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Amplified nanostructure electrochemical sensor for simultaneous determination of captopril, acetaminophen, tyrosine and hydrochlorothiazide

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ABSTRACT

A novel nanomaterial-based voltammetric sensor has been developed for use a highly sensitive tool for the simultaneous determination of captopril (CA), acetaminophen (AC), tyrosine (TY) and hydrochlorothiazide (HCTZ). The device is based on the application of NiO/CNTs and (2-(3,4-dihydroxyphenethyl)isoindoline-1,3-dione) (DPID) to modify carbon paste electrodes. The NiO/CNTs nanocomposite was synthesized through a direct chemical precipitation approach and was characterized with X-ray powder diffraction (XRD), and scanning electron microscopy (SEM). The NiO/CNTs/DPID/CPEs were found to facilitate the analysis of CA, AC, TY and HCTZ in the concentration ranges of 0.07–200.0, 0.8–550.0, 5.0–750.0 and 10.0–600.0 μM with the respective detection limits of 9.0 nM, 0.3 μM, 1.0 μM and 5.0 μM. The developed NiO/CNTs/DPID/CPEs were used for the determination of the mentioned analytes in pharmaceutical and biological real samples.

1. Introduction

Captopril and hydrochlorothiazide are two important pharmaceuticals used individually or together for treating hypertension [1–3]. These compounds have found other applications in the treatment of congestive heart failure and swelling. On the other hand, acetaminophen has been found to decrease the effects of CAP by opposing drug effects. Recent studies have confirmed that nitrated tyrosine (TY) residues and acetaminophen (AC) adducts can be found in necrotic cells after being subjected to toxic doses of AC [4]. In this light, the development of highly sensitive and selective analytical sensors which can be used for the simultaneous determination of CA, AC, TY and HCTZ is of critical value for the analysis of real samples, yet this has remained a challenge due to the inability of most unmodified sensors to separate the oxidation peak potentials of the mentioned analytes.

Applications of modified electrodes are good choice for trace level analysis of biological and pharmaceutical samples [5–11]. In between, carbon paste modified electrodes (CPMEs) have attracted a great deal of attention on the part of electrochemical researchers working in the area of the determination of electro-active compounds. This can in part be due to the superior capability of CPMEs in comparison to other electrodes, including their low cost, low non-faradic currents and simple modification routines [12–18]. CPMEs have been suggested as highly sensitive tools for simultaneous determination of electroactive compounds with near over-voltages, in the recent years [19–27]. As an example, Sanghavi and Srivastava reported the application of a carbon-nanotube modified paste electrodes for the simultaneous determination of acetaminophen, aspirin and caffeine in their mixtures [13].

Based on the above mentioned information and in the light of the information presented the current study was based on the development and application of NiO/CNTs/DPID/CPEs for the voltammetric determination of CAP. The developed voltammetric sensor was also found to be able to sense and distinguish signals of CA, AC, TY and HCTZ in mixed samples for the first time. The device further enjoyed advantages including good performance and high sensitivity which made it applicable for use in the determination of CA, AC, TY and HCTZ in tablet and urine samples.

2. Experimental

2.1. Apparatus and materials

Captopril, acetaminophen, tyrosine, hydrochlorothiazide, graphite powder, paraffin oil, nickel nitrate, carbon nanotubes and sodium hydroxide were purchased from Sigma-Aldrich Company without any further treatments. Nickel nitrate, carbon nanotubes and sodium hydroxide were used for synthesized of NiO/CNTs nanocomposite according to my previous reported [28]. The crystallinity of the NiO/CNTs were examined through X-ray diffraction (XRD) using a STOE

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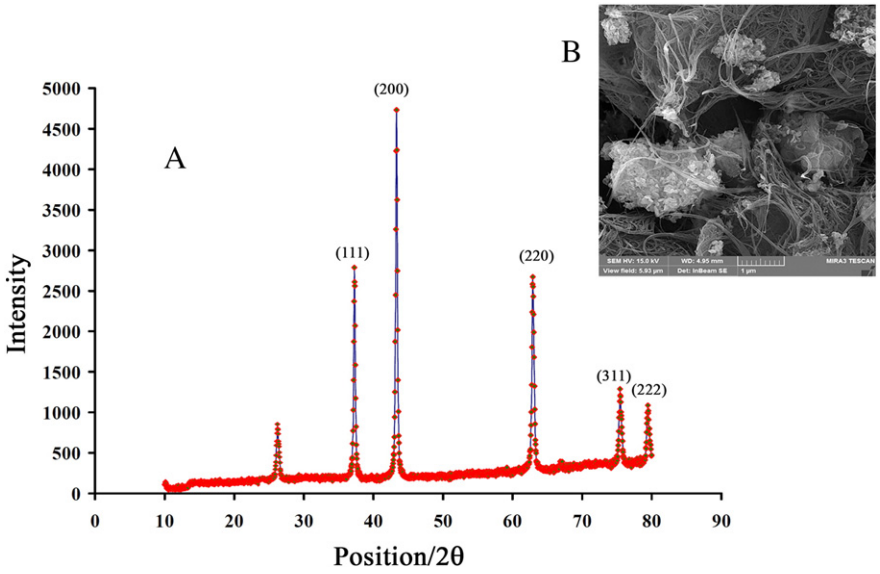

Fig. 1. A) XRD pattern of as-synthesized NiO/CNTs. B) SEM image of as-synthesized NiO/CNTs.

diffractometer with a Cu-Kα radiation source, in the range 20–80° (2θ). For investigating the morphology of the NiO/CNTs nanocomposites a digital scanning electron microscope (YKY-EM3200) was used. All of the electrochemical investigations were performed using an Autolab potentiostat/galvanostat PGSTAT 35 (Eco chemie Utrecht, Netherlands). An Ag/AgCl/KCl_{sat}, a platinum wire and the developed NiO/CNTs/DPID/CPEs were respectively used as the reference, electrode and electrodes.

