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Simultaneous determination of tyrosine, acetaminophen and ascorbic acid using gold nanoparticles/multiwalled carbon nanotube/glassy carbon electrode by differential pulse voltammetric method

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ABSTRACT

A simple strategy for simultaneous determination of tyrosine (Tyr), acetaminophen (AC) and ascorbic acid (AA) based on gold nanoparticles/multiwalled carbon nanotube nanocomposite modified glassy carbon electrode (AuNPs/MWCNT/GCE) is reported. Scanning electron microscopy (SEM), electrochemical impedance spectroscopy (EIS), cyclic voltammetry (CV) and differential pulse voltammetry (DPV) were used to characterize the performance and microstructure of the sensor. The AuNPs/MWCNT/GCE displayed excellent electrochemical catalytic activities. The oxidation overpotentials of Tyr, AC and AA decreased significantly and their oxidation peak currents increased dramatically at AuNPs/MWCNT/GCE.DPV was used for the simultaneous determination of Tyr, AC and AA in their ternary mixture. Under the optimized experimental conditions Tyr, AC and AA give linear response over the range of 0.4–80.0 μmol L^{-1}, 0.09–35.0 μmol L^{-1} and 1.0–150.0 μmol L^{-1}, respectively. The lower detection limits were found to be 0.21 for Tyr, 0.03 for AC and 0.76 μmol L^{-1} for AA. The practical application of the modified electrode was demonstrated by measuring the concentration of Tyr, AC and AA in blood serum and pharmaceutical samples.

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1. Introduction

Tyrosine (4-hydroxyphenylalanine, Tyr), acetaminophen (AC) and ascorbic acid (AA) are biochemical compounds which play important roles in various biological processes. Usually, they are co-existing substances in biological matrices. Tyr is an essential aromatic amino acid and vital constituent of proteins, which is indispensable in human nutrition for establishing and maintaining a positive nitrogen balance [1]. Its absence could produce albinism, hypochondria, or depression. In contrast, high Tyr concentration in culture media increases sister chromatid exchange. Thus, Tyr has been of great interest [2]. Tyr and other amino acids play roles in inducing dementia such as Alzheimer's disease [3]. Alteration of Tyr concentration is also related to atherosclerosis [4] and lung diseases [5]. Acetaminophen (*N*-acetyl-*P*-aminophenol or paracetamol, AC) is a long-established substance being one of the most extensively employed drugs in the world. It is an antipyretic and analgesic drug commonly used against mild to moderate pains or for reduction of fevers [6]. In the therapeutic doses, it is a very effective and safe analgesic and considered as a substitute of aspirin. However, it has been associated with liver necrosis in humans and experimental animal softer high dose exposure [7]. Overdoses of AC can lead to the accumulation of toxic metabolites, causing severe and sometimes fatal hepatotoxicity and nephrotoxicity. Thus, it is necessary to develop efficient analytical techniques for the determination of AC [8–10]. Ascorbic acid (AA) is a soluble vitamin widely present in many biological systems and in multivitamin preparations, which is commonly used to supplement in adequate dietary intake and as an anti-oxidant [11,12]. Altering the concentration of these species is cause to several diseases, so their determination in human fluids such as blood serum, blood plasma and urine is much essential.

Various analytical techniques for individual or simultaneous determination of Tyr, AC and AA have been reported in literature such as fluorometry [13], chromatography [14–18], spectrophotometry [19,20] and capillary zone electrophoresis [21–23]. These methods are time consuming and/or expensive and often need the pretreatment step. Also, some of them suffer from low sensitivity and selectivity in the corresponding determinations. To overcome these defects, electrochemical methods are used extensively for the elegant and sensitive properties such as selectivity, reproducibility, low cost and simplicity of this approach. However, one major problem is that the oxidation peaks of AA and AC are too close at unmodified electrodes, which results in overlapping voltammetric responses and making their simultaneous detection highly difficult. To overcome this problem, it is necessary to make further efforts for the fabrication of electrochemical modified

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electrodes that can be used for simultaneously determination of these compounds.

In recent years, chemically modified electrodes have attracted large interest due to their potential applications in various analyses [24–26]. Carbon nanotubes (CNTs) have attracted considerable attention because of their excellent properties such as high electrical conductivity, good chemical stability, and extreme mechanical strength [27]. It has been shown that application of CNTs results in extraordinary advantages over conventional electrodes, including enhanced mass transport (via thin layer diffusion besides the semi-infinite planar diffusion), catalysis, highly effective surface areas, high porosity, more adsorption, and reactive sites. Furthermore, control over the electrode macro environment can provide an important and feasible platform for electroanalysis, particularly in the design of the modified electrodes for electrochemical sensing [28,29]. Gold nanoparticles (AuNPs) have emerged as a promising new material and their widespread application in the fields of electronics, catalysis and biosensors has received worldwide attention in recent years. AuNPs, due to their large aspect ratio, biocompatibility and high electrical conductivity have also been widely employed as a modifier in voltammetry for analysis of various species [24,30].

