



Eco-friendly one-pot synthesis of gold decorated reduced graphene oxide using beer as a reducing agent



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ABSTRACT

A facile, eco-friendly and economical approach was demonstrated for the synthesis of gold-decorated reduced graphene oxide nanocomposites (rGO–Au_{nano}) using beer as a reducing agent via a hydrothermal method. The phenolic compounds of beer play a key role in the reduction of graphene oxide and the gold precursor. The obtained rGO–Au_{nano} was characterized by X-ray diffraction, UV–vis absorption spectroscopy, electron microscopy, atomic force microscopy and the electrochemical impedance spectroscopy. Analysis revealed that the electron-transfer resistance of rGO–Au_{nano}/GCE was much lower than that of the GCE and GO/GCE. The proposed nanocomposites have excellent electrocatalytic properties for catalytic reduction of O₂ in solution.

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1. Introduction

The discovery of graphene by Andre Geim and Konstantin Novoselov in 2004 has led to great and sustained interest in graphene and its applications [1]. Graphene a single aromatic sheet of sp²-bonded carbon possesses unique electrical, optical, thermal, mechanical and catalytic properties which make it attractive for various applications. To fabricate functional graphene, the surface of graphene must be modified using reducing agents such as hydrazine and sodium borohydride. This results in the synthesis of reduced graphene oxide (rGO). The chemical action also allows exfoliation of the graphene thereby improving its properties [2,3]. However, strong chemical reducing agents are toxic and unstable. Several research groups have investigated the synthesis of rGO by employing an ecofriendly and non-toxic approach for large scale industrial production for various applications [4–6]. Considering the environmental hazards caused by the use of chemical reducing agents for the reduction and exfoliation of GO, several green

reducing agents have gained considerable research interest for the facile production of nanomaterials. For example, reducing sugar [7], protein bovine serum albumin [8], heparin [9], and poly (diallyldimethylammonium chloride) [10] have shown promising results in the reduction of graphene oxide (GO). Therefore, an effective, low-cost, non-toxic and an eco-friendly green reducing agent is needed in the graphene industry. Surprisingly, these green reducers also act as stabilizing agents which also help to improve the stability and dispersion of rGO in the liquid phase. One such green reducing agent is beer which is produced by the fermentation of barley or malt grains.

Beer is the third-most popular drink worldwide after water and tea. The major chemical constituents present in beer are maltose, dextrin, acetate, lactate, alanine, ethanol, and phenolic compounds [11]. Apart from its reducing property, beer is also known to possess antioxidant and antimicrobial properties due to its ability to reduce free radical formation and scavenge free radicals [12,13]. It has been reported that the sources of natural antioxidants in beer are phenolic compounds. The phenolic compounds in beer are biocompatible and mainly consist of syringic acid, coumaric acid, ferulic acid, protocatechuic acid, vanillic acid, caffeic acid, *p*-hydroxybenzoic acid, tannic acid, epicatechin and quercetin. These compounds are excellent antioxidant agents because they readily react with reactive oxygen species such as free radicals. Upon oxidation, the phenolic groups are converted to their corresponding quinone forms which

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are also biocompatible in nature [14]. Phenolic compounds (electron-donating substituents) have been previously used for the green synthesis of nanoparticles [15,16]. In recent years an increasing number of research reports have been published regarding the green synthesis of graphene decorated with novel metal nanoparticles such as Pt [17], Ag [18] and Au [19]. The decoration of graphene with these nanoparticles may lead to the synthesis of novel metal matrix composites with improved mechanical, electrical and thermal properties via cooperative interactions. In this paper, we demonstrate a simple facile and novel route to prepare rGO sheets decorated with gold nanoparticles in a beer-formulated aqueous media.

2. Materials and methods

All chemicals were of analytical reagent grade and were used without further purification. High purity graphite powder (particle size < 0.1 mm) was used in the experiment for the synthesis of graphene. Other chemical reagents like H_3PO_4 , H_2SO_4 , $KMnO_4$, HCl, KOH, KCl, $K_3[Fe(CN)_6]$, $K_4[Fe(CN)_6]$, ethanol (96%) and ammonium hydroxide (25%) were obtained from Fluka (Buchs, Switzerland). Barley beer (0% alcohol) of Tuborg brand (Germany) was obtained from local supermarket. Double distilled water was used throughout the experiments to avoid contamination.

