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Electrochemical sensor for chloramphenicol based on novel multiwalled carbon nanotubes@molecularly imprinted polymer

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ABSTRACT

Herein, we present a novel electrochemical sensor for the determination of chloramphenicol (CAP), which is based on multiwalled carbon nanotubes@molecularly imprinted polymer (MWCNTs@MIP), mesoporous carbon (CKM-3) and three-dimensional porous graphene (P-r-GO). Firstly, 3-hexadecyl-1-vinylimidazolium chloride (C16VimCl) was synthetized by using 1-vinylimidazole and 1-chlorohexadecane as precursors. Then, C16VImCl was used to improve the dispersion of MWCNT and as monomer to prepare MIP on MWCNT surface to obtain MWCNTs@MIP. After that, the obtained MWCNTs@MIP was coated on the CKM-3 and P-r-GO modified glassy carbon electrode to construct an electrochemical sensor for the determination of CAP. The parameters concerning this assay strategy were carefully considered. Under the optimal conditions, the electrochemical sensor offered an excellent response for CAP. The linear response ranges were 5.0×10^{-9}–5×10^{-7} mol L^{-1} and 5.0×10^{-7}–4.0×10^{-6}, respectively, and the detection limit was 1.0×10^{-10} mol L^{-1}. The electrochemical sensor was applied to determine CAP in real samples with satisfactory results.

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1. Introduction

Chloramphenicol (CAP) is a broad-spectrum antibiotic. It is effective against a wide variety of gram-positive and gram-negative bacteria and is widely used for the treatment of infectious diseases in humans and animals (Falagas et al., 2008; Turton et al., 2002). However, it also has serious side-effects on human beings such as gray baby syndrome, leukemia, and aplastic anemia (Falagas et al., 2008; Turton et al., 2002). For this reason, many countries such as USA, Canada and China have banned the use of CAP in the food producing animals and set the maximum residue limit for food-safety control programs (Falagas et al., 2008). Thus, it is important to develop sensitive and selective methods for the determination of this compound in food production.

At present, several analytical techniques are used for the determination of CAP, including high-performance liquid chromatography (Chen et al., 2009), gas chromatography–mass spectrometry (Shen et al., 2009) and electrochemical technique (Borowiec et al., 2013; Pilehvar et al., 2012; Yadav et al., 2014; Zaidi 2013; Zhuang et al., 2014). Among them, electrochemical method offers the advantages of simplicity and low cost in comparison with other methods (Zaidi, 2013). Many electrodes were applied to the directly electrochemical determination of CAP, such as activated carbon fiber electrode (Agüí et al., 2002), singlewall carbon nanotube (SWCNT)–gold nanoparticle (GNP)–ionic liquid (Xiao et al., 2007) and nitrogen-doped graphene (r-GO) decorated with GNP (Borowiec et al., 2013) modified glassy carbon electrodes (GCEs), self-assembled monolayer modified gold electrode (Codognoto et al., 2010) and platinum electrode (Zhuang et al., 2014), but their selectivity and/or sensitivity were still not satisfactory. To solve this issue, some recognition receptors such as antibody (Chullasat et al., 2011; Liu et al., 2013; Zhang et al., 2011), aptamer (Pilehvar et al., 2012; Yadav et al., 2014; Yan et al., 2012) and molecular imprinted polymer (MIP) (Alizadeh et al., 2012; Zhao et al., 2012) were used to improve the sensing performance. MIP is cheap and stable in comparison with antibody and aptamer. However, the MIPs prepared with conventional methods for the determination of CAP had less effective binding sites as many binding sites were buried in polymeric matrix (Alizadeh et al., 2012; Zhao et al., 2012). To improve this situation, surface molecularly imprinting technique was developed (Chen et al., 2011; Cheong et al., 2013; Sharma et al., 2012).

