



Nano composite system based on fullerene-functionalized carbon nanotubes for simultaneous determination of levodopa and acetaminophen



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ABSTRACT

In this paper we report an electrochemical sensor based on fullerene (C₆₀)-functionalized carbon nanotubes composite for the sensitive determination of the levodopa (LD) and acetaminophen (AC). A modified electrode by fullerene-functionalized carbon nanotubes (CNTs) composite has been fabricated and its electrochemical behavior was investigated by cyclic voltammetry, chronoamperometry and differential pulse voltammograms. The modified electrode shows electrocatalytic activity toward LD oxidation in phosphate buffer solution (pH 7.0) with a reduction of the overpotential of about 270 mV and an increase in peak current. These conditions are sufficient to allow determination of LD and AC in the samples at fullerene-functionalized carbon nanotubes composite. The differential pulse voltammetry data showed that the obtained anodic peak currents were linearly dependent on the LD concentrations in the range of 0.5–2000 μM, with the detection limit of 0.035 μM. The prepared electrode was successfully applied for the determination of LD and AC in real samples.

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1. Introduction

Carbon based nanomaterial are extensively used in electrochemistry, particularly in modification of electrodes due to their low cost in comparison to metal nanoparticles, relative high surface area, good conductance, favorable electronic properties and electrocatalytic effect make them adequate for the construction of electrode [1–3]. Carbon based nanomaterial can be formed from a variety of graphite products such as carbon nanotubes (CNTs), carbon fibers, fullerene, graphene and highly ordered paralytic graphite. CNTs continue to receive considerable attention in electrochemistry [4–6]. CNTs have been the subject of numerous studies in chemical, physical, and materials areas due to their novel structural, mechanical, electronic, and chemical properties [7]. Chemical and physical functionalization endows CNTs with diverse functions and paves the way for their future applications in different fields [8–10]. C₆₀ appears to be attractive to functionalization of CNTs because it is chemically stable, metallic impurity free and relative simple to implement and gives rise to reproducible electrocatalytic

responses [11]. Electrostatic effect C₆₀ have not been widely accepted [12,13]. Compton et al. [14] have shown that the observed electrocatalysis in some cases were caused by either the small amount of graphite impurity in C₆₀ sample, or the oxygenated species formed on the surface of glassy carbon electrodes by “electrode pretreatment”. As an excellent electron acceptor, C₆₀ could be dispersed in nanomaterial, such as CNTs, to facilitate electron transfer. Recently, we show that fullerene-functionalized carbon nanotubes/ionic liquid film modified electrodes offer electrocatalytic effect in respect catecholamines oxidation [15]. We propose that the CNTs surface is best described as porous materials; the smaller blocks allow more material to diffuse around them on the timescale of the voltammetric experiment and undergo electrolysis at the exposed electrode surface. Furthermore, porous C₆₀-CNTs film also exhibits some degree of thin-layer behaviors [15].

The unusual amino acid levodopa (3,4-dihydroxyphenylalanine, LD), is the precursor required by the brain to produce dopamine, a neurotransmitter. LD is widely used as a source of dopamine in the treatment of most patients with Parkinson's disease and epilepsy [16,17]. The most patients with Parkinson's disease are very old, at least older than 34 years, who receive levodopa as an anti-parkinson drug. With its serious side effects with long-term use

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on human health, e.g. gastritis, paranoia, and dyskinesia [18], LD should be given an accurate analysis in both pharmaceutical formulations and biological fluids. Furthermore, Parkinson's disease patients chronically use opiates, acetaminophen and drugs treating neuropathic pain such as antidepressants and antiepileptics. Acetaminophen (AC) is categorized as pain relievers and fever reducers. When used antipyretic, analgesic drug and an effective agent that used worldwide for the reduction of bacterial fevers and also it used for the relief of mild to moderate pain associated with headaches, backaches, arthritis and postoperative pain [19]. AC is metabolized in the liver where it generates toxic metabolites. According to above sentences, simultaneous determination of LD and AC is quite necessary in the diagnosis and treatment of diseases. However, LD and AC have an overlapping oxidation potential on the bare electrodes and AC shows serious interference in determination of LD [20,21]. It is vital to control the amount of LD and AC in real samples to gain lower toxicity and better medicinal effect. So it is essential to develop simple and fast methods for their determination in routine analysis. Among many methods for determination of LD and AC in biological samples, electrochemical methods due to the advantages of relatively low cost, fast response, simple instrumentation, high sensitivity, facile miniaturization, and low power requirement have shown to be a powerful tool [22–24].

In the present work, we have developed an electrochemical sensor based on the fullerene-functionalized carbon nanotubes (C_{60} -CNT/GCE) for the oxidation of LD. The catalytic effect of C_{60} -CNT/GCE was investigated for electro oxidation of LD. Furthermore, DPV was used to evaluate the analytical performance of LD in the presences of AC at C_{60} -CNT/GCE and good limit of detection was obtained by this sensor. Also the modified electrode was used successfully as an electrochemical sensor for simultaneous determination of LD and AC in real sample as a sensitive, selective, simple and rapid method.

