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Hierarchical and hybrid RGO/ZIF-8 nanocomposite as electrochemical sensor for ultrasensitive determination of dopamine

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ABSTRACT

A hierarchical and hybrid nanocomposite of reduced graphene oxide/zeolitic imidazolate framework-8 (RGO/ZIF-8) has been successfully fabricated by the RGO as template for the in situ growth of ZIF-8. The morphology and structure of the resulted RGO/ZIF-8 nanocomposite were characterized by using scanning electron microscopy (SEM) and X-ray diffraction (XRD). It was demonstrated that ZIF-8 nanoparticles anchored and grew well on the surfaces of RGO. The electrocatalytic activity of the RGO/ZIF-8 towards the dopamine (DA) was investigated using cyclic voltammetry (CV) and differential pulse voltammetry (DPV). The results demonstrated the fabricated RGO/ZIF-8 sensor exhibited high sensitivity for the determination of DA, owing to the synergistic effect from highly electrical conductivity of RGO and porosity of ZIF-8. Under the optimal conditions, the linear response range for DA was from 1.0×10^{-7} to 1.0×10^{-4} mol L^{-1} and the detection limit of DA was estimated to be as low as 3×10^{-8} mol L^{-1}. Moreover, the prepared electrochemical sensor also showed excellent selectivity for DA detection in the presence of ascorbic acid (AA) and prospective results in real samples detection.

1. Introduction

Dopamine (DA) is one of the most effective neurotransmitter in mammalian central nervous systems, which mediates the transmission of messages between brain and neurons [1,2]. An abnormal dopaminergic neuron process is directly related to motive functions of the central nervous system which may lead to the neurological illnesses. Therefore, the rapid, accurate and sensitive determination of DA is important in clinical analysis. Various methods have been established for the sensitive detection of DA [3,4], such as liquid chromatography, capillary electrophoresis, chemiluminescence, fluorescence and colorimetric methods. Electrochemical method has been applied in the determination of DA due to its higher accuracy, faster response, lower instrumental and operation expenses [5,6]. However, ascorbic acid (AA) usually coexists with DA in the central nervous system and both of them are oxidized at the nearly same potential. Therefore, many kinds of nanomaterials have been applied to fabricate novel modified electrodes for the selective and sensitive determination of DA [7–10].

Metal-organic frameworks (MOFs) are a new class of crystalline porous materials which are constructed by connecting metal ions with organic links. Compared with other porous materials, MOFs have unique properties including of great surface area, stable chemical properties, and adjustable pore sizes and functionalities, which have attracted enormous interests. Zeolitic imidazolate frameworks (ZIFs), an attractive subclass of MOFs, exhibit the facility of synthesis, great thermal, hydrothermal and chemical stabilities compared with most of other MOFs [11–13]. They have shown great potential in gas storage, separation, chemical sensing, drug delivery and catalysis [14,15]. Recently, ZIF-8, which is a kind of ZIFs built from zinc ions and 2-methylimidazolate, has emerged as one kind of promising materials for constructing electrochemical sensors. For example, Ma et al. [16] investigated the first application of ZIF material which acted as a matrix for developing electrochemical biosensors. However, poor conductivity of ZIF-8 largely restricts their application in the electrochemical sensing area [17]. To enhance the electrical conductivity, the construction of conductive MOFs-based composites by introducing excellent conductive and electroactive materials into MOFs is an effective method. Various conducting materials such as metal nanoparticles [18,19] and conductive polymer [16] have been introduced into MOFs-based electrochemical sensors. For example, Hosseini et al. [19] have prepared Au-SH-SiO_2 nanoparticles supported on metal-organic framework (Au-SH-SiO2@Cu-MOF) and established a highly active and excellent

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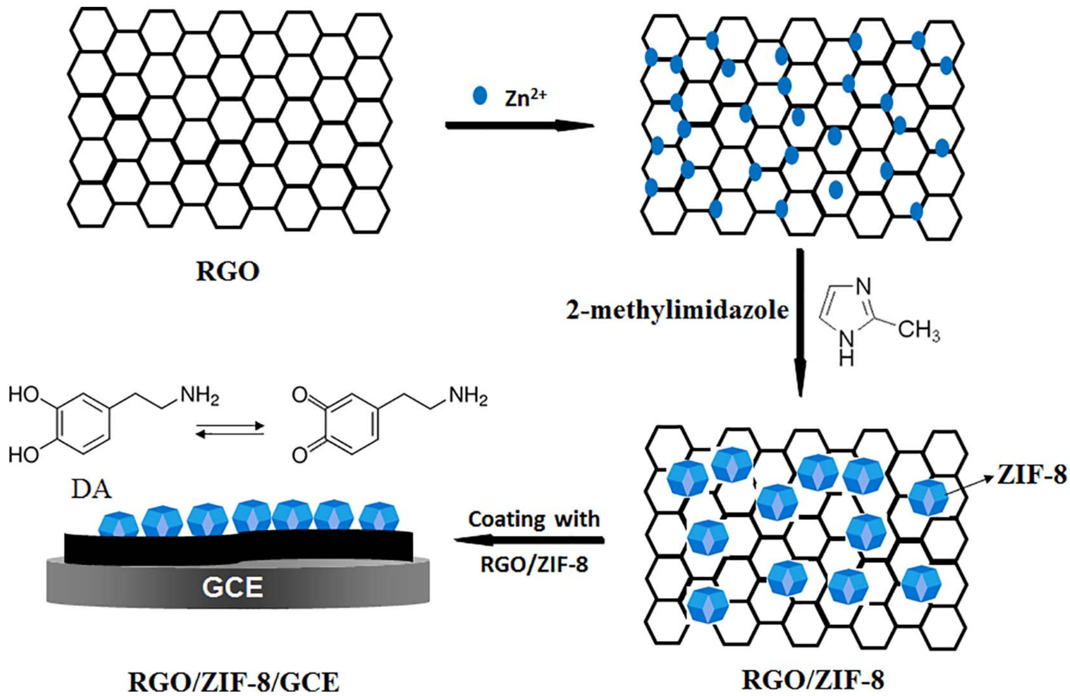

