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Regular Article

Pt nanoparticles supported on nitrogen-doped porous graphene for sensitive detection of Tadalafil

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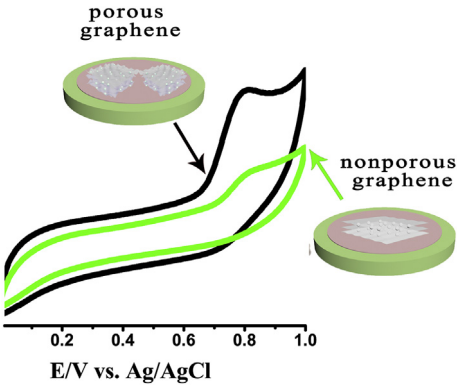
GRAPHICAL ABSTRACT



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ABSTRACT

Graphene (GR) is one of the most promising candidates for utilization in the electroanalytical field because of its superior electrocatalytic activity, excellent electronic conductivity, and high chemical stability. However, the GR sheets usually tend to stack together with π-π interaction. The spontaneous stacking leads to the aggregation of the GR sheets and imposes a negative feedback in the surface area of the GR, which obviously limits its electrochemical application. In this study, nitrogen-doped porous GR (NPGR) with different pore sizes is prepared by using silica (SiO_2) as a template. The NPGR exhibits high surface area and porous structure, fulfilling the requirement for supporting materials. Being a support, the structural uniqueness and N dopants of NPGR facilitate the deposition of Pt nanoparticles (Pt NPs). The Pt NPs/NPGR composites integrate the structural properties of NPGR and catalytic properties of Pt NPs. A selective and sensitive electrochemical sensor was successfully developed for sensitive determination of Tadalafil (TAD), showing a concentration range of 1.30–488.9 μM and limit of detection of 0.268 μM.

1. Introduction

Graphene (GR), a single graphite layer with sp^2-hybridized carbon, is considered as one of the most promising materials that have

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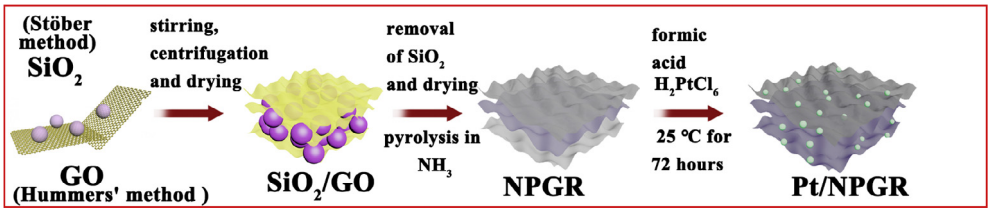
attractive applications in many research areas because of its high surface area, high conductivity, excellent thermal and chemical stability, and low cost [1]. To extend its application, some heteroatoms, such as nitrogen (N), boron, sulfur, fluorine, and phosphorous, are generally doped into GR to improve the electrocatalytic properties of GR [2–6], which is attributable to the rich edge plane like-sites/defects effectively induced by introduction of heteroatoms. Additionally, by supporting of nanoparticles (NPs) on GR surface, the electrochemical activity of NPs can be dramatically improved because of the synergistic interaction between the NPs and GR. For example, NiCo [7] and Pt NPs [8,9] supported GR are capable for catalyzing the oxidation of methanol with high performance. However, the major weakness of the GR is the inescapable aggregation owing to the strong π–π stacking propensity among the nanosheets [2,10–12]. The irreversible stacking leads to the partial aggregation of GR sheets, thus seriously affecting its surface area and electrochemical behavior. Because of the irreversible aggregation, NPs supported GR layers are easily secluded inside the GR layers and the catalytic efficiency of supported NPs is restrained. Hence, it is of significant importance to decrease the aggregation of GR layers and increase the surface area of GR. These efforts are centered in intercalation of nanostructured carbon materials, modification of GR and porous GR (PGR) [13–16]. With regard of PGR, the porous structure and the high surface area of PGR favor the availability of the electrocatalytic sites and in turn effectively enhance the catalytic activity of the PGR-based electrochemical sensors [12,17–25]. Because of large surface area and structural uniqueness of PGR, NPs supported on PGR are not hidden by GR layers and efficiently exposed to the targets of interest [10,18,26–29]. These electrochemical biosensors or sensors based on PGR are widely investigated in several analytical fields, such as environment monitor, disease diagnosis and food security [3,4,14,30–32].

