

Graphene-quantum dot hybrid materials on the road to optoelectronic applications



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ABSTRACT

With its unique properties, graphene has demonstrated not only high potential for various applications, but is also the subject for studying various fundamental physics. However, due to the lack of a band gap in intrinsic graphene, it is very difficult to separate the charges from photo-excitons and manipulate them for useful purposes within graphene itself. This obstructs its use as active material in photovoltaics (PVs) and photodetectors (PDs). This drawback can be overcome by either chemically or physically combining graphene with semiconductor quantum dots (QDs) to form QD-graphene hybrid materials. These hybrid materials possess designable multifunctional or even completely new properties, which are synergetic combinations of the outstanding properties of graphene with the tunable optoelectronic properties of semiconducting QDs. As a result, various achievements in using QD-graphene hybrid materials as active materials in PV and PD applications have been recently demonstrated. This review will provide comprehensive discussions on recent developments in semiconducting QDs-graphene hybrid materials designed towards PVs and PDs applications. First, the synthesis approaches to QD-graphene hybrid materials will be summarized. In the second part, charge transfer processes occurring within these hybrid materials will be discussed, since it is the basic mechanism behind their applications in PVs and PDs. Finally, recent developments in applications of QD-graphene hybrid materials in PVs and PDs will be addressed.

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1. Introduction

Graphene, a single-atomic layer of sp^2 carbon atoms, has recently been attracting tremendous attention within the scientific community owing to its unique properties such as high conductivity, optical transparency and mechanical stability [2,3]. A single graphene layer for example can absorb 2.3% light power of any wavelength [1]. With the discovery of graphene, Geim and Novoselov were awarded the Nobel Prize in Physics in 2010. So far, several methods are utilized to synthesize graphene. Pristine graphene was firstly obtained by using Scotch tape to peel off a single or few layers of graphene sheets from bulk graphite which was reported by Geim et al. [4]. Alternatively, the chemical reduction of graphene oxide (GO) is a method for obtaining graphene-like structures in high quantities [5], while chemical

vapor deposition (CVD) is the method of choice to produce thin and continuous large area graphene films [6]. Already, various applications utilizing graphene based materials have been reported, such as transparent electrodes and harvesting layers in solar cells [7–9], catalytic electrodes in fuel cells [10,11], supercapacitors [12,13], electrodes in transistors [14], and photodetectors [15,16]. Despite the promising properties and potential applications, intrinsic graphene has no a band-gap, thus graphene transistors cannot be fully switched off, which obstructs its application for digital electronics. Therefore, recently a new research area on graphene hybrid materials evolved where properties of semiconducting materials are combined with the outstanding properties of graphene. Meanwhile, semiconducting nanocrystals (NCs), also known as quantum dots (QDs) have easily tunable optical and electrical properties [17]. Moreover, when QDs absorb a photon with higher energy than their band gap energy, more than one exciton can be produced through exciton-multiplication, enhancing photocurrent—solar energy conversion [18–20]. Therefore, QDs already demonstrated their potential for

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various applications such as photovoltaics (PVs) [21–25] and photodetectors (PDs) [17,26–28]. Semiconductor-graphene hybrid materials possess designable multifunctional or even completely new properties. Such synergetic properties also lead to various novel applications such as in catalysis or even biological applications which have recently been reviewed elsewhere [29,30]. Moreover, semiconductor nanoparticles are often synthesized in solution with ligand shells to prevent aggregation; this in turn decreases the effectivity as catalysts and charge transfer processes in fuel cells and solar cells applications [31]. Furthermore, when nanoparticles are incorporated into devices and the synthesis ligands are exchanged ideally by a monolayer of more conductive ligands, the nanoparticles tend to agglomerate [32] leading to a significant reduction of their active surface area. This general problem can be overcome by using graphene as framework to support the nanoparticles, keeping them separated and avoiding aggregation. Additionally it favors charge transfer processes at the nanoparticle-graphene interface as well as charge transport processes making them ideal candidates for interlayers or electrode support materials in photodetectors or photovoltaics. The current review will provide comprehensive discussions on recent developments in semiconducting QDs-graphene hybrid materials designed towards PVs and PDs applications. So far, there exist only very few review articles discussing this topic [33,34]. With the pronounced light absorption ability, QDs can convert light energy into electron-hole pairs, also known as excitons. The excitons can then move to a donor-acceptor interface where they can be separated and transferred to graphene. In solar cell applications, these separated charges are transported to the respective electrodes via graphene. This review is focusing on CdSe, CdS, and PbS QDs-graphene hybrids since they have been well investigated showing great potential for optoelectronic applications. In the first part, the synthesis routes to QD-graphene hybrid materials will be summarized. In the second part, charge transfer occurring within these hybrid materials will be discussed, since it is the basic mechanism for their applications in PVs and PDs. Finally, recent developments in applications of QD-graphene hybrids in PVs and PDs will be presented.

