



# Poly(ionic liquids) functionalized polypyrrole/graphene oxide nanosheets for electrochemical sensor to detect dopamine in the presence of ascorbic acid

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## ABSTRACT

Novel poly(ionic liquids) functionalized polypyrrole/graphene oxide nanosheets (PILs/PPy/GO) were prepared by the polymerization of 1-vinyl-3-ethylimidazole bromide (VEIB) on the surface of N-vinyl imidazolium modified PPy/GO nanosheets. Due to the synergistic effects of GO with well-defined lamellar structures, conductive PPy and biocompatible PILs, PILs/PPy/GO modified glassy carbon electrode (GCE) presented the excellent electrochemical catalytic activity towards dopamine (DA) with good stability, high sensitivity and wide linear range in the presence of ascorbic acid (AA) with high concentration. PILs played an essential role for the simultaneous determination of DA and AA in a mixture, whose existence effectively improved the transmission mode of electrons and resulted in the different electrocatalytic performance towards the oxidation of DA and AA. It is indicated that PILs/PPy/GO nanosheets can act as a good steady and sensitive electrode material for the development of improved DA sensors.

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## 1. Introduction

Thin film with two-dimensional (2D) nanostructures has been considered as a kind of promising modified electrode material with good stability and excellent sensitivity for the application to build electrochemical sensors and biosensors, due to its good environmental stability, thinner thickness and large surface area (Janáky and Visy, 2013; Jiang et al., 2004; Pang et al., 2000; Zhang et al., 2014). Graphene is a monolayer nanosheet composed of sp<sup>2</sup>-hybridized carbon atoms arranged in hexagonal honeycomb lattice, which is well known as the thinnest nanomaterial in the world and has attracted tremendous attentions due to its exceptional thermal mechanical, and electrical properties with promising applications (such as developing an ultra-high-resolution electrochemical biosensor with single-DNA resolutions (Akhavan et al., 2012) and the electrochemical detection of leukemia (Akhavan et al., 2014a; Akhavan et al., 2014b)), since it was separated from graphite (Geim, 2009; Geim and Novoselov, 2007; Novoselov et al., 2004). Recently, novel composite film materials

composed of conducting polymers (CPs) and graphene (G) or graphene oxide (GO) have attracted a tremendous amount of attentions and become a research focus, because they possess both excellent properties of conducting polymers and graphene or graphene oxide, such as high electric conductivity at room temperature, long term environmental stability, good electrochemical activity and biocompatibility (Guiseppe-Elie, 2010; Lee et al., 2006) of CPs, as well as unique electrical and chemical properties of G or GO (Kuila et al., 2011; Liu et al., 2012b). Therefore, CPs/G or CPs/GO nanocomposite materials can be applied in many fields such as energy storage, supercapacitors or electrochemical sensors and biosensors for the detection of certain special substances, for instance, polyaniline/graphene (PANI/G) (Gómez et al., 2011) and polypyrrole/graphene oxide (PPy/GO) (Zhu et al., 2012) exhibited good electrochemical properties and cycling performance, which should be promisingly used for the fabrication of inexpensive, high-performance electrochemical supercapacitors; poly(3,4-ethylenedioxythiophene)/graphene oxide (PEDOT/GO) nanocomposite modified electrode exhibited lowered impedance and increased charge storage capacity as well as improved sensitivity to the oxidation of dopamine (DA) in the presence of ascorbic acid (AA) and uric acid (UA) (Weaver et al., 2014). However, most CPs/G or CPs/GO nanocomposites were prepared by electrochemical deposition (Chang et al., 2012; Si et al., 2011; Zhu et al., 2012)

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which limited their high-volume production. CPs/G or CPs/GO nanocomposites synthesized by chemical precipitation technique or liquid/liquid interfacial polymerization were often used for supercapacitors (Bora and Dolui, 2012; Gómez et al., 2011), but rarely for electrochemical sensors and biosensors due to the hydrophobicity and poor dispersibility of the pristine CPs on the surface of electrode.

Poly(ionic liquids) (PILs) have attracted great interest during the past few years, because that they combined the unique properties of ionic liquids (ILs) (such as low flammability and toxicity, enhanced ionic conductivity, high polarity, good chemical stability, etc.) (Holbrey and Seddon, 1999) with the intrinsic polymer properties and presented the advantages of both ionic liquids and polymers (Marcilla et al., 2006), which can be applied in various fields. For instance, glucose oxidase adsorbed on PILs-Au nanoparticle composites exhibited direct electron transfer and bioelectrocatalytic properties towards the oxidation of glucose (Lee et al., 2012); Ag nanoparticles-PIL-graphene sheets were utilized to fabricate a nonenzymatic hydrogen peroxide sensor (Wang and Yun, 2013), PILs functionalized graphene (PILs-G) with the immobilized glucose oxidase (GOD) modified electrode exhibited excellent direct electrochemical response for glucose with good sensitivity and wide linear range (Zhang et al., 2011), and so on. Therefore, PILs as a modifier may contribute more excellent performance to electrode materials for electrochemical sensors and biosensors.