Scheme 1. Schematic diagram of the preparation procedure of the RGO/ZIF-8 nanocomposite and RGO/ZIF-8/GCE.

selective sensor for the determination of L-cysteine.

Among the conducting materials, graphene is one of the most popularly and promising candidate as modified material for electrochemical analysis due to its superior electronic conductivity, exceptional electron transfer rate, excellent chemical, thermal and mechanical stability [20–23]. Wang et al. have prepared PDA/ZIF-8@rGO composites for the immobilization of glucose oxidase to determinate glucose [24]. The introduction of graphene accelerates the electron transfer between the composites and the electrode and improves the electrochemical detection sensitivity. However, the MOF/graphene composites were prepared using physical blend method in that report, which was not stable and effective for electron transfer. Recently, Kim et al. have achieved the preparation of graphene/ZIF-8 nanocomposites by in situ growing ZIF-8 on graphene [25]. Due to the synergistic effect of graphene and ZIF-8 crystals, the researchers obtained the fast mass transfer and high electrical conductivity. These excellent properties made the graphene/ZIF-8 nanocomposite become a candidate as modified material for electrochemical sensing. A large scale of researches on the preparation and electrocatalysis application of graphene-ZIF nanocomposites have been reported in recent years [26,27]. However, there are few reports about applying graphene/ZIF nanocomposites to the electroanalytical field.

In this work, we have prepared a hierarchical and hybrid nanocomposite of RGO/ZIF-8 (reduced graphene oxide/zeolitic imidazolate framework-8) by the RGO-templated growth of ZIF-8. The morphology and structure of the resulted RGO/ZIF-8 nanocomposite were characterized by using scanning electron microscopy (SEM), X-ray diffraction (XRD). Then, the resulted RGO/ZIF-8 nanocomposite was cast on a glassy carbon electrode (GCE) for the fabrication of sensor which was used for electrochemical determination of DA. Owing to the synergistic effect from highly electrical conductivity of RGO and porosity of ZIF-8, the electrochemical performances for DA detection were significantly improved. And the RGO/ZIF-8/GCE was successfully applied to the fast determination of DA in human serum with satisfactory recoveries. Therefore, the RGO/ZIF-8-based sensors held great promise for electrochemical sensing.

2. Experimental

2.1. Chemicals and materials

Graphite was provided by Qingdao Fujin Graphite Co., Ltd. (Qingdao, China). $Zn(NO_3)_2{\cdot}6H_2O$, 2-methylimidazole (2-mim) were obtained from Aladdin Reagent Co., Ltd. (Shanghai, China). DA and AA were obtained from Shanghai Reagent Factory (Shanghai, China). HCl, NaOH and ethanol were obtained from Sinopharm Chemical Reagent Co., Ltd. Phosphate buffer solutions (PBS) with different pH values (from 4.0 to 8.0) were prepared by mixing 0.02 M NaCl, $NaH_2PO_4{\cdot}2H_2O$ and $Na_2HPO_4{\cdot}12H_2O$ and adjusting the pH by adding HCl or NaOH. All chemicals were of analytical grade and used without further purification. Doubly distilled water (DDW) was used throughout the experiments, and the experiments were performed at room temperature.

2.2. Apparatus

Scanning electron microscopy (SEM) images were obtained by JEOL JSM-7001F (Japan). X-ray diffraction (XRD) was operated on a Rigaku Ultima IV diffractometer (Japan). Cyclic voltammetry (CV) and differential pulse voltammetry (DPV) were performed on CHI-660C electrochemical workstation (Shanghai Chenhua Instrument Co., Ltd., China) with a conventional three-electrode system: an Ag/AgCl electrode (Ag/AgCl) was used as the reference electrode, a platinum wire was used as the auxiliary electrode, and the bare or modified glassy carbon electrode was used as the working electrode.

2.3. Preparation of ZIF-8 and RGO/ZIF-8 nanocomposite

ZIF-8 was synthesized in an aqueous system according to the previous literature [28]. First, 0.744 g of $Zn(NO_3)_2{\cdot}6H_2O$ and 12.3 g of 2-mim were dissolved in 10 mL and 90 mL of DDW respectively. The 2-mim solution was added dropwise into the $Zn(NO_3)_2$ solution with vigorous stirring and the mixture was stirred for 24 h. Then, the precipitate was collected by centrifugal separation at 6000 rpm for 10 min and wash with a mixture of ethanol and DDW for repeated three times. The product was dried at 60 °C for 48 h under vacuum to obtain ZIF-8.

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