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Amperometric detection of hydrazine utilizing synergistic action of prussian blue @ silver nanoparticles / graphite felt modified electrode

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ABSTRACT

In this study, a triple-component hydrazine sensor (PB@Ag/GF) was fabricated with freestanding graphite felt (GF), silver nanoparticles (Ag) and prussian blue (PB). The Ag nanoparticles were electrodeposited on GF ultrasonically (Ag/GF), and acted as a catalyst of the chemical deposition of PB. The electrode was characterized by scanning election microscopy (SEM), infrared spectroscopy (IR), X-ray diffraction (XRD), and energy-dispersive X-ray spectroscopy (EDS). The electrochemical behavior of PB@Ag/GF was measured by cyclic voltammetry and amperometric measurements. The sensor displayed a prominent electrocatalytic activity toward hydrazine oxidation, with a fast response time of 2 s, a low detection limit of 4.9×10^{-7} mol L^{-1} and very high detection sensitivity of 26.06 A mol^{-1} L.

1. Introduction

Hydrazine ($N_2H_4{\cdot}H_2O$) and its derivatives have wide applications in industry, agriculture, and can be used as explosives, antioxidants, photographic developer, oxygen scavengers and propellants [1], but they have also been recognized as carcinogenic and hepatotoxic substances, which could cause liver and kidney diseases, even cancer or genetic damages [2]. All these traits make their detection and quantization problems of considerable importance, notably in agriculture and pharmaceutical industry. The traditional methods reported for the detection of hydrazine are potentiometric [3], chemiluminescence [4], coulometric [5], and spectrophotometric methods [6], which are all expensive and require high demands on devices. Therefore, developing more convenient and cheaper methods for hydrazine detection is important. Based on these considerations, we have chosen to study carbon-based chemically modified electrodes with excellent electron transfer performance between the electrode and the electrolyte [7], to find a new way for hydrazine detection which is both convenient and less cost.

Prussian Blue (PB) and its analogues, which belong to a family of cyanide-based coordination network, have been used as electron-transfer mediators thanks to their outstanding electrocatalytic properties, with applications in many fields, such as ion selective electrodes [8], charge storage devices [9], catalysis [10], and biosensors [11]. The traditional synthetic methodology of Prussian Blue is to mix aqueous solutions of ferric (Fe^{3+}) and ferricyanide ($[Fe(CN)_6]^{3-}$) ions [12]. However the rate of the reaction is very low and the yield of PB nanoparticles is limited. In order to increase the reaction speed, Au and Pt nanoparticles were found to actively promote the process of PB growth [13,14], while the catalytic effect of Ag nanoparticles has not been examined yet.

In this study, graphite felt (GF) was selected as a support because of its cost-effectiveness, its high conductivity, its reasonable chemical stability, and its 3D structure with a high porosity, and thus a high surface area for its functionalization by PB growth [14,15]. To our knowledge, studies reporting GF as working electrode for application in hydrazine sensing has not been reported. Here, we report a novel and original sensor (PB@Ag/GF) for direct electroanalytical determination of hydrazine. The sensor electrode was built in two steps. Firstly, the Ag nanoparticles were deposited on the surface of GF using a simple potentiostatic method under ultrasons. Secondly, Prussian Blue was catalytically deposited on the nanoparticles from a mixture of $K_3[Fe(CN)_6]$,