Surface imprinted polymers have higher binding capacity, faster mass transfer and binding kinetics (Sharma et al., 2012, 2013; Yang et al., 2014). However, these advantages most depend on the supporting materials, which should possess large surface,

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good conductive property and easy functionalization. Multiwalled carbon nanotube (MWCNT) is an ideal supporting material based on these criterions (Chen et al., 2011; Sharma et al., 2013). Thus, it was widely applied for this purpose, but the functional monomer was firstly immobilized on MWCNT surface by using chemical bonding before the preparation of MWCNTs@MIP in most reports (Anirudhan and Alexander, 2014; Bali Prasad et al., 2013; Chi et al., 2012; Lee and Kim, 2009; Moreira et al., 2011; Prasad et al., 2010; Tong et al., 2013; Zhang et al., 2012; Zhao et al., 2014). Recently, Qian et al. reported the in situ chemical oxidative polymerization of pyrrole on MWCNT surface for the preparation of novel MWCNTs@MIP (Qian et al., 2014). Comparing with other methods, this one was simpler, but the obtained quantity of MIP was not enough as expected. Therefore, the facile preparation of MIPs on MWCNT surface is still in need of exploration.

Here, we synthetized 3-hexadecyl-1-vinylimidazolium chloride (C16VimCl), which possessed double functional groups for the preparation of MWCMTs@MIP. C16VimCl could improve the dispersion of MWCNT and act as monomer to form MWCNTs@MIP. Then, the obtained MWCNTs@MIP was coated on the mesoporous carbon (CKM-3) and three-dimensional porous graphene (P-r-GO) modified GCE to construct an electrochemical sensor for the determination of CAP. In this strategy, C16VimCl facilitated the preparation of MWCNTs@MIP, and the CKM-3 and P-r-GO made the sensor more sensitive. To the best of our knowledge, this is the first time to use such a functional monomer to prepare MWCNTs@MIP for the determination of CAP.

2. Experimental

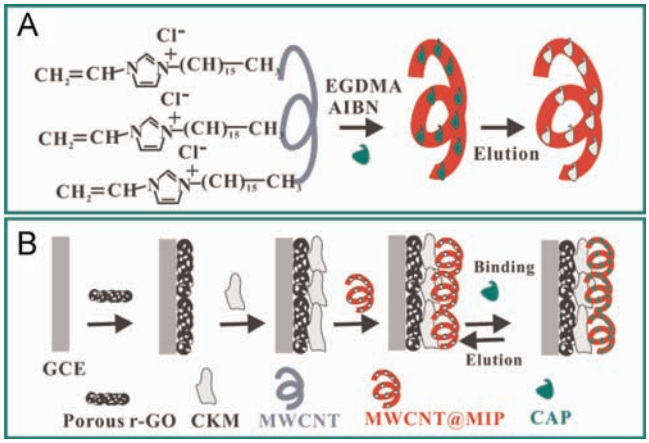
2.1. Apparatus and reagents

All electrochemical experiments were performed with a CHI 660D electrochemistry workstation (Shanghai CH Instruments Co., China). A three-electrode system was used, consisting of a modified glassy carbon electrode as working electrode, a saturated calomel electrode (SCE) as reference electrode and a platinum foil as counter electrode. Scanning electron microscope (SEM) was performed using a Zeiss (German) with an accelerating voltage of 10 kV. Transmission electron microscopy (TEM) was carried out by using a JEM-2100 transmission electron microscope with an accelerating voltage of 200 kV. ^{1}H NMR spectra were recorded with a Varian Mercury 400 spectrometer and the solvent used was deuterated chloroform ($CDCl_3$). Fourier transform infrared (FTIR) absorption spectra were recorded with a model Nexus-670 spectrometer (Nicolet, USA).

Carboxylic acid-functionalized multiwalled carbon nanotube (MWCNT) and mesoporous carbon (CKM-3) were supplied by Nanjing XFNANO Materials Tech Co., Ltd. (Nanjing, China). Chloramphenicol (CAP), thiamphenicol, florfenicol, 1-vinylimidazole, 1-chlorohexadecane, 2.2-azobisisobutyronitrile (AIBN), ethylene glycol dimethacrylate (EGDMA) were purchased from Sigma (St. Louis, MO, USA). Unless stated otherwise, other reagents used were analytical grade. The milk samples and honey samples were purchased from local supermarket. The support electrolyte was 0.1 mol L^{-1} phosphate buffer solution (PBS, pH=7.0), which was prepared with NaH_2PO_4 and Na_2HPO_4.

