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Modified glassy carbon electrode with Polydopamine-multiwalled carbon nanotubes for simultaneous electrochemical determination of biocompounds in biological fluids

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ABSTRACT

In this study, a simple modified glassy carbon electrode with Polydopamine-multiwalled carbon nanotubes (GCE/PDA-MWCNTs) is described for selective and sensitive simultaneous voltammetric determination of dopamine (DA), acetaminophen (AC), and xanthine (XN). A detailed investigation by field emission scanning electron microscopy, Fourier transform infrared spectroscopy and electrochemistry methods such as cyclic voltammetry (CV), electrochemical impedance spectroscopy, differential pulse voltammetry (DPV) and chronoamperometry are performed in order to elucidate the preparation process and properties of the GCE/ PDA-MWCNTs. The proposed modified electrode displays intense and indelible electrooxidation response for simultaneous determination of DA, AC, and XN to three well-separated peaks in the potential range from 0.0 to 0.8 V/Ag/AgCl using CV and DPV methods in phosphate buffer solution with pH 7.0. Under the optimum conditions, detection limits of 20, 30 and 50 nM were obtained for DA, AC and XN, respectively. Moreover, GCE/PDA-MWCNTs was successfully used for simultaneous determination of DA, AC and XN in real samples.

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1. Introduction

Dopamine (DA) is in mammalian central nervous system and acts as a hormone as well as a neurotransmitter [1]. Low levels of DA may lead to Parkinson [2]. Also, its low levels in urinary excretion indicate chronic renal parenchymal disease [3]. Acetaminophen (AC) is a popular anti-fever and pain drug [4], high levels of which cause liver damages [5]. Nearly 5% of AC in urinary eliminates remains unchanged [6]. Xanthine (XN) is a product on the pathway of purine degradation. High levels of XN in human body fluids point to xanthinuria, a pathological state [7]. These analytes usually coexist in biological fluids [8], therefore simultaneous determination of these analytes is important for inspection of their physiological functions and disease diagnosing.

Polydopamine (PDA) is a mussel-inspired adhesive polymer which was synthesized for the first time by Lee et al. in 2007 [9]. PDA is mainly prepared by spontaneous oxidative polymerization of dopamine in weak alkaline conditions [10]. Dopamine is stable toward oxidation in acidic media but this oxidative polymerization can be catalyzed in the mild acidic media in the presence of transition metal ions [11]. Moreover, the spontaneous oxidative polymerization of dopamine can be performed in strong acidic conditions by using the hydrothermal method [12]. Another route for PDA synthesis is electropolymerization of dopamine [13]. This multifunctional polymer has many interesting properties which increase its application in many research areas such as battery, fuel cell, solar cell, drug delivery and biosensors. PDA forms an adhesive thin film on many surfaces such metals, metal oxides, ceramics and organic substrates [9]. The PDA coating mechanism is not fully revealed but extant evidence suggests that PDA adheres to metal and metal oxides via co-ordination bonds [14,15]; also, due to having catechol groups in its structure, it is able to adhere to organic surfaces via Michael addition or Schiff base reaction [16]. PDA was used for immobilization of metal and metal oxide nanoparticles on various substrates [17,18] as well as for functionalization of many surfaces via Michael addition or Schiff base reaction with amine and thiol functional groups of organic molecules [19]. PDA forms complexes with metal ions; this chelating ability has been used to remove metal ions from aqueous solutions [20]. PDA has numerous applications in electrode modification. For example, silver nanoparticle- PDA-graphene composite was used for DNA electrochemical bio sensing [21]. Cui et al. reported the application of PDA capsules for intracellular anticancer drug delivery [22]. Nam

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et al. used PDA as sensitizer in dye-sensitized solar cell [23]. Ryou et al. prepared a PDA modified composite separator for high-power lithium-ion battery [24].

Carbon nanotubes are carbonic nanostructures with unique properties such as large surface area, good thermal and mechanical stability as well as good electrochemical conductivity. These unique properties have inspired researchers to use this material as excellent support to prepare composites for development of sensors and biosensors [25]. Yongxin li et al prepared some Nano composites such as overoxidized polypyrrole directed single wall carbon nanotubes and Single-stranded deoxyribonucleic acid-wrapped single-walled carbon nanotubes to modify GCE for simultaneous electrochemical determination of biomolecules [25,26]. However, in this work, we use the easy, one-step dopamine self-polymerization technique to generate a thin layer of PDA on MWCNTs. To the best of our knowledge, for the first time these PDA-coated MWCNTs were used to modify the glassy carbon electrode (GCE) for sensitive simultaneous electrochemical determination of DA, AC and XN in human's urine and blood serum samples.

