Journal of Materials Science and Nanomaterials

Zeming, J Mater Sci Nanomater 2022, 6:3

Mini Review Open Access

Facile Electrochemical Sensor for Sensitive and Selective Determination of Guaifenesin, Phenylephrine and Paracetamol on Electrochemically Pretreated Pencil Graphite Electrode

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Abstract

Guaifenesin (GFS), phenylephrine (PHE) and paracetamol (PAR), drugs used in combination for the relief of cold and flu symptoms, were determined at electrochemically pretreated pencil graphite electrode. Differential pulse voltammetry (DPV) was used for the first time for the concomitant determination of the target compounds based on the electro-oxidation of PAR at 0.43 V, PHE at 0.74 V and GFS at 1.14 V in Britton–Robinson buffer pH 6.0. Under optimized experimental con-ditions, two linear ranges were obtained for PAR (2.50 × 10−6 M–1.00 × 10−5 M and 1.00 × 10−5 M–1.00 × 10−4 M) and for PHE and GFS linearity was proved between 5.00 × 10−6 M–2.00 × 10−4 M and 2.50 × 10−6 M–2.00 × 10−4 M, respectively. The detection limits were 8.12 × 10−7 M for PAR, 1.80 × 10−6 M for PHE and 8.29 × 10−7 M for GFS.The selective and sensitive DPV method and the electrochemically treated electrode [1].

Keywords: Paracetamol; Hypertension; Phenylephrine

Introduction

Guaifenesin (GFS), phenylephrine (PHE) and paracetamol (PAR) are active ingredients frequently combined in pharmaceutical formulations administered for short-term treatment of cold and flu symptoms (pain, nasal congestion, headache, fever and chesty cough). In such over-the-counter drugs, guaifenesin (3-(2-methoxyphenoxy)-1,2-pro-panediol) is an expectorant [2], phenylephrine (2-methylamino-1-3(-hydroxyphenyl) ethanol is used as a decongestant and paracetamol (N-(4-hydroxyphenyl) acetamide) acts as an analgesic and antipyretic agent . The quality control of such preparations is of special importance considering that the overdoses can cause a range of hepatic damages (PAR), depression of the central nervous system (GFS) or severe hypertension and tachycardia (PHE). Therefore, the rigorous quantitative determination of these chemical compounds is of great interest. However, a challenge in the analysis of pharmaceutical preparations consists in the simultaneous detection, without preliminary separation, of more active ingredients with similar physiochemical properties, from the complex formulations that contain a wide range of excipients [3].

Despite the fact that there are many published papers on the individual electrochemical determination of the three mentioned drugs, but also in combination with other active sub- stances, the literature survey revealed that there is no reported electrochemical method for the simultaneous analysis of the ternary mixture of GFS, PHE and PAR in pharmaceutical formulations. Thus, PAR was determined together with GFS in the presence of ascorbic acid or oxomemazine hydrochloride using modified carbon paste electrodes. There are more electrochemical methods proposed for the determination of PAR and PHE, these active ingredients being quantified in their binary mixtures or together with chlor- pheniramine maleate, dextromethorphan, and cetirizine, ascorbic acid or lo- ratadine. In all these studies chemically modified electrodes based on carbon paste or glassy carbon, but also boron-doped diamond electrodes were used. A cheaper and simple alternative to these modified electrodes that require expensive reagents and additional preparation steps is the pencil graphite electrode (PGE). Further, the low cost and disposable use that eliminates the tedious cleaning procedures, PGE benefits from the excellent properties of composite graphite. PGE was used as PAR electrochemical sensor in pharmaceutical formulations and different biological samples, the literature data on this subject being presented in a review paper [4, 5]. For GFS deter- mination, PGE was modified with silver nanoparticles and poly (L-cysteine).

PHE quantification there is no study that uses this type of electrode. Therefore, the main objective of the present study was to develop a differential pulse voltammetric (DPV) method which was able to quickly and selectively determine, in a single anodic scan, GFS, PHE and PAR from pharmaceutical formulations using the electrochemically pre-treated PGE (PGE*). Moreover, the electrochemical behavior of all phar- maceutical active ingredients was studied at the PGE* surface.

