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Invited Review

Graphene materials-based energy acceptor systems and sensors

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ABSTRACT

Graphene materials have recently attracted considerable attention because of its extraordinary mechanical, electronic, thermal and optical properties, leading to the wide application of graphene such as in biology and energy areas. In recent years, energy-transfer based optical biosensors using graphene materials as the energy acceptors have become the focus of researches, which take the advantages of the high surface area and ultrahigh luminescence quenching efficiency of graphene materials. These sensors have extensively covered the detection of DNA, protein, enzyme activity, metal ions and other small molecules. In this review article, we aim to provide a comprehensive discussion on the development of the graphene materials-based energy acceptor systems and sensors, sorting the sensors according to the probes with which the energy acceptors are assembled to or conjugated with the luminescent energy donors. At the end we also present an overview of future perspective and possible challenges in this rapidly developing area.

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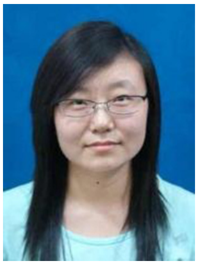

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1. Introduction

Graphene, an increasingly concerned star nanosized material first reported in 2004 [1], is a single-layer planar sheet consisting of sp^2-hybridized carbon atoms perfectly ordered in a honeycomb lattice [2,3] (Fig. 1). The unique physicochemical and structural properties [4–6] of graphene make it stand out and applicable to various technologies such as field emission display [7,8], nanoelectronics and high-frequency electronics [9–11], transparent conductors [12] and energy storage [13,14]. What is particularly noteworthy, graphene has proved to be a promising material in bio/chemo sensing in its short history of research, thanks to its high electron transfer rate, high quenching efficiency, ultra-large planar structure and large surface area, excellent carrier mobility and carrier capacity, and good mechanical properties [15].

As the partially oxidized pattern of graphene, graphene oxide (GO) is also featured with its single-atom thickness and shows similar optical properties as graphene. Therefore, the reported optical sensors use either graphene or GO as the energy acceptor; and the item 'graphene materials' in this article include both the two patterns and does not differentiate them. But it should be clarified that GO is decorated with oxygen-containing groups, which enable covalent conjugation with other molecules and endow GO with improved water solubility as compared to graphene [17]. On the other hand, graphene was reported to have broader absorption spectrum and higher electron transport property than GO, hence higher luminescence quenching efficiency [18].

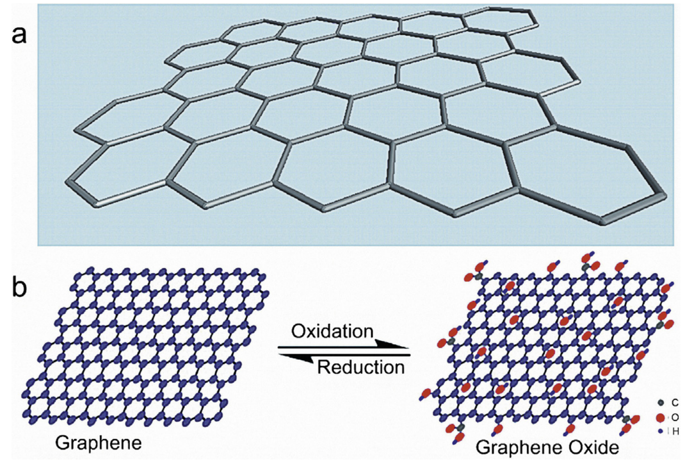

Fig. 1. (a) The planar six-member ring lattice structure of graphene. (b) The interconversion between graphene and graphene oxide (GO). Reprinted with permission from Ref. [16]. Copyright (2012) American Chemical Society. Similar to graphene, GO is featured by its single-atom thickness. But it is decorated with oxygen-containing groups, which enable covalent conjugation with other molecules.

The optical sensors with graphene materials as the energy acceptors have already demonstrated great success in recent years mainly because of the following strong characteristics of the material: (1) the ultrahigh luminescence quenching efficiency; (2) the large planar surface which allows simultaneous adsorbing and quenching of multiple probes to achieve multicolor detection in the same solution; (3) the excellent capabilities for direct and easy wiring with biomolecules due to the π–π stacking and hydrophobic interactions [17]. These sensors have been applied for detection of DNA, protein, enzyme activity, metal ions and other small molecules with favorable performances.

This review is intended to provide a comprehensive discussion on the development of the optical sensors using graphene materials as the energy acceptors in recent years emphasizing the detection mechanisms of the sensors. Finally we present an overview of future perspective and possible challenges in this rapidly developing area. We expect to attract broader interest and promote greater development in this area. To point out, since graphene materials do not luminesce in the energy donor–acceptor pairs, it generally acts as a dark quencher in the energy transfer systems. As such the energy acceptor is typically referred to as a 'quencher' in most literatures, and also in this article.

2. Luminescence quenching in graphene materials

The energy of the electrons of graphene is linearly correlated to the wave vector near the crossing points in the Brillouin zone [19]. Dirac equation describes the relativistic particles, which match the behavior of the charge carriers of graphene better than Schrödinger equation does [9]. Therefore, graphene shows high conductivity and luminescence quenching ability [20]. The ultra-high quenching efficiency of graphene materials is attributed to three possible mechanisms: (1) Förster resonance energy transfer (FRET), (2) surface energy transfer (SET), (3) photo-induced electron transfer [21].

The occurrence of FRET requires a pair of suitable energy donor and energy acceptor which have appropriate spectral overlap (the donor emission and the acceptor absorption) and are in proximity of ca. 1.0–10.0 nm (Fig. 2). In the process that the energy transfers from the excited donor to the acceptor in a non-radiative manner through a dipole–dipole interaction [22], the transfer efficiency

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