

Electrochemical determination of carbamazepine in the presence of paracetamol using a carbon ionic liquid paste electrode modified with a three-dimensional graphene/MWCNT hybrid composite film

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ABSTRACT

In this research work, a carbon ionic liquid paste electrode (CILPE) based on butyl-3-methylimidazolium bis (trifluoromethylsulfonyl) imide ([bmim] NTF₂) was fabricated and further modified with graphene/multiwall carbon nanotube (GR/MWCNT) hybrid composite. The modified electrode (GR/MWCNT/CILPE) was used for measurement of carbamazepine (CBZ) in the presence of paracetamol (PA) with an excellent electrochemical catalytic behavior. The application of the electrode was investigated by cyclic voltammetry and differential pulse voltammetry. The oxidation peak currents of CBZ and PA were linear at the ranges of 1–60 μ M and 2–80 μ M, respectively. Also, the detection limits for CBZ and PA were 0.233 μ M and 0.262 μ M, respectively. The proposed sensor was successfully applied for the determination of CBZ and PA in tablet and urine samples.

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1. Introduction

Carbamazepine (CBZ) (Fig. 1a), is an anticonvulsant, antiepileptic and mood stabilizing drug which is used primarily in the treatment of epilepsy and bipolar disorder. It can also be administered to patients who have other illnesses including schizophrenia, neuromyotonia, attention-deficit hyperactivity disorder (ADHD), and post-traumatic stress disorder [1,2]. As a result of its widespread use, CBZ determination in biological and pharmaceutical samples is of great importance. Several analytical methods have been used for determination of CBZ in pharmaceutical formulations and biological samples including capillary electrophoresis [3], liquid chromatography [4–6] and electrochemical methods [7,8]. Although liquid chromatography has been widely recommended for determination of CBZ, because of its high sensitivity and selectivity [9], but it is time consuming, solvent usage intensive and requires expensive devices and maintenance.

Paracetamol (PA), (Fig. 1b) has been used comprehensively all over the world as the reducer of fever and as a pain killer for the relief of moderate pain associated with backache, headache, arthritis and post-operative pain [10]. However, the overdose of PA can lead to the accumulation of toxic metabolites which may cause hepatotoxicity and nephrotoxicity [11]. Several analytical methods such as spectrophotometry [12], chromatography [13], capillary electrophoresis [14] and

electrochemical methods [15,16] have been developed for determination of paracetamol in solutions. Whereas, combination drugs consisting of paracetamol and carbamazepine are widely used as analgesic and antipyretic agents for treating the symptoms of different painful processes, thus a simple, rapid and reliable method that could be easily and widely used for the simultaneous quantitative estimation of these compounds is necessary.

Among the different analytical methods, electrochemical determination has proved to be sensitive and reliable for determination of numerous electroactive components [17–19]. On the other hand, the poor ability of the bare electrodes in direct electrochemical activity of different electroactive materials has led to interest in the use of mediators and modified electrodes to catalyze the electrochemical oxidation and/or reduction of them [20,21].

MWCNTs have been widely used in electrochemistry due to their unique one-dimensional (1D) structural, electronic, and physical properties. In the field of chemically modified electrode, one of the most important characteristics of MWCNTs is their reported ability to promote electron-transfer process [22–26].

Graphene (GR) a 2D carbon material comprised of a single sheet of hexagonally packed carbon atoms [27,28], has attracted tremendous attention because of its unique nanostructure and extraordinary electrocatalytic properties such as high surface area, excellent conductivity, and high mechanical strength [29]. Based on its properties, GR is considered as an ideal electrode material for electrochemical and biosensing [30]. However, the excellent properties of GR emerge only in a planar

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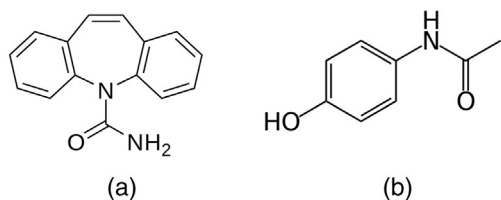


Fig. 1. Chemical structure of carbamazepine (a) and paracetamol (b).

direction. The properties of MWCNTs emerge in the axial direction while providing current density, high specific surface area, and thermal conductivity. Thus, a GR-MWCNT hybrid that combines the unique properties of the two carbon allotropes in all directions and provides a high surface area per unit volume for increased catalyst loading could be an ideal electrode material [31,32].

Ionic liquids (ILs) have received great interests recently because of their unusual properties as liquids [33–35]. ILs are composed of only ions, they show very high ionic conductivity, nonvolatility, and chemical stability. As new green media, ILs have many unique electrochemical properties, such as higher ionic conductivity and wider electrochemical windows [36–39]. CILPE is a new type of working electrode that is prepared by using an ionic liquid as a binder and modifier in the traditional carbon paste electrode (CPE). Due to the presence of IL in the carbon paste, CILPE had shown the advantages such as increased conductivity,

easy preparation, good reversibility, high sensitivity and the ability to lower the over-potential of the electroactive compounds [40]. CILPE can also be further decorated to get the modified electrode. Different kinds of the modified CILPE had been prepared and used for electrochemical sensing with excellent performance [41,42].