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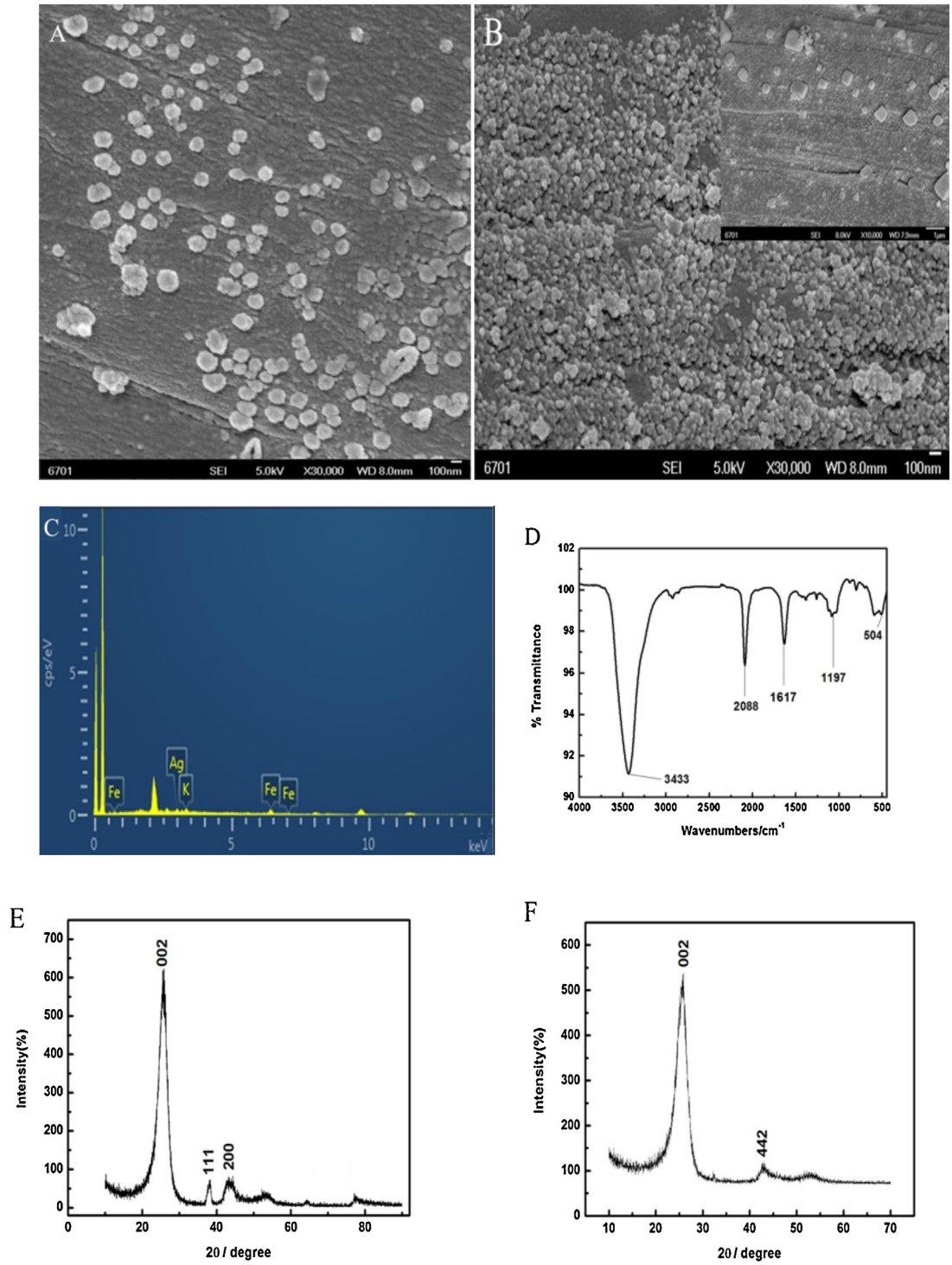

Fig. 1. SEM images of Ag/GF (A) and PB@Ag/GF (B). Inset: SEM image of PB/GF; EDS spectrum of PB@Ag/GF (C); FT-IR spectrum of PB@Ag/GF (D); XRD pattern of Ag/GF (E) and PB@Ag/GF (F).

$FeCl_3$, KCl and HCl, resulting in the formation of the PB@Ag/GF electrode. This electrode was characterized by various techniques and a complete electrochemical characterization was performed, using cyclic voltammetry (CV) and amperometry. Electrochemical studies showed that the sensor has a good performance for the determination of hydrazine, in terms of anodic current increment, regarding the oxidation of $N_2H_4 \cdot H_2O$.

2. Experimental

2.1. Chemicals and Apparatus

Polyacrylonitrile-based graphite was obtained from Haoshi carbon fiber Institute, without any further treatment. All other chemicals were obtained from Tianjin Guangfu Fine Chemical