

Rational Design of Gold Nanoparticle/graphene Hybrids for Simultaneous Electrochemical Determination of Ascorbic Acid, Dopamine and Uric Acid



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Abstract: Gold nanoparticles (AuNPs) stabilized by citrate with different sizes (17, 28 and 46 nm) were assembled onto the poly(dimethyl diallyl ammonium chloride) (PDDA)-functionalized graphene nanosheets (GNS), respectively, and three kinds of Au/PDDA/GNS hybrids were thereby fabricated through electrostatic interaction. The structure, morphology and composition of the resulting hybrids were further characterized by transmission electron microscope (TEM), ultraviolet-visible (UV-vis) spectra, and X-ray photoelectron spectra (XPS) measurements. TEM characterization revealed that the AuNPs with the size of 17, 28 and 46 nm were densely dispersed onto the PDDA-functionalized GNS. Cyclic voltammetry (CV) and differential pulse voltammetry (DPV) measurements showed that the obtained Au/PDDA/GNS nanocomposites with high electrocatalytic activity could simultaneously determine ascorbic acid (AA), dopamine (DA) and uric acid (UA). The high sensitivity towards DA and UA determination was due to the electrocatalytic effect of Au/PDDA/GNS hybrids and the electrostatic attractions between Au/PDDA/GNS and DA, Au/PDDA/GNS and UA. The proposed sensor exhibited good selectivity and sensitivity, and could be applied to determine UA in human urine with satisfactory results.

Key Words: Graphene; Gold nanoparticle; Ascorbic acid; Dopamine; Uric acid

1 Introduction

Recently, numerous efforts have been devoted to the design and controllable fabrication of hybrid materials which integrate graphene nanosheets (GNS) with metal nanoparticles (Au, Pt, Pd), because of their remarkable catalytic properties^[1–4]. Generally, the fabrication of metal nanoparticle-decorated GNS hybrids was based on two approaches: grow of metal nanoparticles on the GNS surface through in-situ reduction reaction^[5,6], and mixing of chemically pre-synthesized metal nanoparticles with molecule functionalized graphene^[7,8]. However, in the first approach, the resulting hybrids normally aggregated in solution, leading to low loading and non-uniform dispersion of nanoparticles.

So the second approach is prevailing. To improve the properties of graphene, tailoring graphene through covalent and non-covalent interactions is demonstrated as an effective strategy. For example, linear positively charged poly(diallyldimethylammonium chloride) (PDDA) have been used to prevent graphene from aggregation via noncovalent functionalization. Due to the positively charged property of PDDA, the PDDA-functionalized GNS is supposed to be beneficial for interacting with the negatively charged metal nanoparticles through electrostatic interaction. For example, negatively charged gold nanoparticles (AuNPs) can be self-assembled onto the PDDA-functionalized graphene with high uniformity to form the Au/PDDA/graphene hybrid used in the catalytic systems, such as electrochemical sensors, fuel

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cells, and disease detection, and so on^[9–11]. Despite the successful synthesis of Au/PDDA/graphene hybrids, the loading amount of AuNPs in these hybrids is usually uncontrollable, which is supposed to be unable to get the insights of the related reaction mechanism. Therefore, developing effective and simple routes to fabricate Au/PDDA/graphene hybrids with controllable amount of AuNPs is still challenging.

It is well known that ascorbic acid (AA), dopamine (DA) and uric acid (UA) are important biomolecules in body fluids such as blood and urine, and play an important role in the metabolic system of human bodies^[12,13]. Therefore, simultaneous determination of these compounds is particularly important. It is well known that the oxidation peaks of AA, DA and UA appear at almost the same potential on conventional bare electrode, which results in overlapping voltammetric responses, and thus makes their discrimination highly difficult. So far, various modified electrodes have been developed in simultaneous determination of AA, DA, and UA, including carbon-based materials modified electrode^[14–16], noble metal/alloy nanoparticles modified electrodes^[17,18], metal oxides modified electrode^[19,20], etc. Due to graphene's remarkable electric properties, graphene-based materials have been fabricated and used to determine AA, DA and UA in ternary mixture^[21–23]. Recently, chitosan-graphene^[24] and graphene-Au nanocomposite-modified electrode^[25] were also applied to selectively determine DA, UA and AA. Yu *et al.*^[26] prepared the PDDA/graphene-modified carbon paste electrode to detect UA in the presence of AA and DA. However, it is still one of the attractive subjects to develop new materials for the simultaneous determination of DA, UA and AA.