2.2. Synthesis of C16VimCl

C16VimCl was synthesized by referring the previous report about the synthesis of 3-hexyl-1-vinylimidazolium bromide (Jana et al., 2013). In a typical reaction, 6.03 mL (i.e. 20 mmol) 1-chlorohexadecane and 1.81 mL (i.e. 20 mmol) 1-vinylimidazole were added to 10 mL dry tetrahydrofuran and let them react for 48 h at 60 °C. The resulting crude product was purified by recrystallization with ethyl acetate for at least three times, giving the desired product as a white powder in 70.0% yield. ^{1}H NMR ($CDCl_3$, δ): 0.94 (t, 3H, $-CH_3$), 1.10–1.41 (m, 26H, $13CH_2$), 1.94 (t, 2H, CH_2), 4.40 (t, 2H, N–CH_2), 5.45 (d, 1H, vinyl CH_2), 5.96 (d, 1H, vinyl CH_2), 7.40 (t, 1H, vinyl CH), 7.60 (s, 1H, imidazole N–CH), 8.02 (s, 1H, imidazole CH–N^+), 11.20 (s, reduced intensity, imidazole N–CH–N^+).



Scheme 1. Schematic diagram of the construction procedure of sensor.

2.3. Preparation of MWCNTs@MIP

The preparation of MWCNTs@MIP was shown in Scheme 1 (Part A). Firstly, 0.1136 g (i.e. 4.00 mmol) C16VimCl and 40 mg MWCNTs were dispersed into 40 mL methanol/water (4/1, V/V)) by ultrasonication for 30 min to obtain a homogeneous suspension. Then, 0.0323 g (i.e. 1.00 mmol) CAP was completely dissolved into above suspension and let them prepolymerize for 60 min. After that, 10 mg AIBN and 3.1 mL (i.e. 20.00 mmol) EGDMA were dispersed into above solution. The resultant mixture was stirred at 60 °C for 24 h under nitrogen protection. Finally, the product was eluted by methanol/acetic acid (9/1, V/V) until no template molecule was detected in the washing solutions. For comparison, MWCNTs@NIP was prepared by the same manner except the absence of CAP. At the same time, other MWCNTs@MIPs were prepared by changing the mole ratio of CAP to C16VimCl (i.e. 1:2, 1:3, 1:5 and 1:6), the mole ratio of CAP to EGDMA (i.e. 1:15, 1:18, 1:22 and 1:25) and the mass ratio of MWCNT to C16VimCl (1:2, 1:2.5, 1:3 and 1:3.2), respectively.

2.4. Preparation of porous r-GO

Graphene oxide (GO) was prepared by a modified Hummers method (Liang et al., 2009) and characterized as our previous report (Yang et al., 2014). According to the report (Chen and Qiao 2013), porous GO (P-GO) was obtained by oxidizing and etching GO with $KMnO_4$ and HCl, but we lengthened oxidation time to 12 h in order to acquire bigger pore and better three-dimensional structure. Specifically, 100 mL of 0.5 mg mL^{-1} GO suspension was mixed with 0.5 g $KMnO_4$ under magnetic stirring for 12 h. The above solution was merged with 30 mL HCl (36%, wt%) and 30 mL H_2O_2 (30%, wt%) for 3 h. After that, the products were separated by centrifugation, washed with water and dried in a vacuum oven at 60 °C.

Next, 200 mL of 0.3 mg mL^{-1} P-GO suspension was mixed with 2.4 mL ammonia (28%, wt%) and 0.24 mL hydrazine hydrate (85%, wt%), followed by heating at 95 °C for 12 h. Then, the products were separated by centrifugation, washed with water and dried under a vacuum oven at 60 °C.

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