In this study, a three-dimensional graphene/MWCNT hybrid composite was fabricated and further modified on a 1-butyl-3-methylimidazolium bis (trifluoromethylsulfonyl) imide ([bmim] NTF₂) based carbon ionic liquid paste electrode (CILPE). The modified electrode was further used for the detection of CBZ in the presence of PA. To the best of our knowledge, the electrochemical detection of CBZ by using CILE has not been reported in the literature. Based on the electrochemical response of CBZ and PA on GR/MWCNT/CILPE, a new electrochemical method was established for simultaneous determination of CBZ and PA with good electrocatalytic activity, high sensitivity, good reproducibility, long-term stability and low cost.

2. Experimental

2.1. Chemicals

CBZ was purchased from Alborz Bulk pharmaceutical Co. (Tehran Iran), PA was obtained from Borhan Darou Co. (Mashhad Iran). 1.0×10^{-2} M stock standard solutions of CBZ and PA were prepared and stored at 4 °C. Working standard solutions were prepared by

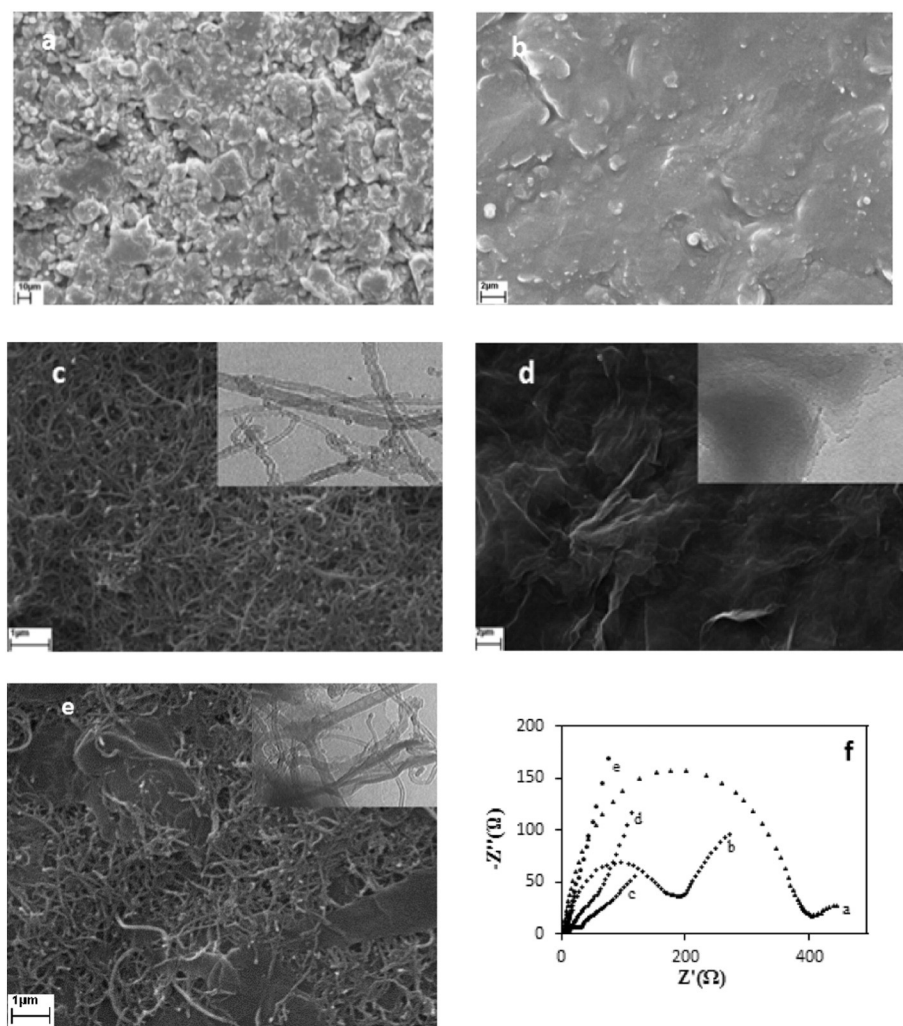


Fig. 2. SEM images of (a) CPE, (b) CILPE, (c) MWCNT/CILPE, (d) GR/CILPE, (e) GR/MWCNT/CILPE (inset TEM image), (f) EISs of (a) CPE, (b) CILPE, (c) GR/CILPE, (d) MWCNT/CILPE, (e) GR/MWCNT/CILPE in 5.0×10^{-3} mol L⁻¹ [Fe(CN)₆]^{-3/-4} containing 0.1 mol L⁻¹ KCl.

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