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Simultaneous Alkali Metal Doping and Thermal Annealing for the Purpose of Engineering the Electrical and Optical Properties of Graphene Oxide

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Abstract

In order to extend the application of graphene oxide (GO) in the area of electronic industries, enhancing the electrical properties of GO as a cost-effective alternative for graphene seems mandatory. Engineering the electrical properties of GO can be achieved in two different approaches: the oxygen functional group reduction and doping GO with chemical dopants. Here, both approaches were utilized to tune the electrical properties of GO toward its application as cathode [1-15]. First, GO was doped with alkali metal dopants, and later, the doped samples were thermally reduced. Energy-dispersive X-ray spectroscopy (EDX) and X-ray photoelectron spectroscopy were utilized to study the chemical composition of the doped samples. The even distribution of the dopants on the GO surface presented via the EDX elemental map, with no sign of particle development. After doping GO with alkali metals followed by thermal reduction, the sheet resistance of the doped samples was decreased from 311.0 kΩ/sq to as low as 32.1 kΩ/sq. Moreover, the optical properties of GO were effectively engineered via the different doping agents. The ultra-violet photoelectron spectroscopy showed that the shift of the work function of GO was as high as 1.74 eV, after doping followed by thermal reduction.

Introduction

In order to extend the application of graphene oxide (GO) in the area of electronic industries, enhancing the electrical properties of GO as a cost-effective alternative for graphene seems mandatory. Engineering the electrical properties of GO can be achieved in two different approaches the oxygen functional group reduction and doping GO with chemical dopants. Here, both approaches were utilized to tune the electrical properties of GO toward its application as cathode; first, GO was doped with alkali metal dopants, and later, the doped samples were thermally reduced. Energy-dispersive X-ray spectroscopy (EDX) and X-ray photoelectron spectroscopy were utilized to study the chemical composition of the doped samples.

Subjective Heading

After the dilemmas regarding the costly methods of producing graphene, difficulties of transferring methods, and small scale of production, graphene oxide was introduced as a low-cost two-dimensional alternative that could fit well into the different application demands via variable tuning techniques. Easy and cost-effective producing methods of GO and its hydrophilic characteristics introduced it as a relevant material that can efficiently be utilized as a coating or thin film on an industrial scale via the conventional water-based deposition techniques. utilized a spray coating approach for depositing a large-scale film of GO for use in electronic devices. used spin coating as a simple solution-based method for depositing GO on silicon oxide substrate as an active material for molecular sensors. deposited GO on an aluminum surface via electrophoretic deposition to take advantage of the chemical stability of the coating and its corrosion barrier characteristics in saline solutions.

Discussion

Among the different techniques of the oxygen functional group reduction, the thermal reduction has been attracted attention since there is no need for toxic chemicals or any advanced facilities The effectiveness of thermal reduction on the oxygen functional groups removal and improving the electrical properties of GO via thermal reduction was already investigated via the different research Wang et al. investigated the thermally reduced GO in solid-state dye-sensitized solar cells as window electrodes, which they could achieve a high conductivity (550 S/cm). Becerril et al. reduced a GO thin film via different reduction techniques, which among them the thermal reduction, was more promising and resulted in the rGO thin film with the sheet resistance of 10^2–10^3 Ω/sq. They claimed that the solution-processed rGO films had the potential to be used as transparent electrodes. Chen et alreported a highly conductive, free-standing rGO film reduced via thermal reduction of GO film, which, based on their observations, the size of the sp^2 domains were decreased due to the thermal reduction.reported a gentle and direct thermal treatment of GO under argon or H_2 atmosphere for producing conductive rGO. Based on their experience, thermal annealing was successfully reduced the different oxygen functional groups of GO, resulted in recovering the sp^2 network structures properly, and enhanced the conductivity of rGO (8100 S/m). Therefore, thermal annealing was introduced as a favorable approach for enhancing the electrical properties of GO.