2.2. Synthesis of DPID

In this procedure phthalic anhydride was reacted with 3-hydroxytyramine hydrochloride (dopamine hydrochloride) in an acetic acid/pyridine medium under reflux and the product, i.e. DPID was found to have a melting point in the range of 167–168 °C. The chemical structure of the product was studied through common spectroscopic techniques. The FT-IR spectra of the product included characteristic signal bands at 3238, 3051, 2947 , 1768, 1692, 1651, 1612, 1529, 1466, 1433, 1403, 1367, 1266, 1242, 1198, 1152, 1119, 1089, 1003, 952, 870, 531, 494. Further the ^{1}H NMR (CD_3OD) spectra showed peaks at 2.84 (t, 2H, C**H$_2$**CH_2N)), 3.85 (t, 2H, CH_2C**H$_2$**N), 6.51 (d, 1H, phenyl), 6.65 (d, 2H, phenyl), and 7.82 (d, 4H, phthalimide) and the ^{13}C NMR (CD_3OD) spectra contained peaks at 168.2, 144.9, 133.9, 131.9, 129.6, 122.6, 119.7, 115.5, 114.9, 39.2, and 33.3. Anal. Calcd. for $C_{16}H_{13}NO_4$ (283.08): C, 67.84, H, 4.63, N, 4.94. The yield of the DPID preparation process was about 88% and the process revealed great advantages over the previously reported DPID preparation techniques (about 18%).

2.3. Preparation of the sensor

To prepare the sensors 10.0 mg of DPID was hand mixed with 890.0 mg of graphite powder and 50.0 mg of NiO/CNTs in a mortar and pestle. Next 12 drops of nujol oil were added to the mixture and it was mixed well for 50 min until a uniformly-wetted paste was obtained. The resulting paste was then packed into a glass tube and an electrical contact was established through inserting a copper wire down the glass tube into the rear side of the paste.

2.4. Preparation of real samples

To prepare the tablet samples, eight tablets (from HCTZ, AC and CAP) were grinded to form a uniform powder. Next suitable amounts of the powder were dissolved in 100 mL water samples under sonication.

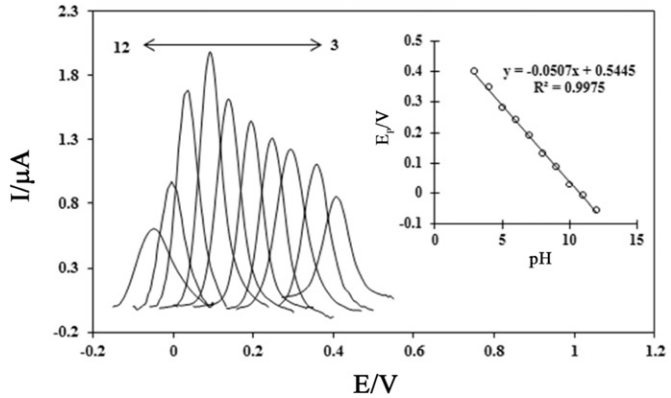

Fig. 2. Left; SWVs of NiO/CNTs/DPID/CPE at various buffered pHs. The numbers 1–10 correspond to 3.0, 4.0, 5.0, 6.0, 7.0, 8.0, 9.0, 10.0, 11.0 and 12 pHs, respectively. Right; Plot of Ep vs. pH for NiO/CNTs/DPID/CPE.

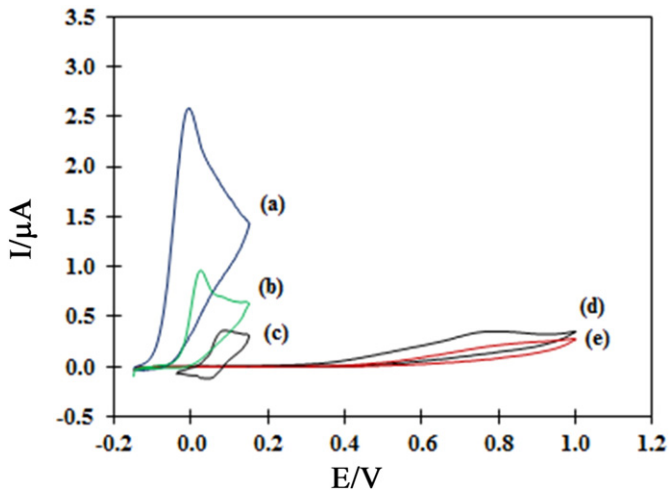

Fig. 3. Cyclic voltammograms of (a) NiO/CNTs/DPID/CPE in the buffer solution pH = 9.0, (b) DPID/CPE in the presence of 100 μM captopril, (c) NiO/CNTs/DPID/CPE in the presence of 100 μM captopril, (d) NiO/CNT/CPE in the presence of 100 μM captopril, and (e) CPE in the presence of 100 μM captopril. In all cases the scan rate was 10 mV s^{-1} and pH = 9.0.