



Graphene oxide: Exploiting its unique properties toward visible-light-driven photocatalysis

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ABSTRACT

The oxygen-containing functionalities on the aromatic scaffold of graphene oxide (GO) enable these materials to facilitate both ionic and non-ionic interactions with a huge range of molecules. Endowed with a plethora of extraordinary properties, GO demonstrates remarkable potential in the field of photocatalysis. Furthermore, the tunability of its physicochemical properties by the facile exploitation of surface modifications and edge defects renders GO an even more intriguing nanomaterial. In this paper, the fundamental aspects on the manipulation of GO for the enhancement in photocatalytic performance will be thoroughly discussed. To mediate further research and development in this area, the current technical challenges faced as well as future research directions will also be included in this paper.

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1. Introduction

At present, frontier scientific and technological research into the fields of energy and environmental protection are becoming increasingly challenging due to the need for key materials with high efficiency and functionality. Ignited by immense technological potential, the research on advanced carbon nanomaterials, particularly graphene has experienced an extensive growth over the past decade [1–4]. Fig. 1 depicts the exponential increase in the number of publications based on graphene and its derivatives from 2004 to 2014. As a simple hexagonal lattice of carbon atoms, graphene provides a highly electronic and thermally conductive surface, which is also the thinnest known material and the strongest ever reported [5].

One specific branch of graphene research is focused on chemically derived graphene oxide (GO) [6,7]. As a newly emerging two dimensional (2D) material, GO possesses many intriguing properties, including mechanical, electronic, optical, electrochemical and chemical reactivity [8]. GO sheets are composed of two primary regions: (1) the hydrophobic π -conjugated sp^2 domains and (2) the sp^3 domains with hydrophilic oxygen-containing functional

groups. The widely accepted structure of GO depicts carboxylic acid at the edges, and hydroxyl and epoxide groups residing at the basal plane [6]. Despite the distortion of the intrinsic conjugated π system, these complex cocktails of functionalities can provide potential advantages for employing GO in numerous applications. The polar oxygen-containing groups on GO render it strongly hydrophilic, allowing it to be dispersed in a number of solvents, particularly in water [9]. Furthermore, the physicochemical properties of GO can be tuned by engineering its atomic and chemical structures [10]. From a broader perspective, the fascinating properties and tunability inherent to GO sheets are very promising for facilitating a wide range of applications, including but not limited to electronics (conductive films and sensors) [11], composite materials [12], clean energy devices [13] and biology [14]. Several reviews on these topics have been reported in literature [15–20]. However, to the extent of our knowledge, no review articles on the application of GO in the field of photocatalysis have been published.

Photocatalysis has become one of the most studied fields due to the increasingly cogent call for environmental remediation [21]. The direct conversion of solar energy into chemical energy through this system provides an opportunity to simultaneously address both the environmental and energy issues faced today. While many different photocatalytic materials such as semiconductors, transition-metal complexes *etc.* have been developed [22], the possibility of employing pure GO nanosheets for efficient

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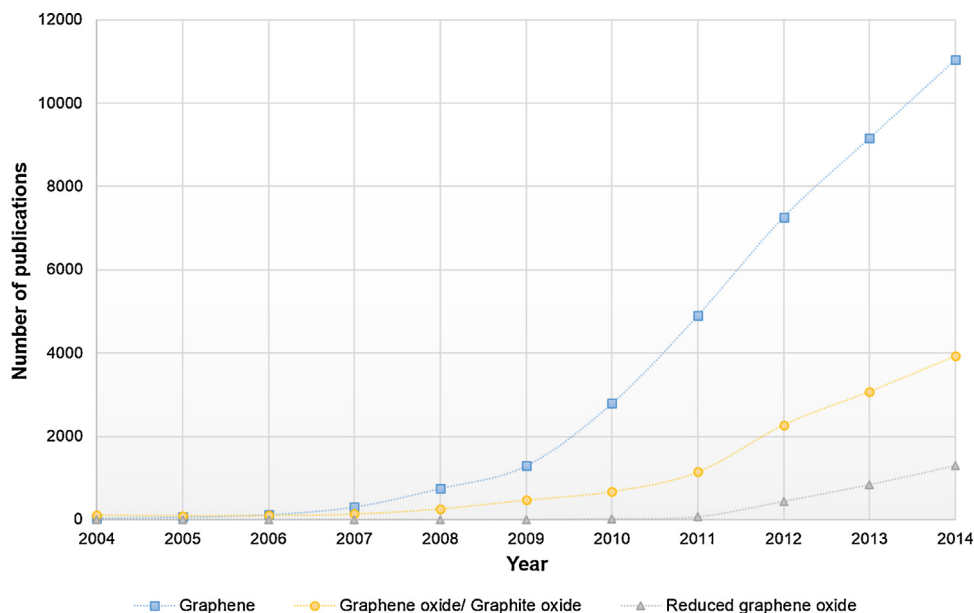