2. Synthesis of QDs-graphene (rGO) hybrid materials

The hybrid materials can be prepared by decorating of graphene with QDs via different routes, which can be categorized into two main methods: *In situ* decoration and *ex situ* decoration.

2.1. In situ decoration of rGO with QDs

In this method, the QDs are grown directly on the surface of graphene or their derivatives. The functional groups of GO and rGO, such as alcohols (C–OH), carbonyl (C=O), and carboxyl (COOH) serve as anchor points for QDs attachment through electrostatic interactions. In an early work, Juárez et al. [35] studied the mechanism of CdSe QD attachment to carbon nanotubes (CNT). Several factors including the presence of water, chloride ions, and the interaction of QDs with the sp^2 carbon lattice of CNT determine the formation of the QD-CNT hybrids. The authors demonstrated the formation of CdSe QDs graphene hybrid materials. In a later report, rGO/CdSe nanocomposites were synthesized by introducing rGO directly into the reaction solution during the synthesis of CdSe QDs [36]. By this method, single-crystal CdSe QDs with sizes of a few tens of nanometers were obtained, and were anchored onto the rGO surface forming CdSe QD decorated rGO nanocomposites. Photoinduced charges from CdSe QDs were proven to be transferred to rGO by photoluminescence quenching experiments. This charge transfer results in a dramatic enhancement in photoresponse of photodetectors based on these nanocomposites, and therefore exhibited their promising application in optoelectronics. In this work, GO was reduced by hydrazine to yield rGO. Afterwards, rGO was dispersed in 1,2 dichloroethane (DCE), which was then introduced into the reaction mixture of CdSe QD precursors: cadmium stearate/hexadecanediol/trioctylphosphine (Cd(SA)₂/HDA/TOPO). Therefore, the solubility of rGO in DCE and in the reaction solution is critical to obtain an efficient decoration. The mechanism for the formation of the nanocomposites has been described as follows. Initially, CdSe QDs are formed freely in the solvent. Subsequently they attach to the rGO surface via the Cd-rich (001) facet. This is because Cd-rich facets are able to react with the p-system based on noncovalent bonding, moreover the electronegativity of remaining oxygen-containing groups on rGO also facilitate the attachment of CdSe QDs *via* Cd-rich facets. The size, shape, and crystalline structure of CdSe QDs decorated on rGO sheets can be controlled via a facile microwave irradiation synthesis [37]. This method enables simultaneously the reduction of GO and the controlled synthesis of phase-selected CdSe QDs on rGO sheets. Depending on the synthesis ligand type, one can obtain either cubic CdSe QDs using a TOPO-HDA ligand mixture or hexagonal CdSe QDs without using the TOPO-HDA synthesis ligands, as shown in Fig. 1. In contrast with above mentioned work [36], the proposed mechanism for obtaining these structure was explained that the CdSe QDs are grown directly on the surface of

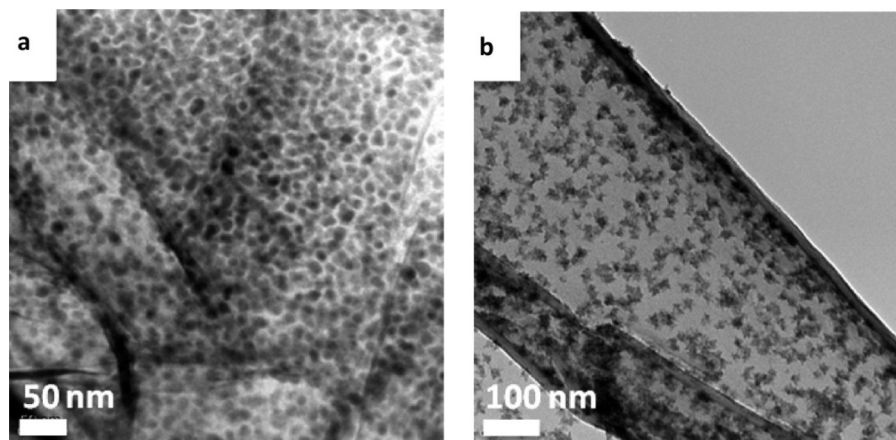


Fig. 1. TEM images of CdSe QDs with (a) cubic and (b) hexagonal structures. The QD decorated graphene sheets were prepared by microwave irradiation of cadmium oleate and TOP-Se in DMSO (hexagonal) and with the addition of TOPO and HDA (cubic). Adapted with permission from Ref. [37], Copyright (2010) American Chemical Society.

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