasi-solid-state-electrolyte; RGO, reduced GO or reduced graphene oxide; RIE, reactive ion etching; SAED, selected-area electron diffraction pattern; SDBS, sodium dodecyl benzene sulfonate; SEI, solid electrolyte interphase; SEM, scanning electron microscopy; SOFC, solid-oxide fuel cells; SSA, specific surface area; STM, scanning tunneling microscopy; TCE, transparent conducting electrode; TEM, transmission electron microscopy; vdW, van der Waals; R_{CT}, charge-transfer resistance; FF, fill factor; δ_T, Hildebrand solubility parameter; V_{OC}, open-circuit voltage; γ, surface tension; R_S, sheet resistance; J_{SC}, short-circuit current density; T, transparency (optical).

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By summarizing the current state-of-the-art as well as the exciting achievements from laboratory research, this Review aims to demonstrate that real industrial applications of graphene-based materials are to be expected in the near future. (1346 references).

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1. Introduction

In response to the limited fossil fuel resources, emerging ecological concerns and global warming, ever-increasing energy consumption and human reliance on energy-based appliances, the emerging development of clean, renewable, and sustainable energy techniques has become increasingly important than ever. In this context, current energy systems including solar cells, [1–3] fuel cells, [4–6] lithium batteries [7–9] and supercapacitors, [10–12] have

attracted much attention in academia and industry alike. It is well known that these energy devices in general possess an active or electrolyte layer sandwiched by two electrodes with their overall performance intrinsically and sensitively dependent on the materials used [13]. Over the past decade, an exciting set of emerging nanomaterials such as nanocarbons, metal-containing compounds and functional nanocomposites with unusual nanoscopic structure-dependent properties, have been largely developed. These materials are promising for fundamental research and

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