2. Experimental

2.1. Apparatus and reagents

Electrochemical measurements were performed using a computerized potentiostat/galvanostat (SAMA 500, Iran). All electrochemical studies were performed with a three electrodes assembly including a saturated calomel electrode (SCE), platinum wire and C_{60} -CNT/GCE as a reference electrode, counter electrode and working electrode, respectively. A Metrohm 781 pH/ion meter was used for pH measurements. All solutions were freshly prepared with double-distilled water. LD, AC, fullerene and reagents were analytical grade from Aldrich. Pure graphite powders (Merck) and paraffin oil (DC 350, Merck) were used as binding agents for the graphite pastes. Multiwall carbon nanotubes were purchased from Plasma Chem. Phosphate buffer solutions (0.1 M) was prepared from 0.1 M H_3PO_4 - NaH_2PO_4 , and the pH was adjusted with 0.1 M H_3PO_4 or NaOH.

2.2. Preparation of the electrode

Purified MWCNTs and C_{60} (MWCNTs: C_{60} = 2:1) with a total amount of about 1.0 mg were dispersed in 10 mL toluene in an ultrasound bath for 30 min to give a 0.1 mg/mL suspension. Prior to the modification, a GCE was polished with alumina powder followed by rinsing thoroughly with water. Then 15 μ L of the suspension was directly cast on a GCE and the solvent was then dried. This C_{60} -MWCNTs film modified GCE was subjected to potential scanning in acetonitrile solution containing 0.1 M tetrabutylammonium hexafluorophosphate as supporting electrolyte between

0.0 and -2.0 V until reversible multistep electron transfer reaction was obtained. The resulting C_{60} -MWCNTs film electrode was then washed with acetonitrile several times to remove the electrolytes and dried in a stream of hot air, which was similar to our group's previous work [15].

3. Results and discussion

3.1. Characterization of the C_{60} -CNT composites

Fig. 1A shows the TEM image of C_{60} -CNT composite, which indicated that C_{60} -CNT composite was obtained with C_{60} deposited on CNTs. As the TEM image in Fig. 1A shows clearly, the C_{60} -CNT composite film comprises small nanoparticles of C_{60} uniformly dispersed on the CNTs. We also added the composites into toluene and subjected the suspension to ultrasonication, but the toluene solution did not turn purple, a characteristic color of C_{60} . These results indicate that C_{60} was been strongly bounded to CNTs.

3.2. Electrocatalytic oxidation of LD at C_{60} -CNT composite

Fig. 2 depicts the CV responses for the electrochemical oxidation of 0.5 mM LD at unmodified GCE (curve b), CNT modified GCE (CNT/GCE) (curve c) and C_{60} -CNT/GCE (curve d). As it is seen, while the anodic peak potential for LD oxidation at the CNT/GCE (curve c) and unmodified GCE (curve b) are 380 and 401 mV, respectively, the corresponding potential at C_{60} -CNT/GCE is ~ 110 mV. These results indicate that the peak potential for LD oxidation at the C_{60} -CNT/GC electrodes shift by ~ 270 and 291 mV toward negative values compared to CNT/GCE, and unmodified GCE, respectively. However, C_{60} -CNT/GCE shows much higher anodic peak current for the oxidation of LD compared to CNT/GCE, indicating that the combination of C_{60} deposited on carbon nanotubes has significantly improved the performance of the electrode toward LD oxidation. The presence of C_{60} on CNT surface promotes electron transfer between CNTs and C_{60} molecules in the C_{60} -CNT composite. Moreover, the change in the porosity of this electrode, combined with possible thin-layer effects can be used to explain the observed improvement on the electrochemical response in electroanalysis of LD [15]. In a thin layer effects, a planar working electrode and a planar insulating surface are separated by a small distance. A small volume of solution is trapped between these surfaces in a thin layer. In a potential sweep experiment, current is drawn as the electron transfer becomes kinetically and thermodynamically favorable. If the layer of solution becomes significantly depleted of the electroactive species on the experimental timescale then the current will be diminished. This can lead to peak separations considerably less than that expected by using the simple semi-infinite planar diffusion model. We propose that similar behavior will be observed at the C_{60} -CNT/GCE; the pockets of

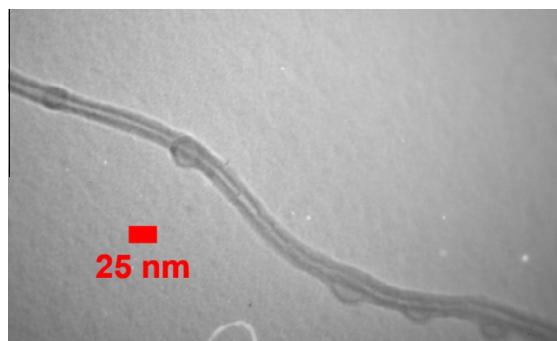


Fig. 1. TEM images of C_{60} -CNT composite.

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