The oxygen functional group reduction of GO is not the only approach for altering the electrical properties of this material. Naghdi et al. reviewed the different approaches to modify the electrical characteristics of GO, which among the different modification techniques, one method is doping the GO layers with chemical agents. Chemical doping was approved to be a significant factor to affect the electrical properties of the graphene family The chemical dopants such as n-doping agents (electron donors), were proven to increase the electron concentration in both graphene and GO structure and

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reduce its work function While the p-dopants (electron acceptors) were presented as a factor for decreasing the charge carrier density, increasing its work function and prepare it for application as anode or hole injection electrodes in applications such as organic thin film transistors

They showed that the n-doped graphene was a great candidate as cathode for organic light-emitting diodes. Yao used Bphen:Cs_2CO_3 as the n-doping agent and reduced the work function of graphene from 4.5 eV to 4.3 eV for its application as cathode for molecular organic light-emitting diodes. Therefore, it can be concluded that both oxygen functional group reduction and chemical doping of GO layers must be taken into account for producing low sheet resistance and low work function materials for application as a cathode in the electrical and optoelectronic applications.

In this work, the combination of these methods on the electrical and optical properties of GO is investigated. First, the GO layers were doped with the alkali metal dopants, and later, the doped samples were thermally reduced. The chemical composition of the samples was investigated using EDX and XPS techniques. The effect of the dopants and thermal reduction on the morphological characteristics of the samples was investigated using the HR-SEM and OM images. XPS and Raman spectroscopy clarified the type of dopants. The effect of the dopants and thermal reduction on the optical and electrical properties of the samples was investigated thoroughly in the last section of this work. Two-dimensional (2D) metal oxides and chalcogenides (MOs & MCs) have been regarded as a new class of promising electro- and photocatalysts for many important chemical reactions such as hydrogen evolution reaction, CO_2 reduction reaction and N_2 reduction reaction in virtue of their outstanding physicochemical properties. However, pristine 2D MOs & MCs generally show the relatively poor catalytic performances due to the low electrical conductivity, few active sites and fast charge recombination. Therefore, considerable efforts have been devoted to engineering 2D MOs & MCs by rational structural design and chemical modification to further improve the catalytic activities. Herein, we comprehensively review the recent advances for engineering technologies of 2D MOs & MCs, which are mainly focused on the intercalation, doping, defects creation, facet design and compositing with functional materials. Meanwhile, the relationship between morphological, physicochemical, electronic, and optical properties of 2D MOs & MCs and their electro- and photocatalytic performances is also systematically discussed. Finally, we further give the prospect and challenge of the field and possible future research directions, aiming to inspire more research for achieving high-performance 2D MOs & MCs catalysts in energy storage and conversion fields.

X-ray photoelectron spectroscopy (XPS)

To further investigate the doped-GO samples and analyze their elemental composition, XPS was utilized before and after thermal annealing. The XPS survey spectra of the samples before and after thermal annealing is presented in and the detail information are presented in , which provides useful evidence about the atomic composition and carbon to oxygen (C/O) ratios of the samples. The survey spectra of the doped samples have pronounced alkali metal peaks, indicating successful doping of GO. From the atomic composition results presented in , it can be concluded that the ratio of carbon to oxygen (C/O) of the GO was increased after alkali metal doping that was in accordance with the results presented for EDX The suggested results were due to partial reduction of the GO and also the functional groups of GO were replaced via the alkali metal elements

Raman

To recognize the number of graphene layers, presence of defects and disorders, and the effect of doping and thermal reduction on the graphene structure, Raman spectroscopy as a non-destructive method can be useful. To identify GO via Raman technique, two prominent Raman features of GO, D band at 1350 cm^{-1} (the defects or edges) and G band at about 1584 cm^{-1} (the first order scattering of the E_{2g} mode) are beneficial. Here the GO layers were doped with alkali metals and later were thermally reduced. Therefore, by studying the Raman pattern of the samples, first, we identify the success of the doping process and its effect on the structure of GO regarding the defects. Moreover, we investigate the effect of thermal reduction on the doped samples.

Environmental crisis together with the continued growth in energy demand has stimulated the rapid development of catalysis in environmental and energy applications Over the past decades, tremendous efforts have been devoted to fabricating novel catalytic materials for energy production and environmental remediation. Among them, hydrogen evolution reaction (HER), CO_2 reduction reaction (CO_2RR) and N_2 reduction reaction (NRR) have been recently extensively investigated for tackling global energy and environmental issues. Up to date, two kinds of main catalysis techniques have been used for promoting the above-mentioned chemical conversions: electro- and photocatalysis. Traditionally, thermal catalysis rely on high temperature or pressure to achieve the molecular activation and reactions . In comparison, the electro- and photocatalytic reactions happen at a relatively mild condition. This review mainly focuses on the sustainable and emerging electro- and photocatalytic catalysis, providing an alternative to thermal catalysis