Tadalafil (TAD) is a PDE5 inhibitor used in pill form for treating erectile dysfunction [33,34]. Sensitive detection of TAD is of great importance for the pharmaceutical analysis of drug release characteristics and routine quality control monitoring [35]. In the literatures, several studies have been reported the TAD detection in biological and pharmaceutical samples. The most methods already published are high performance liquid chromatography (HPLC) with ultraviolet detection [36–38], gas chromatography with mass spectrometry (MS) [39], mass spectrometry [40], fluorescence [41], capillary electrophoresis [42], spectrofluorometry [33], and spectrophotometry[43,44]. Although they provide good precision and sensitivity [34], these techniques have some disadvantage, such as need of expensive equipment and time-consuming extraction procedures. Electrochemical method has many advantages, such as simplicity, cheap cost, simplicity, and relatively short time for analysis compared with other techniques, in addition to high selectivity and sensitivity, which makes it an attractive choice for pharmaceutical analysis [45]. There are few studies of electrochemical determination of TAD. For example, Li's group reported the determination of TAD at the *p*-sulfonated calix[6]arene (SCX6)/reduced graphene oxide (RGO) in glassy carbon electrode (SCX6@RGO/GC)



Scheme 1. Preparation pathway of Pt/NPGR-*X*.

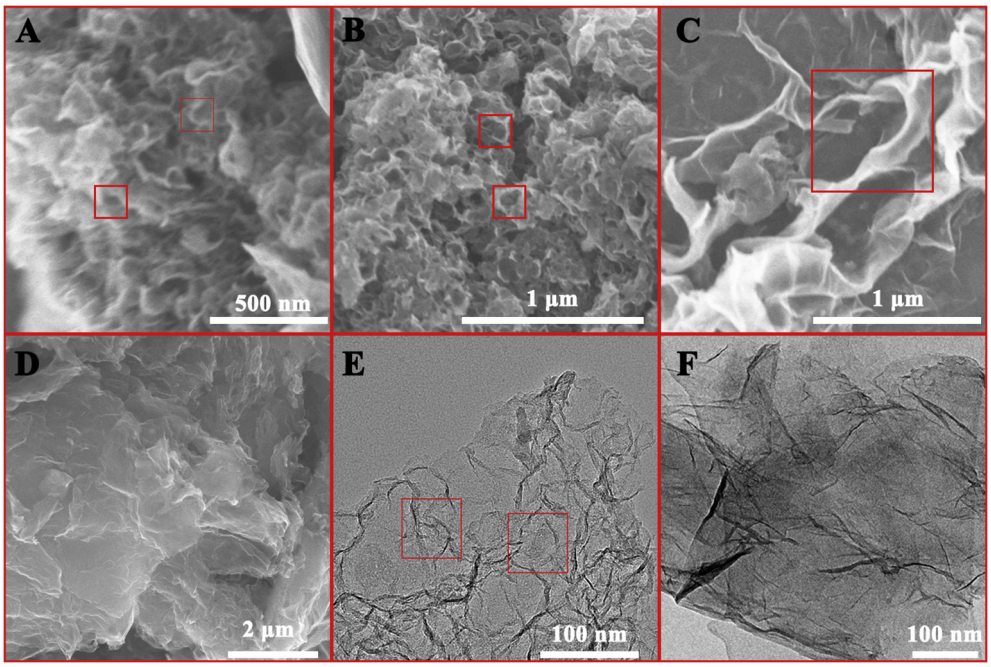


Fig. 1. SEM images of (A) NPGR-120, (B) NPGR-200, and (C) NPGR-310. TEM images of (E) NPGR-120 and (F) GR.