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Original Article

Voltammetric sensor for tartrazine determination in soft drinks using poly (*p*-aminobenzenesulfonic acid)/zinc oxide nanoparticles in carbon paste electrode

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ABSTRACT

Zinc oxide nanoparticles (ZnO NPs) and *p*-aminobenzenesulfonic acid (*p*-ABSA) were used to fabricate a modified electrode, as a highly sensitive and selective voltammetric sensor, for the determination of tartrazine. A fast and easy method for the fabrication of poly *p*-ABSA (P*p*-ABSA)/ZnO NPs-carbon paste electrode (P*p*-ABSA/ZnO NPs-CPE) by cyclic voltammetry was used. By combining the benefits of P*p*-ABSA, ZnO NPs, and CPE, the resulted modified electrode exhibited outstanding electrocatalytic activity in terms of tartrazine oxidation by giving much higher peak currents than those obtained for the unmodified CPE and also other constructed electrodes. The effects of various experimental parameters on the voltammetric response of tartrazine were investigated. At the optimum conditions, the sensor has a linear response in the concentration range of 0349–5.44 μM, a good detection sensitivity (2.2034 μA/μM), and a detection limit of 80 nM of tartrazine. The proposed electrode was used for the determination of tartrazine in soft drinks with satisfactory results.

1. Introduction

Food additives are commonly used in processed foodstuffs to improve appearance, flavor, taste, color, texture, nutritive value, and preservation. Synthetic colorants, compared with to natural dyestuffs, has been extensively used in food industries in the past four decades because of their higher brightness, more stability, cheapness, and the wider range of shades [1,2].

Generally, synthetic dyes contain azo (N = N) functional groups and aromatic ring structures; therefore, they are harmful to human health [3]. Thus, each synthetic food colorant has been evaluated by the Food and Agricultural Organization and World Health Organization [4]. Tartrazine is one of these azo colorants that may be present in common food products, which can cause allergies, migraines, eczema, anxiety, diarrhea, and even cancer if they are excessively

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consumed [5,6]. Facing with increasing legal restrictions, food dye determination has become an analytical challenge. Until now, several methods, such as spectrophotometry [7,8], high performance liquid chromatography (HPLC) [9,10], and capillary electrophoresis [11], have been used for the single or simultaneous determination of tartrazine. On the other hand, electrochemical techniques has obtained much attention and exhibited promising application in food safety analysis due to its portability, excellent sensitivity, automation, short analysis time, low power consumption, and inexpensive equipment [12–18]. Till date, different electrodes have been developed for the single or simultaneous electrochemical determination of different food colorants [19–30].

Carbon electrodes are widely used in electroanalysis due to their low background current, wide potential window, chemical inertness, low cost, and suitability for various sensing and detection applications. Among carbon electrodes, carbon paste electrode (CPE) has a particular importance. The ease and speed of preparation and obtaining a new reproducible surface, very low background current (compared with graphite electrode), low cost, feasibility to incorporate different substances during the paste preparation, and porous surface are some advantages of CPEs over all other carbon electrodes [31]. Therefore, over the past years, CPEs containing various modifiers have been prepared and applied in the determination of different analytes.

The key point to obtain a good and reliable electrochemical sensor is the kind of materials that constitute the detection platform. In this field, the synergy between electrochemical sensors technology and nanomaterials is expecting to bring interesting advantages in the field of new electrochemical transducing platforms beside their use as electrochemical labels or tags for signal enhancement [32]. Sensors based on nanostructured materials take advantages of the increased electrode surface area, increased mass-transport rate, and fast electron transfer compared with electrodes based on bulk materials [32,33]. In addition to novel properties, nanomaterials open up new approaches to fabricate the electrodes cost effectively by minimizing the needed materials and waste generation [34].

Metal oxide nanoparticles have received much attention recently. Various examples of the electrochemical applications are the modification of electrodes with metal oxide nanoparticles in order to develop electrochemical sensors. Among the different metal oxide nanoparticles, zinc oxide (ZnO) is an attractive semiconductor, with a wide band gap (3.37 eV). ZnO has a large excitonic energy, low-cost synthesis, biocompatibility, good electrochemical activities, nontoxicity, high-electron communication features, and high mechanical strength [35]. Nanostructured ZnO has been used previously for the fabrication of different sensors and biosensors [35,36].

On the other hand, polymer-modified electrodes have obtained important attention among the researchers for sensor and biosensor applications because polymeric films have good stability and reproducibility. Electropolymerization is a good method to prepare polymer-modified electrodes as adjusting electrochemical parameters can control film thickness, permeation, and charge transport characteristics.

In this study, we have combined the advantageous features of polymer modification, metal oxide nanoparticles, and carbon paste technology with the aim of electrocatalytic oxidation of tartrazine using poly (*p*-aminobenzenesulfonic acid) (P*p*-ABSA) as a polymer, but with some changes in the carbon paste preparation. The performance of the developed electrode was studied by cyclic voltammetry, and close surface examination was made by scanning electron microscopy (SEM). The fabricated modified electrode facilitated the electron transfer for tartrazine, resulting in the increase of oxidation signals. The performance of the fabricated electrode in tartrazine electroanalysis with respect to sensitivity, selectivity, and linear concentration range was evaluated and discussed. The applicability of the electrode was demonstrated by determining tartrazine in processed soft drinks.

2. Experimental

2.1. Apparatus

Electrochemical measurements were performed on an AUTOLAB modular electrochemical system (ECO Chemie, Utrecht, The Netherlands) equipped with a PGSTAT 12 module and driven by general purpose electrochemical system software (GPES version 4.9) in conjunction with a conventional three-electrode system and a personal computer for data storage and processing. A bare or modified CPE as the working electrode, Ag/AgCl electrode as the reference electrode, and a platinum wire as the auxiliary electrode, were employed in the measurements. SEM images were recorded with KYKY-EM-3200 SEM system (China) at an accelerating voltage of 25 kV.

2.2. Reagents

2.2.1. Chemicals and reagents

Tartrazine was purchased from Aldrich (Canada), and dissolved in water to prepare the standard solution. Desired concentrations were obtained by diluting the standard solution with double distilled water. ZnO nanoparticles (ZnO NPs) were synthesized using solid–vapor phase thermal sublimation techniques. All other chemicals were of analytical grade and used directly. Double distilled water was used throughout the preparation of solutions.

2.2.2. Preparation of modified electrode

First, the ZnO NPs-CPE was prepared by hand mixing 65% graphite powder, 5% ZnO NPs, and 30% paraffin oil in an agate mortar to get a homogeneous carbon paste. Next, the paste was packed in the end of a plastic syringe (2 mm in diameter). A copper wire inserted into the carbon paste provided an electrical contact. The polymer film-modified ZnO NPs-CPE was fabricated by cyclic voltammetry in the potential range −0.6 to +1.8 V at a sweep rate of 100 mV/s in 0.1 M phosphate buffer solution (PBS) pH 7.0 containing 2 mM *p*-ABSA in the presence of 0.5 M KCl for 15 cycles [37]. The obtained modified electrode (P*p*-ABSA/ZnO NPs-CPE) was washed with double distilled water to remove the physically adsorbed material. Stepwise electrodes were also prepared with the same procedures described above for comparison purposes.