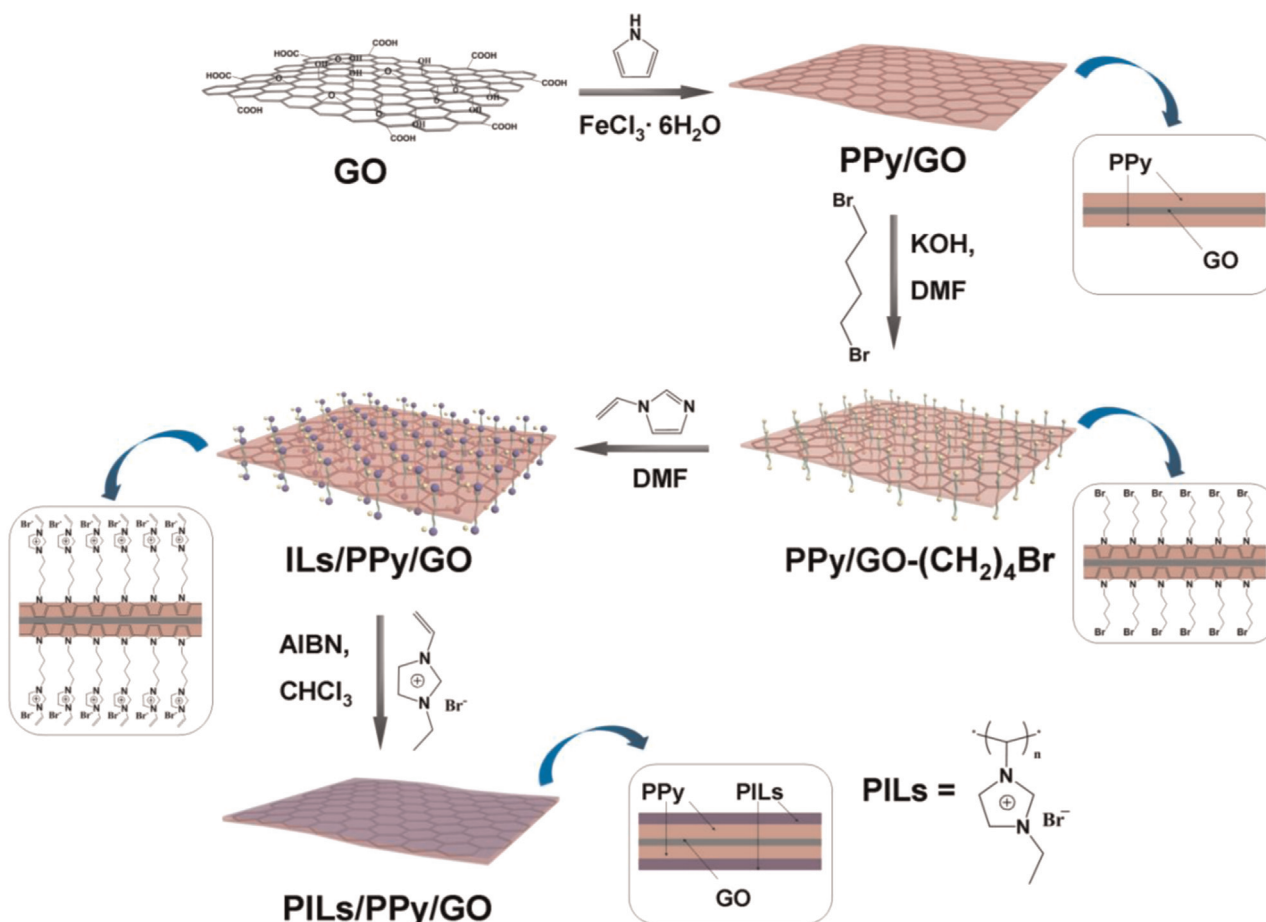
Herein, novel ternary composites, PILs/PPy/GO nanosheets, which combined the advantages of GO, PPy and PILs, were firstly prepared by the polymerization of 1-vinyl-3-ethylimidazolium bromide (VEIB) on the surface of N-vinyl imidazolium modified

PPy/GO nanosheets (Scheme 1). Due to the synergistic effects of GO with well-defined lamellar structures, conductive PPy and biocompatible PILs, PILs/PPy/GO modified glassy carbon electrode (GCE) presented the excellent electrochemical catalytic activities towards dopamine (DA) with good stability, high sensitivity and wide linear range in the presence of ascorbic acid (AA) of high concentration. The PILs played an essential role for the simultaneous determination of DA and AA in the mixture, whose existence obviously changed their surface charge property to electropositivity, resulting in the significant improvement of PILs/PPy/GO dispersibility in aqueous solution, transmission mode of electrons and different electrocatalytic performance towards the oxidation of DA and AA. These research results indicated that PILs/PPy/GO can act as a steady and sensitive electrode material, especially for detecting of dopamine in the presence of ascorbic acid.

## 2. Material and methods

### 2.1. Material

Pyrrrole (Py) (Sinopharm Chemical Reagent Co. Ltd.,  $\geq 98.0\%$ ) and Bromoethane (Sinopharm Chemical Reagent Co. Ltd.,  $\geq 98.0\%$ ) are chemical grade. N-vinyl imidazole was purchased from Yancheng Medical Chemical Factory (China), which were distilled under vacuum before use. High-purity graphite powder was purchased from Sinopharm Chemical Reagent Co. Ltd.. All the other reagents were analytical grade, and used without further purification, including  $\text{KMnO}_4$  (Tianjin Baishi Chemical Co. Ltd,



Scheme 1. The reaction procedure for the preparation of PILs/PPy/GO nanosheets.

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