3. Experimental

Graphite oxide was synthesized using graphite powder by the modified Hummer's method [20]. Briefly 5.0 g of graphite powder was dispersed in 120 mL of concentrated H_2SO_4 maintained at 0 °C under stirring. Then 15.0 g of $KMnO_4$ was added gradually to the mixture which was kept in an ice bath to ensure that the temperature remained around 0 °C. Next, the temperature was increased to 0 °C and the mixture was stirred for 30 min before being diluted gradually with 225 mL of deionized water. The mixture was re-diluted with 700 mL of deionized water and treated with 3% hydrogen peroxide. The colour of the mixture changed to yellow-brown during the drop-wise addition of H_2O_2 . The mixture was filtered and washed with an HCl solution (5%) and then repeatedly washed with water until a neutral pH of the filtrate was obtained. This solution was centrifuged at 3000 rpm for 10 min and then the filtrate was re-dispersed in water and centrifuged several times. Finally the dark brown GO powder was obtained after drying in a vacuum oven at 50 °C for a day. The graphite oxide was then exfoliated by ultrasonication. For this purpose GO powder dispersed in a known volume of water was subjected to ultrasonication for 60 min to yield a stable suspension of GO. The suspension was then centrifuged at 3000 rpm for 30 min to remove any aggregates remaining in the transparent light brown exfoliated GO suspension. Then 10 mL of beer and 100 μ L of $NaAuCl_4$ (0.01 mM) were added into 10 mL of the above solution and the resulting solution was mixed by ultrasonication for 30 min. The mixture was transferred into a Teflon-lined stainless steel autoclave and reacted at 90 °C for 5 h. The resulting rGO-Au_{nano} suspension was filtered and washed with a large amount of water. Finally a rGO-Au_{nano}-modified glassy carbon electrode (GCE) sensor was fabricated. The end surface of the GCE (i.d. = 3.0 mm Metrohm-Herisau, Switzerland) was polished on a synthetic cloth successively with 0.3, 0.1 and 0.05 μ m alumina slurries (Struers, Copenhagen, Denmark) to obtain a mirror finish and then was cleaned in a water-ethanol solution (1:1) under ultrasonication. 6 μ L of the resulting rGO-Au_{nano} solution (0.4 mg mL⁻¹) was dropped onto a GCE and allowed to dry in ambient air for 2 h. The fabricated biosensor was stored at 4 °C in a refrigerator when not in use.

4. Characterization

X-ray diffraction (XRD) spectra were obtained using a D8ADVANCE (Bruker, Germany) using Cu K α (1.5406 Å) radiation. UV-vis absorption spectra were recorded using a single beam Pharmacia UV-vis spectrophotometer (Ultrospec model 4000). The morphological characterization of the prepared composites was examined by SEM (Philips Instrument, Model XL-30-Eindhoven, The Netherlands), TEM (HITACHI H-8100 EM). The surface morphology of the products was characterized using a DME DualScope Scanner DS 95-200 (Herlev, Denmark) atomic force microscopy (AFM). Cyclic voltammetry was performed using an Autolab potentiostat-galvanostat model PGSTAT30 (Utrecht, The Netherlands) with a conventional three electrode set-up in which a bare glassy carbon electrode (GCE) or modified GCE an Ag|AgCl|KCl_{sat} and a platinum rod served as the working, reference and auxiliary electrodes, respectively. Electrochemical impedance spectroscopy (EIS) experiments were carried out using a Zahner Zennium workstation and the output signal was acquired with the Thales z (Zennium release) software. The frequency was ranged from 0.1 Hz to 100,000 Hz with the signal amplitude of 5 mV. The resistance of nanocomposites were measured by two point probe method. The measured total resistance of GO, rGO and rGO-Au_{nano} at 1 V was 0.001 k Ω , 0.6 k Ω and 1.7 k Ω , respectively.

5. Results and discussions

5.1. X-ray diffraction

Fig. 1 shows the XRD (D8 ADVANCE (Bruker, Germany) pattern of exfoliated GO obtained using Cu K α 1.5406 Å radiation. (a) The results show that GO exhibits a diffraction peak centered at a 2θ angle of 13.01° corresponding to the (002) plane with an interlayer spacing of 0.78 nm which indicates complete oxidation of graphite into the exfoliated GO [21]. After chemical reduction with beer, the diffraction peak signal of the exfoliated rGO at a 2θ angle of 13.01° disappears completely and a very broad peak signal around a 2θ angle of 23.0° is (b) observed. This indicates that most of the oxygen-based functional groups were removed due to the reduction process [22]. It should be noted that the diffraction peak signal in rGO is rather broad and significantly different from that of the (002) graphite signal (c) at 26.44° (JCPDF card # 12-0212). Also, the diffraction peaks of the cubic phase gold nanoparticles along with their Miller indices (JCPDF card # 03-0921) were

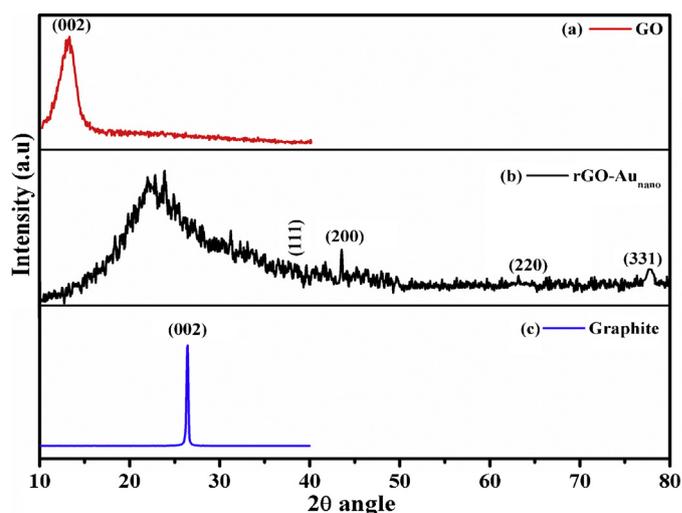


Fig. 1. Typical XRD patterns obtained for (a) exfoliated GO; (b) rGO-Au_{nano} and (c) pristine graphite respectively.

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