Materials and Method

Reagents and Apparatus The stock standard solutions of 1.00 × 10−2 M PAR, PHE and GFS were daily prepared by dissolving the corresponding reagent purchased from Merck in double distilled water and were stored in the refrigerator until further use. The chemicals needed to obtain Brit- ton–Robinson (BR) supporting electrolyte solutions (acetic acid, phosphoric acid, boric acid and sodium hydroxide) were also acquired from Merck. Sachets with powder for oral solution containing paracetamol (500 mg), guaifenesin (200 mg) and phenylephrine hydrochloride (10 mg) were bought from a local pharmacy. Cyclic voltammetry (CV) and DPV were performed using an analytical system model Auto lab PGSTAT 128 N controlled by Nova 1.11 software (Ecochemie B.V., Netherlands) [6]. A glass

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Received: 2-May-2022, Manuscript No: JMSN-22-71168; **Editor assigned:** 06-May-2022, Pre-QC No: JMSN-22-71168 (PQ); **Reviewed:** 20-May-2022, QC No: JMSN-22-71168; **Revised:** 24-May-2022, Manuscript No: JMSN-22-71168 (R); **Published:** 30-May-2022, DOI: 10.4172/jmsn.100040

Citation: Zeming H (2022) Facile Electrochemical Sensor for Sensitive and Selective Determination of Guaifenesin, Phenylephrine and Paracetamol on Electrochemically Pretreated Pencil Graphite Electrode. J Mater Sci Nanomater 6: 040.

cell containing 10 mL of solution and a three electrode system were used: PGE* as working electrode (if not stated otherwise), Ag/AgCl (3.00 M KCl) and platinum wire as reference and counter electrodes, respectively. Rotring graphite pencil leads with different levels of hardness (2H, H, HB, B and 2B) and diameter of 0.50 mm constituted the working electrode. The length of the lead inserted into the solution was 1.00 cm, PGE being pre- pared according to our previous works. All pH measurements were carried out with a Consort C6010 pH/mV-meter (Fisher Scientific, Merelbeke, Belgium) at room temperature.

Sample Analysis

Three sachets containing the pharmaceutical mixture were examined, five replicate samples being analysed from each sachet. The content of one sachet with powder for oral solution was prepared according to the label instructions: it was dissolved in 250 mL warm water and then allowed to cool to room temperature. The thus obtained solution was further diluted with the appropriate supporting electrolyte such that the concentration of the sample subjected to the voltammetric measurement fell within the linear range. For the quantitative determination of GFS, PHE and PAR the standard addition method was applied. Thereby, three different volumes of the stock standard solution were added into the volumetric flasks containing the same diluted sample volume, each time the final concentration falling into the linear range. Taking into consideration that the de- clared contents of the target compounds in the pharmaceutical formulation were significantly different, the standard addition method was performed for each analyte at a time [7, 8]. Differential pulse voltammograms were recorded forth diluted sample solution and for each of the solutions obtained after the additions were made.

Results and Discussion

Electrochemical Behavior of PAR, PHE and GFS at PGE* It is well-known that simultaneous determination of electro active compounds is sometimes difficult due to their voltammetric responses overlapping [9-15]. In the present study DPV measurements were realized in order to evaluate the electrochemical responses of PAR, PHE and GFS in BR buffer solution pH 6.00 at PGE and PGE*, respectively. In order to verify the possibility of simultaneous determination of the three compounds, electro- chemical experiments were firstly performed for each analyte. Thus, at PGE*, in the solution containing PAR a well-defined anodic voltammetric response was obtained at 0.43 V. For PHE, the electrochemical signal was at 0.74 V, while GFS presented an anodic peak at 1.14 V. In the differential pulse voltammogram recorded for the drugs mixture solution distinct electrochemical signals were observed at the same potentials as in the individual voltammograms, which can be attributed to the oxidation of PAR, PHE and GFS, respectively. The significant differences between the peak potentials of the analytic (0.31 V and 0.40 V for PAR-PHE and PHEGFS, respectively) made possible the simultaneous determination of PAR, PHE and GFS in their mixture solution.

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