Fig. 1. Plot detailing the number of publications on graphene and its derivatives between 2004 and 2014; data were obtained using Scopus: keywords “graphene”; “graphene oxide OR graphite oxide” and “reduced graphene oxide”; respectively.

photocatalysis is extremely thought-provoking. Hence, in the paper, we will present a concise account on some fundamental aspects of GO and discuss how its properties can be controlled and tuned to benefit the process of photocatalysis. Advancements on the use of GO as a photocatalytic material will also be thoroughly reviewed. Finally, a brief summary and an outlook on the major challenges and opportunities for future research will be presented. It should be noted that historically, GO sheets have functioned mainly as heterogeneous supports for semiconductor nanoparticles [23–25], and its use as a standalone photocatalytic material is very rare. Hence, the main objective of this paper is to provide some insights into the possibility of employing bare GO sheets as efficient “carbocatalysts” in photocatalytic applications as varied as dye degradation, water splitting and so forth.

2. Unique properties of GO—key criteria affecting photocatalytic performance

2.1. Structural features

In essence, the structure GO can be viewed as graphene but covalently decorated with oxygen-containing groups on its planes and edges. In that account, GO still physically retains its original 2D nature and the benefits entailed with this structure. First and foremost, 2D materials associate to large specific surface area, yielding a theoretical value of as high as $\sim 2630 \text{ m}^2/\text{g}$ as seen in the case of pristine graphene [26]. In fact, through atomic force microscopy (AFM), the apparent thickness of single-layer GO was found to be around $\sim 1 \text{ nm}$ (Fig. 2(a)), which is a small deviation to the thickness of pristine graphene that varies in the range 0.3–0.6 nm [27–29]. This difference is accounted to the presence of bounded oxygen functionalities and the out-of-plane protrusion of sp^3 -hybridized carbon atoms. From a photocatalysis viewpoint, this massive area is helpful as GO can host a great number of active sites that can be directly accessible to the reactive species. In addition to that, the 2D structure of GO can also eliminate the occurrences of volume recombination from which ordinary bulk semiconductor suffers, that will be detrimental to the efficiency of the photocatalytic process [30]. This is understandable since the atom-thin structure of GO guarantees the rapid transfer of carriers to the surface, thus

leaving more carriers at disposal which can be successively consumed by acceptor molecules to drive the desired redox reaction.

Unlike having a definite physical structure, the chemical structure of GO is somewhat ‘fluid’, as its stoichiometry varies depending on sample-to-sample variability and their synthesis protocol [8]. This is further made complex by the inhomogeneities in the GO structure in which the precise identity and distribution of oxygen functional groups remain an ambiguous subject. A widely adopted model of GO structure [31,32] depict that epoxide and hydroxyl are the dominant oxygen functionalities on the basal plane, while carboxylic acid, ketones, phenols and lactols groups accommodate at edge terminations (Fig. 2(b)). Together, these cocktails of oxygen functionalities act in concert to endow properties, which although relevant to GO are not typical of pristine graphene. The interaction of the basal plane with oxygen-functionalized groups leads to the modification of the π - π conjugation and thus the electron density distribution, yielding the differences in its electrical and chemical properties, which benefit photocatalysis.

2.2. Electrical and optical properties

Even today, the precise electronic band structure of GO has not been clearly elucidated owing to the nano-scale inhomogeneities of the structure. The functionalization of graphene with oxygen groups involves a complex interplay of phenomena which influences graphene’s collective electrical, optical and chemical attributes. Essentially, the covalent addition of oxygen functionalities on the basal plane of graphene converts the original, unsaturated planar sp^2 - of carbon atoms into a tetrahedral sp^3 -electronic hybridization [34]. These in turn impose a structural disorder on the lattice, as shown in Fig. 3, which acts as transport barriers, since they interrupt the continuity of π -network which ordinarily allows classical carrier transport to occur. This removal of π electrons plays a mechanistic role in the change of electric properties by introducing a band gap *via* the symphonious removal of electronic states and emergence of new energy states from functionalizing oxygen groups. In that context, GO is viewed as an electronically hybrid material containing both conducting π -states from graphitic sp^2 -hybridized carbons and a large energy gap between the σ -states from its sp^3 -hybridized carbon

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