On the other hand, photoelectrochemical (PEC) cells are demonstrated to be efficient routes for solar to electrochemical energy transformation, in which two electrodes serving as cathode and anoe, aqueous electrolyte, and an external circuit are needed to induce incoming photons generating electronic charge carriers under bias PEC routes work on the principle of not only harvesting sunlight but employing applied potentials as well. Similar to that in photocatalysis, photo-electrocatalysts with proper bandages are indispensable for sufficient absorption of the solar spectrum to initiate the catalytic reactions.Favourable photo-electrocatalysts should achieve both efficient photocarrier generator and high charge/carrier mobility, but these requirements are typically incompatible in one single material Therefore, adopting a co-catalyst is one typical route for efficient photogenerated charge accumulation and transportation, as well as providing sufficient active sites for catalytic reactions, similar with in photocatalysis . Photo-electrocatalysis is beyond the scope of this review paper. Therefore, we mainly focus on electro- and photocatalysis in the following parts. There have been many reported catalytic materials such as metals, metal oxides, metal chalcogenides, phosphates, carbides, and Particularly, two-dimensional (2D) metal oxides and chalcogenides (MOs & MCs) have attracted more and more attention because of their unique properties such as planar morphology, catalytic edge effects and tunable band-gap energies, which have been widely investigated for electro- and photocatalytic applications. Such semiconductor-based electrocatalysts show interesting catalytic behaviours. Fundamental electrochemistry uses Schottky-analogue junction to model the kinetics, in which the charge transfer process across the electrode–electrolyte interface is affected by the band alignment between the semiconductor and the redox species classical electron transfertheories, the interface is regarded as fully active during electrochemical reactions It is hard to accumulate a sufficient carrier concentration to achieve high conductivity since

the Schottky-analogue junction tends to transform into the ohmic characteristic once the electrochemical reaction starts However, experimental results indicate the occurrence of both "active" and "inert" regions, and a mixed interface as certain edges and defective planes are with extremely high activity. Other emerging models such as the self-gating phenomenon explain the extremely high carrier accumulations in ultra-thin 2D semiconductor catalysis Ascribed to the electronic-conduction modulation, ultra-thin 2D semiconductors with intrinsically low charge carrier concentrations can obtain high surface conductance. For example, the current decays with the thickness of MoS_2, indicating a penetration depth of the surface conductance at 1% of the max current is ~25 nm . Meanwhile, n-type semiconductor catalysts (MX_2, M = Mo, W, Re, Sn; X = S, Se; ZnO and SnO_2) are turned on by a negative potential to be suitable for cathodic reactions such as HER and CO_2RR. On the contrary, p-type semiconductor catalysts such as Ni- and Co-based oxides/(oxy)hydrooxides, Cu_2O, CuO, Cu_2S, Mn_3O_4, WSe_2, and $WSe_{1.8}Te_{0.2}$, are suitable for the anodic reactions (i.e., OER). Bipolar semiconductors (e.g., Ta/Nb-doped MoS_2) can be utilized for both reactions since they can be turned on by both positive and negative potentials.

Sheet resistance

The sheet resistance of GO and alkali metal doped-GO samples was measured from three different points on the samples' surface in ambient conditions utilizing a four-point probe system. Based on the different researches, doping graphene with alkali metals (that are n-type dopants) slightly increase the sheet resistance of the graphene layers in comparison with the p-dopants that decrease the sheet resistance while in the case of GO, n-doping reduces the sheet resistance of the GO layers Additionally, the oxygen functional groups cover the GO layers, which by eliminating these groups, the sheet resistance of the rGO decreases dramatically.

Conclusion

As a cost-effective alternative for graphene in advanced optoelectronic and electronic applications, graphene oxide(GO) needs to be enhanced electrically. In the current project, the GO was doped with alkali metal elements and the doped samples were thermally reduced to improve their electrical properties for cathode application. The chemical composition of the samples before and after the doping process and thermal reduction were studied via EDX, XPS, and Raman. The results suggested the successful doping of the GO sample with alkali metal elements, the partial reduction of GO via doping, and the stability of the dopants after thermal annealing at high temperatures.

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Conflict of Interest

The authors declare that they are no conflict of interest.

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