



In-situ fabrication of reduced graphene oxide (rGO)/ZnO heterostructure: surface functional groups induced electrical properties



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ABSTRACT

Reduced graphene oxide (rGO)/ZnO heterostructure was prepared via electrochemical deposition directly on Hummers method derived rGO membranes and the corresponding diodes had been fabricated. Rectifying I–V curve was obtained by modifying the functional groups on the surface of rGO. Further investigation for GO based transistors showed that the conductivity of rGO could vary from n-type to p-type under different annealing conditions. Based on Lerf-Klinowski model and X-Ray photoelectron spectroscopy, it was found that the C-sp², hydroxyl and epoxy in rGO would be responsible for the change of electrical properties. It was also concluded that reasonable p-type conductivity of rGO for obtaining rectifying rGO/ZnO heterostructure occurred while the percentage of C-sp² content was about 54% with the C-sp²/(OH + C–O–C) ratio around 1.6.

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1. Introduction

Wide bandgap oxide such as ZnO has declaimed to be very promising in optoelectronic applications such as solar cells and light emitting diodes, due to its low cost, varieties of synthesis methods and nanostructured morphologies, as well as the tunable optical and electrical properties. However, the difficulty in p-type doping of ZnO due to the rich defect chemistry, has hindered the further development of ZnO-based optoelectronic devices. Accordingly, another candidate to develop ZnO based optoelectronic devices is to use heterojunction device structure [1,2]. The most commonly used p-type materials for ZnO based heterojunction is Mg:GaN because of the same hexagonal structure and the reasonably low lattice mismatch (~1.5%) [1,3]. However, the carrier concentration and mobility of Mg:GaN are significantly lower than that of ZnO, in which case the recombination of carriers would occur on GaN side of the heterojunction, and the device performance are thus dominated by the material properties of GaN.

On the other hand, graphene, a one-atom-thick material with closely packed sp² bonded carbon honeycomb structure, has exhibited excellent and tunable conductivity. Graphene has a rich defect chemistry inducing vacancy, impurity, surface adsorbate,

grain boundary and topological defects [4–20], which results in varieties of the electrical and mechanical properties of graphene. For example, boron and phosphine can be used as the p-type and n-type dopant, respectively [5–7]. It has also been reported that p-type conductivity of graphene could be introduced in two ways: broadened band gap and confinement of electron transport due to quasi-one-dimensional size effect [4,14], or the adsorption of ambient gases [20]. Accordingly, the adjustable and controllable conductivity of graphene might provide an alternatively new approach to p-type materials for ZnO heterojunctions.

Among various kinds of graphene synthesis, Hummers method has provided a low cost, fast and productive approach for graphene via oxidizing graphite to dispersed GO sheet, which can be further reduced to reduced GO (rGO) via annealing or chemical reduction [17]. Different from graphene obtained by vapor deposition and exfoliation, the Hummers derived rGO has abundant hydration related defects uniquely. Recently, Lerf-Klinowski model has been commonly used to describe the structure of GO [18], which proposed that there are three hydration related functional groups forming chemical bonds with C atom: hydroxyl, epoxy and carboxyl. It has also been reported that annealing can promote the generation or extinction of these hydration related functional groups via different temperature, pressure and gas ambience [19–21].

Meanwhile, those kind of oxygen related functional groups such as hydroxyl widely exist as surface defects on the polar facets {0001} of ZnO, which are the dominant facets for growth. Oxygen

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vacancy and interstitial are also very common in metal oxides including ZnO [1]. The similarity in defect chemistry of graphene and metal oxides opens up the prospect of developing graphene/ZnO (or other metal oxides) heterostructures [1,22].

Graphene was usually mixed with metal oxides in powder or stamped to other as-prepared oxides to obtain hybrid materials [23,24], in which cases the surface defects could not promote the interfacial growth, and the interface between graphene and ZnO would not be influenced by only surface defects of graphene and ZnO, but also the gels used for stamping. Moreover, direct growth of ZnO nanostructure on graphene was rarely reported, which usually required a pre-fabricated seed layer [25]. In this work, rGO/ZnO heterostructure with diode architecture is developed by an in-situ deposition of ZnO nanorods directly on Hummers method derived rGO membranes. Electrochemistry, which could be applied in biological and environmental analysis [26–28], is employed to

synthesize ZnO nanorods on rGO. The interface between graphene and ZnO is modified to achieve rectifying I–V properties. It has been found that the post-growth annealing can change the chemical components of absorbed functional groups on the surface of graphene and then affect the electrical property in rGO/ZnO interface.