2. Experimental

2.1. Reagents

Multiwall carbon nanotubes (MWCNTs), with diameters, OD = 20–30 nm, wall thickness = 1–2 nm, length = 0.5–2 μm and purity > 95%, Dopamine (DA) acetaminophen (AC), and Xanthine (XN) were purchased from Sigma-Aldrich Company. Sodium hydroxide and phosphoric acid were purchased from Merck Company. Chitosan (CH) with medium molecular weight, 400,000 Da, was purchased from Fluka Company. Fresh urine and blood serum samples were obtained from the Tohid Clinical Laboratory (Zahedan, Iran) without any pretreatments.

2.2. Instrumentation

Electrochemical measurements were carried out with a SAMA 500 Electro analyzer (SAMA Research Center, Iran) controlled by a personal computer. All electrochemical experiments were carried out in a conventional three-electrode cell at room temperature. A platinum electrode and a silver/silver chloride electrode (Ag/AgCl) were used as the counter and reference electrodes, respectively. FESEM images were taken using a Philips XL30 Scanning electron microscopy. Electrochemical impedance spectroscopy (ESI) was performed with an Autolab PGSTAT 128 N (EcoChemie, Netherlands) potentiostat/galvanostat controlled by NOVA 1.11 software. Electrochemical impedance measurements were performed in 5 mM $[Fe(CN)_6]^{3-/4-}$ prepared in 0.1 M KCl. EIS was performed over a frequency range of 10 kHz to 0.1 Hz with 10 mV amplitude and it is superimposed on the formal potential of the redox probe ($E0 \cong E1/2 = (Epa + Epc)/2$, when Dox = Dred) which calculated from cyclic voltammograms. A Metrohm pH meter, model 744 was also used for pH measurements.

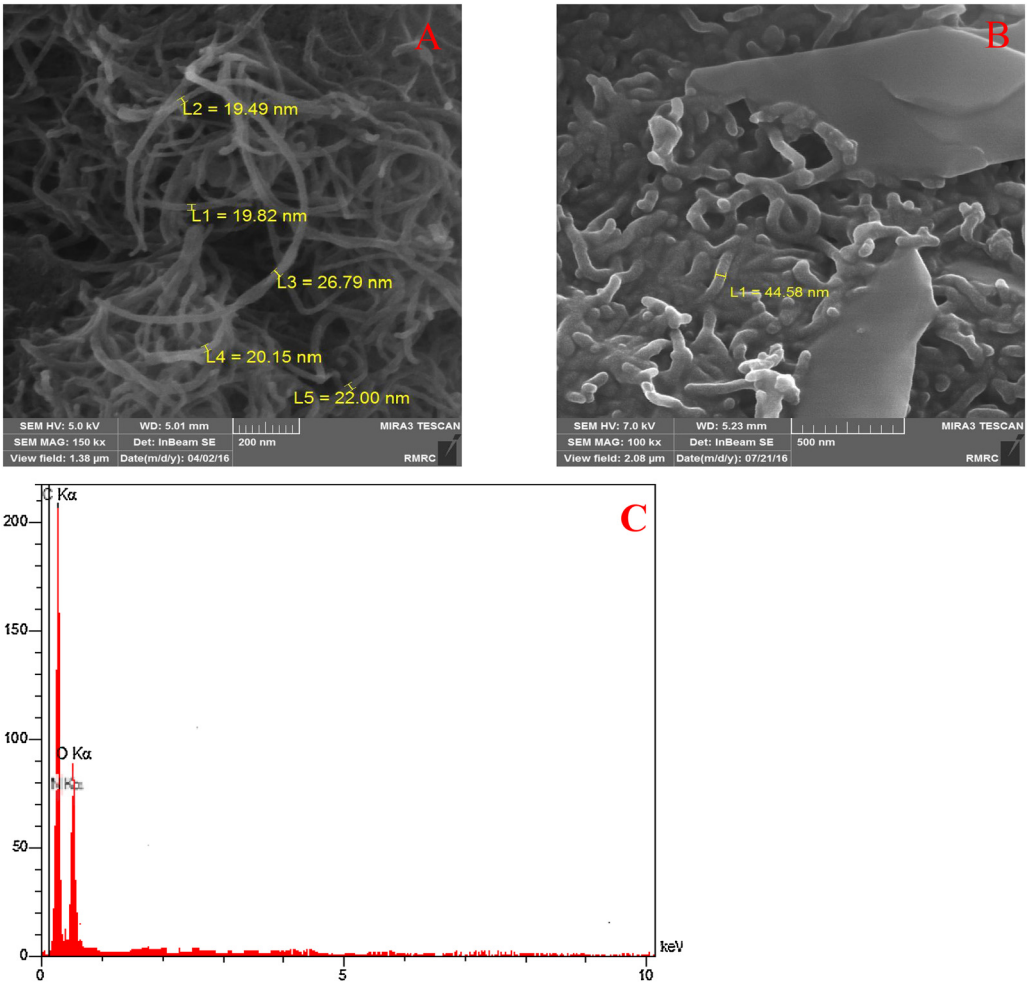

Fig. 1. FESEM of MWCNTs (A) and PDA - MWCNTs composite (B). EDX spectrum of PDA - MWCNTs composite (C).

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