In this current study, AuNPs/MWCNT composite-modified glassy carbon electrode (GCE) was used for the simultaneous determination of Tyr, AC and AA. The electrochemical behaviors of these species at the AuNPs/MWCNT/GCE were investigated using cyclic voltammetry (CV) and differential pulse voltammetry (DPV) techniques. The experimental results indicate that AuNPs/MWCNT hybrid exhibit remarkable ability to increase the electroactive surface area and enhance the electron-transfer between the electrode and the analytes. Finally, to evaluate the utility of the modified electrode for analytical applications, it was used for the voltammetric determination of Tyr, AC and AA in real samples such as commercial tablets and a blood serum. However, we have developed a method that is simple, rapid, selective, and sensitive for simultaneous determination of Tyr, AC and AA in real samples.

2. Experimental

2.1. Chemicals

Tyr, AC, AA, acidic solution of $HAuCl_4$ and the other reagents were obtained from Merck Company and used as received. Multiwalled carbon nanotubes (10–20 nm in diameter, length of 30 μm, purity of 95%) were purchased from Neutrino Company (Iran). All the chemical reagents used were of analytical grade. A Briton–Robinson buffer solution (BRBS) of pH 6.0 served as a supporting electrolyte solution. The pharmaceutical samples, AC (325 mg, Rouz Darou Company, Iran) and AA (250 mg, Darou Pakhsh Company, Iran) tablets, were obtained from local drug stores. All solutions were prepared with double distilled water (DDW).

2.2. Apparatus

Electrochemical measurements were performed on a potentiostat/galvanostat (Autolab PGSTAT302N). It was controlled by a computer using Nova version 1.7 software. A platinum wire was used as the auxiliary electrode. AuNPs/MWCNT/GCE and Ag/AgCl were used as the working and reference electrodes, respectively. A Metrohm Model 713 pH lab (Herisau, Switzerland) was used for pH measurements. The electrode prepared was characterized by scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDS) (VEGA TESCAN).

2.3. Electrode preparation

A bare GCE was polished successively with 0.3 μm Al_2O_3 water slurry using a polishing cloth and it was rinsed with doubly distilled water, sonicated subsequently in a 1:1 aqueous HNO_3 solution, acetone, and doubly distilled water each for 10 min. The freshly cleaned GCE was electrochemically activated in a 1.0 $\mu mol\,L^{-1}$ H_2SO_4 solution at a scan rate of 100 $mV\,s^{-1}$ using 20 times cyclic potential sweeps in the range of −0.6 to 2.0 V. The fabrication of MWCNT is described as follows. The MWCNTs' suspension was prepared by dispersing 1.0 mg MWCNTs in 5.0 mL 3:1:1 mixture of DDW, ethanol and sodium dodecyl sulphate (SDS) under sonication for 30 min. A 10 μL aliquot of black suspension was dropped directly onto the clean GCE surface and dried at room temperature to form a MWCNT film at the GCE surface and prepare an MWCNT–modified GCE (MWCNT/GCE) [31,32]. The formation of AuNPs on the MWCNT/GCE was carried out by cyclic voltammetry (CV) scanning from 0.7 V to 0.0 V, vs. Ag/AgCl, in 0.1 $mol\,L^{-1}$ KCl and 2 $mol\,L^{-1}$ HCl solution containing 250 $mg\,L^{-1}$ $HAuCl_4$ at a scan rate of 50 $mV\,s^{-1}$ for 20 cycles. The obtained electrode was marked as AuNPs/MWCNT/GCE. The modified electrode was taken out and rinsed with water. Through changing the cycle number in electrodeposition process, the amount and the size of the deposited AuNPs can be controlled. Because the size of the nanoparticles significantly influenced the catalytic efficiency [33], 20 cycles was chosen as the optimum cycle number in gold electrodeposition process.

2.4. Pharmaceutical sample solution preparation

Five tablets of the AC and AA were finely powdered in a mortar with pestle. Calculated amounts of the tablets required for AC and AA were separately transferred into a 25 mL volumetric flask and were dissolved in BRBS of pH 6.0. The content of the flask was sonicated for 5 min to affect complete dissolution. Finally the solutions were filtered and suitable aliquot of the clear filtrate were collected and stored in the refrigerator for further uses.

2.5. Serum sample preparation

Serum samples were obtained and stored frozen until the analysis. The serum samples were prepared in two ways. In first way: a 0.8 mL of acetonitrile was added to a 1 mL serum sample to remove serum protein [32], followed by fortification with Tyr, AC and AA dissolved in BRBS to achieve the final concentrations of Tyr, AC and AA as 20 $\mu mol\,L^{-1}$, 10 $\mu mol\,L^{-1}$ and 50 $\mu mol\,L^{-1}$, respectively. After vortexing for 45 s, the mixture was centrifuged for 10 min at 15,000 rpm to remove the serum protein residues. Supernatant was taken carefully and appropriate volumes of this supernatant were transferred into the 25 mL flask and diluted up to the volume with the BRBS (serum 1). In second way: for preparation of serum samples, 1.0 mL of each sample was diluted to 50.0 mL by BRBS (pH 6.0). Then, 20 mL of this solution was transferred to the voltammetric cell and different amounts of Tyr, AC and AA were added and the recovery percent was obtained by DPV technique and standard addition method (serum 2) [8].

3. Results and discussion

3.1. Characterization of AuNPs/MWCNT/GCE

For characterization of surface morphology of AuNPs/MWCNT/GCE a scanning electron microscope was employed. Fig. 1 shows SEM images of MWCNT (Fig. 1A) and AuNPs on the MWCNT film surface (Fig. 1B). It shows a network-like structure of MWCNTs without aggregation was observed

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