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ORIGINAL ARTICLE

A new sensor based on glassy carbon electrode modified with nanocomposite for simultaneous determination of acetaminophen, ascorbic acid and uric acid

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Abstract A chemically-modified electrode has been constructed based on a single walled carbon nanotube/chitosan/room temperature ionic liquid nanocomposite modified glassy carbon electrode (SWCNTs–CHIT–RTIL/GCE). It was demonstrated that this sensor could be used for simultaneous determination of acetaminophen (ACT), uric acid (URI) and ascorbic acid (ASC). The measurements were carried out by application of differential pulse voltammetry (DPV), cyclic voltammetry (CV) and chronoamperometry (CA) methods. Electrochemical studies suggested that the RTIL and SWCNTs provided a synergistic augmentation that can increase current responses by improvement of electron transfers of these compounds on the electrode surface. The presence of the CHIT in the modified electrode can enhance the repeatability of the sensor by its antifouling effect. The modified electrode showed electrochemical responses with high sensitivity for ACT, URI and ASC determination, which makes it a suitable sensor for simultaneous sub-μmol L^{-1} detection of ACT, URI and ASC in aqueous solutions. The analytical performance of this sensor has been evaluated for detection of ACT, URI and ASC in human serum and urine with satisfactory results.

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1. Introduction

Carbon nanotubes (CNTs) are new kinds of porous nanostructured carbon materials, possessing properties such as high electrical conductivity, high surface area, chemical stability and significant mechanical strength (Yakobson and Smalley, 1997;

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Inagaki et al., 2004). CNTs can be used to promote electron transfer reactions when they are used as electrode materials in electrochemical devices (Yin et al., 2005; Yang et al., 2005).

Room temperature ionic liquids (RTILs) have been extensively used as a material for preparation of the modified electrode because of their interesting properties such as low toxicity, wide electrochemical potential window, high ionic conductivity and increasing the sensitivity of response (Zhao et al., 2004, 2007; Lu et al., 2006; Du et al., 2007).

A wide range of polymer and inorganic mesoporous metal oxides have been combined to form nanocomposite materials with unique mechanical, electrical, magnetic and adhesive properties (Gangopadhyay and De, 2000; Chiang and Whang, 2003). Chitosan (CHIT) as a natural polymer is biocompatible, biodegradable, nontoxic, highly hydrophilic with high mechanical strength, low cost, chemical inertness, and good adhesion properties that exhibit an excellent membrane-forming ability. Because chitosan–CNTs can form stable complexes through noncovalent binding, the stability of CNTs in aqueous chitosan solution was greatly improved. Thus, CNTs could be uniformly distributed in the chitosan film (Huang et al., 2002; Wang et al., 2002; Zhang and Zheng, 2008; Babaei et al., 2010a,b, 2011a,b).

Acetaminophen (ACT) is an important medicine, extensively used both in a pure form and in pharmaceutical preparations. It is mainly used as an alternative to aspirin as an analgesic and antipyretic agent that lacks the disadvantageous secondary effects of the salicylates on the gastric mucosa. It is often self-prescribed, without medical control, for relief of moderate pain, fever, lumbar pain, migraine or even non-specific indications. It has been reported to be a useful drug in osteoarthritis therapy. However an overdose of ACT can result in the accumulation of toxic metabolites that may cause severe and sometimes fatal hepatoxicity (Mugford and Tarloff, 1997). It can also cause liver disorders, skin rashes and inflammation of the pancreas (Prabakar and Narayanan, 2007). A number of analytical techniques such as spectrophotometry (Ayora Canada et al., 2000), spectrofluorometry (Vilchez et al., 1995), voltammetry (Babaei et al., 2010a,b, 2011a,b), HPLC (Ravisankar et al., 1998), colorimetry (Knochen et al., 2003) and Fourier transform infra red spectrometry (Ramos et al., 1998), have been proposed for the determination of ACT in pharmaceutical formulations and biological samples.