In this work, AuNPs with different sizes (17, 28 and 46 nm) were assembled onto PDDA functionalized GNS to form Au/PDDA/GNS hybrids through strong electrostatic interaction. The obtained hybrids were further used for the simultaneous determination of AA, DA and UA.

2 Experimental

2.1 Instruments and reagents

PDDA (20% (w/w) in water), DA and UA were obtained from Sigma. AA was purchased from Fluka. Graphite powder (325 mesh), NaH₂PO₄, Na₂HPO₄, NaOH and H₃PO₄ were purchased from Beijing Chemical Factory (Beijing, China). Chloroauric acid (HAuCl₄) was purchased from Shanghai Chemicals, China. All chemicals were used as received without further purification. Phosphate buffer solution (PBS, 0.05 M) was prepared with Na₂HPO₄ and NaH₂PO₄. Aqueous solutions were prepared with ultrapure water (> 18 MΩ cm) from a Millipore system.

Ultraviolet-visible (UV-vis) absorption spectra were carried out on a U-3900 HITACHI UV-vis spectrophotometer,

whereas Raman spectra measurements were performed using a LabRam HR-800 confocal Raman microscope with a 514-nm laser as light source. Hitachi-600 transmission electron microscope with an accelerating voltage of 100 kV was employed to inspect the morphology of samples. X-ray photoelectron spectroscopy (XPS) analysis was performed on an ESCALAB-MKII X-ray photoelectron spectrometer (VG Co.) with Al Kα X-ray radiation as the X-ray source for excitation. Cyclic voltammetric (CV) and differential pulse voltammetric (DPV) measurements were performed by using a CHI920C electrochemical workstation (CHI) and conventional three-electrode system comprising a glassy carbon (GCE, *d* = 3 mm) or modified glassy carbon working electrode, a platinum wire as the auxiliary electrode, and an Ag/AgCl (in 3M KCl) as reference electrode. Before use, the GCE surface was carefully polished to a mirror using alumina slurries with different diameter (1.0, 0.3 and 0.05 mm), successively.

2.2 Preparation of graphene oxide (GO) and PDDA-functionalized GO

GO was synthesized from graphite by a modified Hummers method^[27]. As-prepared GO was dispersed in ultrapure water to give a brown suspension (0.01%, w/w), which was further subjected to dialysis against water to completely remove residual salts and acids for 7 days. Then the suspension was exfoliated for 1 h by ultrasonication and finally the bulk GO powders were transformed into GO nanosheets.

Approximately 0.1 mL of PDDA (35%, w/w) together with 35 mg of NaCl was added into 12 mL of GO (0.5 mg mL⁻¹) aqueous solution. The mixed solution was then further sonicated for 30 min at room temperature. After that, the PDDA functionalized GO was centrifuged and washed by ultrapure water for three times to remove excessive PDDA and NaCl. Finally, the resulted PDDA/GO NS was re-dispersed into 12 mL of water for further use.

2.3 Synthesis of different sizes of AuNPs

AuNPs was synthesized by the classic Frens' method^[28–29]. Typically, different volume (5 mL for 17 nm AuNPs, 1.5 mL for 28 nm AuNPs, and 1.0 mL for 46 nm AuNPs) of sodium citrate aqueous solution (1 %, w/v) was injected into boiling HAuCl₄ aqueous solution (100 mL, 0.01%, w/w) under vigorously stirring. The mixed solution was further refluxed for 30 min to complete the reaction. After that, additional sodium citrate (the total amount of sodium citrate added was kept at 15 mL for all three different sizes of AuNPs) was introduced to make sure the surface of AuNPs had sufficient negative charge for the efficient assembly with PDDA/GO NS.

2.4 Self-assembly of AuNPs with PDDA/GNS

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