



# Electrochemical performance of boron-doped diamond electrode in surfactant-containing media for ambroxol determination



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## ABSTRACT

A novel application of boron-doped diamond electrode is introduced for the determination of ambroxol, a potential antioxidant drug belonging to the expectorant class. The electrochemical oxidation of ambroxol was first investigated by cyclic voltammetry using boron-doped diamond and glassy carbon electrodes in aqueous solutions both with and without the addition of surfactant. The compound was irreversibly oxidized in one/two steps at high positive potentials, resulting in the formation of a couple with a reduction and re-oxidation wave at less positive potentials. Special attention was given to the use of adsorptive stripping voltammetry at a surface of mildly oxidized boron-doped diamond electrode in aqueous solutions over the pH range of 1.0–10.0. Addition of anionic surfactant (sodium dodecylsulfate) to ambroxol-containing electrolyte enhanced the stripping current signal. Using square-wave stripping mode, the drug yielded a well-defined voltammetric response in phosphate buffer pH 2.5 containing  $4 \times 10^{-4}$  M sodium dodecylsulfate at +1.02 V (versus Ag/AgCl) (after 30 s accumulation at +0.50 V). The process could be used to determine ambroxol in the concentration range of 0.05–0.7  $\mu$ M, with a detection limit of 0.010  $\mu$ M (4.2 ng mL<sup>-1</sup>). The suggested method was successfully applied to pharmaceuticals and spiked human urine samples.

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## 1. Introduction

Carbon materials have attracted much interest as possible electrodes for use in the area of electroanalytical chemistry. This is because electrochemical techniques offer advantages of simplicity, being rapid and relatively inexpensive, not requiring large amounts of reagents, and commonly having high sensitivity as regards constructing a sensor format. Glassy carbon (GC), one of the most important and traditional materials, has been widely used for determination of various electroactive compounds over several decades. Recently, new carbon-based materials, such as boron-doped diamond (BDD), carbon nanotubes and graphene nanostructures have been developed for electroanalytical purposes. Because of the commercial availability and advantageous electrochemical and mechanical properties of BDD (sp<sup>3</sup>-bonded carbon electrode material), it has been used as an alternative to other electrodes commonly used for electroanalytical purposes, such as GC, pyrolytic graphite, and carbon fiber or paste over the

last decade [1,2]. BDD electrode has lots of excellent features for its voltammetric use. An extremely wide potential window in aqueous and non-aqueous electrolytes, very low and stable background current, and high resistance to surface fouling due to weak adsorption are the most important ones [3]. These properties of BDD electrode are affected by controlling its surface termination, most commonly either to predominantly hydrogen or oxygen, which plays an important role in the electroanalytical activity of the electrode for some analytes. The recent reports of our research group have shown that some biologically active compounds important in the field of environmental, agricultural, pharmaceutical and food analyses may satisfactorily be determined by the use of BDD electrode [4–11].

On the other hand, surface-active agents (surfactants) play a very important role in voltammetric studies, not only in solubilizing organic compounds but also by providing specific orientation of the molecules at the electrode interface which heavily influences the electrochemical process of electroactive species [12]. By the use of surfactants in the electroanalysis of a wide range of oxidizable organic compounds, the increase in the sensitivity and selectivity of the determination is achieved [13–15]. Additionally, next to the electrode material, the medium containing

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surfactant can prevent electrode from fouling due to its properties described above. Recently, the detection and/or treatment of surfactants in aquatic environment by electrochemical oxidation on BDD electrode have been studied [16,17]. However, to the best of our knowledge, no literature data were found on the usefulness and behavior of BDD electrode in the presence of surfactants for electroanalysis of organic compounds, except for our very recent works that deal with adsorptive transfer stripping voltammetric determination of two lipophilic substances, namely benzo[a]pyrene, a member of the polycyclic aromatic hydrocarbon class [4], and capsaicin, the most abundant capsaicinoid in peppers [5]. The present study is connected with our previous ones; however, in this case a hydrophilic compound such as Ambroxol (AMB) molecule was selected to explore the application of surfactant effect on the surface of BDD.

AMB, *trans*-4-[(2-amino-3,5-dibromobenzyl)amino]cyclohexanol, pharmacologically active metabolite of bromhexine, has been used as an expectorant agent for almost three decades that leads to bronchial secretion due to its mucolytic properties. Drug is administered as a hydrochloride salt in daily doses of 30–120 mg and is available commercially as granules, tablets and syrups. Similar doses have been given by inhalation, injection or rectally [18]. In humans, the drug is mainly metabolized to 3,5-dibromoanthranilic acid and 6,8-dibromo-3-(*trans*-4-hydroxycyclohexyl)-1,2,3,4-tetrahydroquinazoline. These metabolites which are probably pharmacologically inactive, as well as the parent compound are also converted to conjugates, predominantly glucuronides. After oral administration, AMB is excreted in urine (80%), about 70% of a dose as metabolites and their glucuronide conjugates and 10% unchanged. The remaining 10% seems to be excreted in feces [19].