2. Experimental

Graphene was obtained by reduction of Hummers method derived graphene oxide as follows: 0.5 g expanded graphite, 3 g KMnO_4 and 0.5 g NaNO_3 (Xilong Chemical Co., Ltd., China) were mixed in 60 ml concentrated sulfur acid and the solution was stirred for at least 12 h until the color changed to dark green, then the solution was diluted by deionized (DI) water to 200 ml. After that 12 ml H_2O_2 was introduced. Then the solution was centrifuged

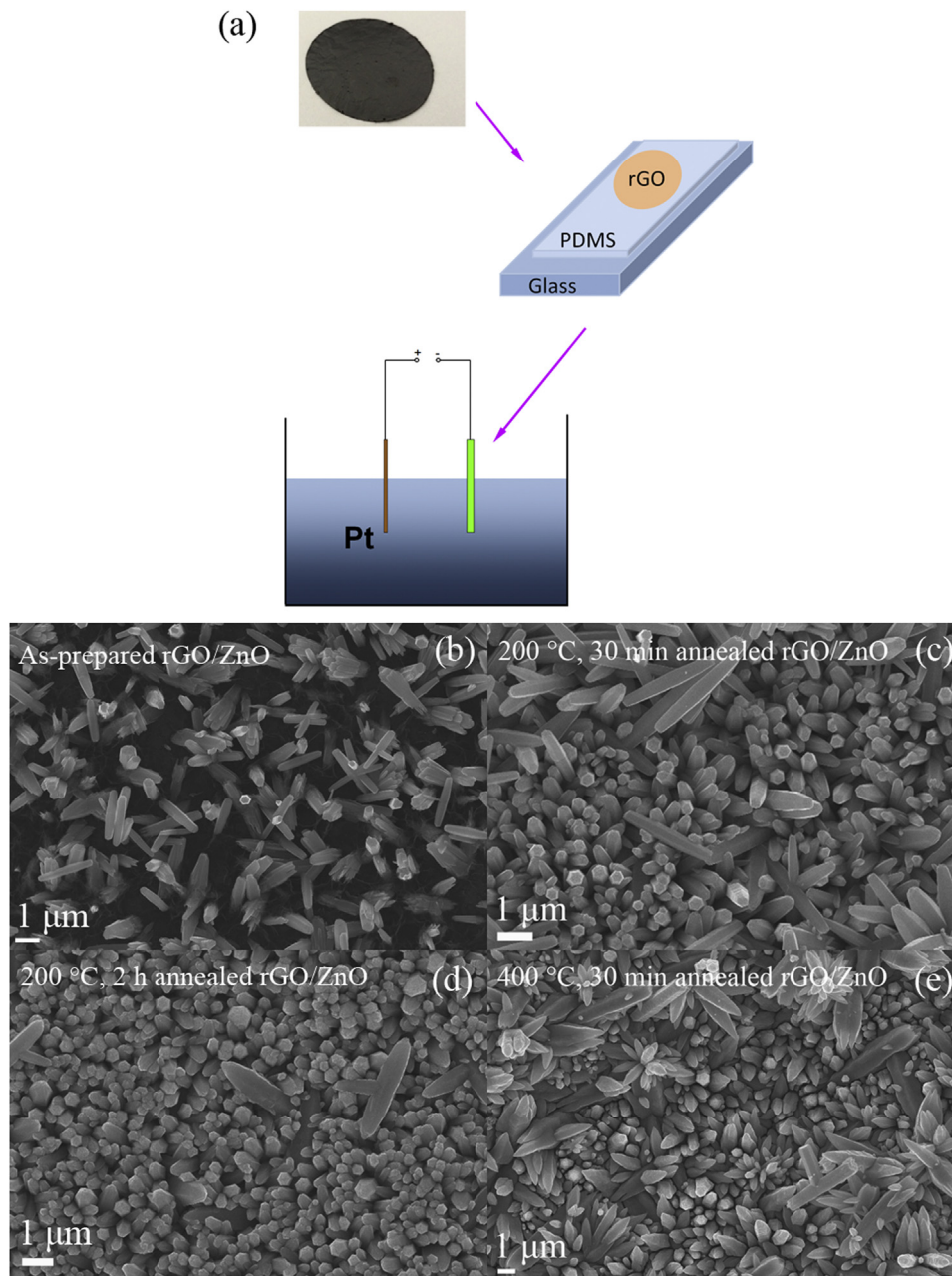


Fig. 1. (a) Schematic of the assembly of rGO substrate and the electrodeposit; SEM images of rGO/ZnO heterostructure i.e. ZnO nanorods on (b) as-prepared; and (c) 200 °C for 30 min.; (d) 200 °C for 2 h; (e) 400 °C for 30 min. annealed rGO substrates.

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