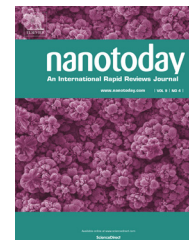




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REVIEW

Nanocarbon-based electrochemical systems for sensing, electrocatalysis, and energy storage



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Summary Carbon materials are important for many electrochemical applications due to their tunable electron-transfer and charge-storage properties. Judicious structural manipulation of carbon to modulate its chemical, electronic, and crystalline properties is key to the rational design of many high-performance electrochemical devices. Here we focus on three types of carbon nanomaterials of recent interest in electrochemistry, namely, carbon nanofibers, carbon nanotubes, and graphene. We concentrate on how structural variations in these carbon nanomaterials impact their electrochemical activities. In this review, following a brief overview of the synthesis methods for each class of carbon nanomaterials, we discuss their electrochemical applications for sensing, electrocatalysis, and energy storage, with emphasis on general carbon structure manipulation strategies that impart specific functionalities to suit each application area. Special attention is devoted to articulating how the electronic structure of carbon influences its electrochemical activity. Through the analysis of different electrochemical devices, we find that some of the modification techniques apply to more than one application area; thus structural manipulation methods in one class of electrochemical devices may be extended to other types.

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Introduction

Carbon materials are used extensively in both analytical and industrial electrochemistry, and are important for many electrochemical applications due to their tunable electron-transfer and charge-storage properties [1,2]. Carbon has several advantages relative to other electrode

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materials, including low cost, a wide working window (i.e., the electrochemical potential range over which the electrode can perform its normal function), and high electrocatalytic activities for a variety of redox-active chemical and biological systems. Structural manipulation of carbon to modulate its chemical, electronic, and crystalline properties is key to the design of many high-performance electrochemical devices, for example, sensors [3], lithium-ion batteries [4], and supercapacitors [5], to name a few. Moreover, carbon surfaces can be modified easily by a variety of noncovalent and covalent methods, thereby broadening significantly the utility of carbon electrodes. Noncovalent modification mostly relies on π -stacking interactions between the adsorbates and carbon surfaces. Common chemical structures that exhibit π -stacking interactions with sp^2 -conjugated carbon surfaces include nucleotide bases [6–9], pyrenes [10–12], porphyrins [13–15], and metallocenes [16,17]. Various types of reactions have been exploited to modify carbon surfaces covalently; most commonly used reactions are diazonium ion reduction [18], amine oxidation [19], photochemical reactions [20], and click cycloaddition chemistry [21].

Here we focus on three types of carbon nanomaterials of recent interest in electrochemistry, namely, carbon nanofibers (CNFs), carbon nanotubes (CNTs), and graphene. Electrochemical applications of the “classical” carbon materials based upon graphite, glassy carbon, and carbon black have been covered extensively in several earlier reviews and monographs [22–24]. In this review, following a brief overview of the synthesis methods for each class of carbon nanomaterials, we discuss their electrochemical applications for sensing, electrocatalysis, and energy storage (including both supercapacitor and lithium ion battery (LIB) applications). The field of nanocarbon electrochemistry has developed rapidly in recent years; this review focuses primarily on general strategies developed for the manipulation of carbon structure, as summarized in Table 1, with emphasis on elucidation of the relevant structure-property relationships for each application area for carbon nanofibers, nanotubes, and graphene. We devote special attention to the intrinsic properties of carbon nanomaterials that give rise to their high electrochemical activities; hence, in the last section of this review we discuss how the electronic structure of carbon influences its performance for sensing, catalysis and energy storage.

Synthesis of CNFs, CNTs and graphene-based materials

Fibrous carbon materials have attracted widespread attention, both fundamentally and for practical applications [25]. In the late 1950s, Bacon succeeded in producing the first high performance carbon fibers [25]. A few years later, in the early 1960s, polyacrylonitrile (PAN)-based carbon fibers were developed by Shindo [26], following which Singer, in 1970, developed pitch-based carbon fibers [27]. Generally, carbon fibers are produced either by pyrolyzing fibers spun from an organic precursor, or by chemical vapor deposition (CVD) [28]. The major spinnable carbon precursors include PAN, pitch, polybenzimidazol (PBI), poly(amic acid) (PAA), phenolic resin, polyvinylpyrrolidone (PVP), polyimide (PI),

poly(vinyl alcohol) (PVA), and cellulose acetate (CA) [29,30]. Recently, electrospinning followed by thermal treatment has emerged as a simple, efficient and scalable method to generate ultrafine carbon nanofibers [30]. Electrospinning has the advantages of simplicity, efficiency, low cost, high yield and high degree of reproducibility of the obtained materials. The versatility of electrospinning is not only related to the nature of the precursors and electrospun materials, but also extends to the fiber assemblies and architectures of the final products. Modifications of solution and processing parameters [31,32] and/or set-up geometries [33] permit the preparation of complex nanostructures with controllable hierarchical features such as nonwoven, aligned or patterned fibers, nanoribbons, nanorods, random three dimensional structures, sub-micron springs and convoluted fibers with controlled diameters. Additionally, the possibility of co-electrospinning different solutions using specially designed spinnerets leads to the elaboration of core–sheath fibers [33,34], hollow fibers [35], porous fibers [36], and multichannel tubes [37].

Carbon is well known to form distinct solid state allotropes with diverse structures and properties. CNTs can be viewed as one-dimensional cylinders composed of a single layer or a few layers of graphene with high length-to-diameter ratios ranging from 10^2 to 10^7 . The first observations of CNTs and their subsequent large-scale synthesis using arc discharge techniques were reported by Iijima and coworkers [38]. Laser ablation was later demonstrated by Smalley et al. [39] to be an alternative method for growing CNTs. Once it was observed that transition metals embedded in graphite electrodes produced carbon nanotubes with higher yield and reproducibility [40], chemical vapor deposition using transition metal nanoparticle catalysts was developed to produce high quality single-walled and multi-walled CNTs (SWCNTs and MWCNTs, respectively) in vertically aligned arrays [41,42]. Vertically aligned arrays can also be grown on metallic and quasicrystalline substrates. The metal nanoparticle catalyst concept was extended later to the synthesis of CNTs by pyrolysis of metal carbonyls in the presence of other hydrocarbons [43]. When optimized in a high pressure carbon monoxide (HiPco) environment, the carbonyl pyrolysis process led to high yield production of SWCNTs [44].

Graphene is the latest carbon nanomaterial to be isolated; this feat was accomplished on an insulating substrate and the allotrope was electrically characterized by Novoselov et al. in 2004 [45]. Graphene is often viewed as the two-dimensional building block for other sp^2 hybridized carbon nanomaterials because, conceptually, it can be rolled or distorted to form carbon nanotubes and fullerenes. In 1962, Boehm et al. [46] reported the existence of very thin carbon foils, produced from the reduction of graphite oxide in a dilute alkaline solution. In 1998, Forbeaux et al. [47] reported a heteroepitaxial growth method that opens up the possibility for isolation of a single graphene layer above a substrate. In addition, some attempts at mechanical isolation of graphene were made prior to 2004 [48]. However, the most definitive evidence of monolayer graphene and its electrical properties was provided by the Manchester group using mechanically exfoliated graphene [45]. Although historically significant, this so-called “Scotch tape” technique for producing graphene lacks sufficient scalability for

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