To date, various methods have been reported for AMB quantification individually or simultaneously in combination with other drugs from pharmaceutical formulations or biological fluids. They are generally based on high-performance liquid chromatography [20–23], UV-visible spectrophotometry [24,25], flow injection analysis [26,27], capillary electrophoresis [28], Raman spectroscopy [29], or potentiometry using membrane selective electrodes [30]. In a very recent work of our research group, first attempt was made to describe colorimetric and atomic absorption spectrometric determination of AMB through ion-pair formation with iron and thiocyanate [31]. Some of the reported methods are prone to some drawbacks, such as expensiveness, complicated and lengthy procedures. AMB is recently included in the British Pharmacopoeia which recommends potentiometric titration as the official method [32].

In addition to its secretagogue action, AMB has also been reported to have non-mucolytic activities such as antioxidant, anti-inflammatory, local anesthetic and surfactant synthesis stimulatory properties in biological systems, due to its ability to scavenge hydroxyl radical, hypochlorous acid, superoxide anion and peroxidative metabolites. Moreover, its some antiviral and antibacterial activities were also shown [33–36]. Further studies are needed to better verify and evaluate its non-mucolytic activities. Thus, it may be helpful to investigate the electrochemical behavior of AMB in order to understand its antioxidant role better as radical scavenger, of consequence, using its additional therapeutic potential to protect the biological targets from oxidative stress. However, a survey of the literature revealed that only four methods have been reported for the voltammetric behavior and determination of AMB [37–40]. All studies were published in the last decade. Previous work of our research group has also demonstrated the effectiveness of GC electrode for the electrochemical oxidation of AMB [38]. The analytical performance of the reported electrodes will be discussed later in detail. On the other hand, a few analytical procedures have been reported for the electrochemical measurement of this drug which

involves high-pressure liquid chromatography [41], and flow injection analysis in combination with amperometric detection [42].

Bearing in mind the relatively new and very limited data on voltammetric determination of AMB and considering the effect of surfactants on the carbon electrode surfaces, the current work aims to throw a more light upon the redox behavior of AMB at surface of BDD in the presence of anionic surfactant and to establish a methodology for its sensitive and rapid determination by using adsorptive stripping voltammetry in pharmaceutical dosage forms and spiked human urine samples. Considerable attention will be given to the demonstration of importance of the electrode pretreatment in surfactant-containing media.

## 2. Material and methods

### 2.1. Apparatus

All voltammetric measurements were carried out with an Autolab PGSTAT 128N potentiostat (EcoChemie, The Netherlands), in connection with a personal computer and controlled by the GPES 4.9 electrochemical software (from EcoChemie). For a better and clearer identification of the original peaks, the experimental square-wave (SW) voltammograms presented were baseline-corrected using the Savicky and Golay filter (level 2) and a moving average application (peak width = 0.01 V) of the GPES software.

Cyclic voltammetry (CV) was employed for preliminary studies on the electrochemical behavior of AMB. Square-wave adsorptive stripping voltammetry (SW-AdSV) was used for the development of electroanalytical methodology and AMB determination in real samples.

A three-electrode-system in a 10-mL one-compartment voltammetric cell was employed consisting a BDD working electrode (Windsor Scientific Ltd.; Ø: 3 mm, diameter), a platinum wire auxiliary electrode, and an Ag/AgCl (3 M NaCl) reference electrode (Model RE-1, BAS, USA) to which all electrode potentials hereinafter are referred. In some cases GC electrode (BAS; Ø: 3 mm, diameter) was also used as working electrode for comparison.

Before each measurement, GC and BDD electrodes were polished manually with aqueous slurry of alumina powder (Ø: 0.01 µm) on a damp smooth polishing cloth (BAS velvet polishing pad), and then ultrasonically cleaned in deionized water thoroughly in order to remove any residual alumina. For BDD electrode, because of its surface doping, this step was always preceded by an anodic polarization process in order to guarantee the decrease of the loss of electrode activity by self-passivating electrode reactions. After the mechanical treatment, BDD electrode was electrochemically activated in an independent electrochemical cell containing the selected supporting electrolyte by applying a potential of +1.8 V for 60 s (in the case of cyclic voltammetric studies, for 180 s).

Successive measurements were carried out by repeating the assay protocol on the working electrode. All measurements were performed in triplicate.

### 2.2. Chemicals

Ambroxol hydrochloride (AMB HCl) in its pure form was received as a gift from Bilim Pharm. Ind. (İstanbul, Turkey), and was used without further purification. Tablet and syrup dosage forms containing the active compound were procured from commercial local pharmacies. Other reagents used were of analytical grade (Merck or Sigma), and their solutions were prepared with deionized water further purified via a Milli-Q system (Millipore). All experiments were carried out at the room temperature of the laboratory. CV measurements were recorded in stock solutions of AMB ( $1 \times 10^{-3}$  M) which were prepared daily in selected

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