Uric acid (URI) is the primary end product of purine metabolism, and considered as a species of great importance in human diagnosis. The typical concentration of URI in blood is in the range of 120–450 μmol L^{-1}. An abnormal concentration of URI can indicate the presence of one of numerous diseases and/or physiological disorders. An elevated concentration of URI is observed in patients suffering from diseases such as gout and hyperuricemia (Dutt and Mottola, 1974; Zen et al., 1998). Because of its clinical relevance, it is crucial to develop simple and rapid methods for URI determination in routine analysis. Various methods have been used to accomplish this, such as enzyme-based systems (Zhao et al., 2009), fluorescence (Galban et al., 2001), chemiluminescence (Yang and Zhang, 2010), capillary electrophoresis (Zhao et al., 2008), liquid chromatography (Perelló et al., 2005) and voltammetry (Inada et al., 2009). A major problem for the electrochemical detection of URI is interfering species such as ACT, which is oxidized at a similar potential to that of URI making direct determination of ACT or URI unreliable.

Ascorbic acid (ASC) an essential nutrient can be found mainly in fruits and vegetables. The body requires ASC to form collagen protein to maintain bones, blood vessels, and skin. Due to its antioxidant and pH regulator properties, this vitamin is present or added to a wide variety of food products and pharmaceuticals. Ascorbic acid is easily oxidized chemically and electrochemically to L-dehydroascorbic acid (Sabzi and Pournaghi-Azar, 2005). ASC is unstable, undergoing oxidation, especially in aerobic conditions, alkaline media, and at exposure to light (Zeng et al., 2005). Ascorbic acid is widely found in association with various biologically and pharmacologically active substances, including acetaminophen (Săndulescu et al., 2000) in various pharmaceutical products as well as in biological fluids (Săndulescu et al., 2000; Zhang et al., 2001; Arvand et al., 2003; Ramesh and Sampath, 2004). An overdose amount of ASC in some people may lead to diarrhea, nausea, skin irritation, burning upon urination, and depletion of copper. In addition ASC can cause adverse reactions when taken with some drugs. Therefore determination of ASC in the presence of some popular medicines like ACT is of major interest (Săndulescu et al., 2000). Several available methods have been reported for the determination of ascorbic acid in pharmaceutical preparations, biological fluids, food and beverages. The methods are: fluorimetry (Yang et al., 2001), HPLC (Kand'ár and Záková, 2008), spectrophotometry (Salkić and Kubiček, 2008) and voltammetry (Satheesh Babu et al., 2010).

The advantages of an electrochemical technique for the determinations of ACT, URI and ASC are high sensitivity, low cost, and rapid measurement time. In this work we outline the use of a single-walled carbon nanotube/chitosan/room temperature ionic liquid nanocomposite modified glassy carbon electrode (SWCNTs–CHIT–RTIL/GCE) as a sensor for this purpose. To the best of our knowledge there has been no report of the use of an electrochemical sensor for the simultaneous determination of ACT, URI and ASC compounds. In addition, the analytical performance of this sensor for the determination of ACT, URI and ASC in human serum and human urine samples was evaluated with satisfactory results.

2. Experimental

2.1. Reagents

All chemicals were analytical grade and used as received. ACT, URI and ASC were obtained from Merck chemical company. Chitosan (MW 1.0–3.0 × 10^5) and Single-walled carbon nanotubes (SWCNTs) were purchased from Acros and Sigma companies, respectively. The purity of SWCNTs was 90% with a surface specific area of 480 m^2 g^{-1}, diameter of 1–2 nm and length of 0.5–2 μm.

1-Ethyl-3-methyl-imidazolium tetra-fluoro borat (EMIM·BF_4) was obtained from Merck chemical company. All ACT, URI and ASC solutions were prepared by diluting the stock standard solutions using 0.1 mol L^{-1} phosphate buffer (pH 7). 0.1 mol L^{-1} Phosphate buffer solution (PBS) was prepared by dissolving appropriate amounts of sodium hydrogen phosphate and sodium dihydrogen phosphate in a 250 mL volumetric flask. pH of solution was adjusted to an appropriate value by addition of 7.5 mol L^{-1} sodium hydroxide solution. Electrochemical experiments were carried out on ACT, URI and ASC in 0.1 mol L^{-1} PBS at pH 7.