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Original Article

Electrochemical oxidation behavior of hydrochlorothiazide on a glassy carbon electrode and its voltammetric determination in pharmaceutical formulations and biological fluids

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ABSTRACT

The electrochemical oxidation behavior of hydrochlorothiazide (HCT) on a glassy carbon as a working electrode was investigated in Britton–Robinson (B–R) buffer pH 3, by using anodic stripping voltammetry (ASV) and cyclic voltammetry (CV). This drug gave a well-defined voltammetric oxidation peak at + 1200 mV versus an Ag/AgCl reference electrode. The electrochemical oxidation process was shown to be irreversible and diffusion controlled, with adsorption characterized over the entire pH range. The optimized conditions, such as accumulation time and potential, scan rate, frequency, pulse amplitude, varying of working electrodes, and instrumental parameters were studied. The calibration graph for HCT was obtained from 4×10^{-6} to 4×10^{-5} M (correlation coefficient = 0.997) using the developed electroanalytical method (ASV). The detection limit of this drug was 4.3×10^{-9} M. ASV and CV techniques with adequate precision and accuracy have been developed and applied for direct determination of HCT in commercial tablets without separation or extraction procedures and biological fluids such as urine and plasma.

1. Introduction

Hydrochlorothiazide (HCT) is a diuretic drug of the thiazide class, which acts by inhibiting the ability of the kidneys to retain water. This reduces the volume of the blood, decreasing blood return to the heart and thus cardiac output and, by other mechanisms, is believed to lower peripheral vascular resistance [1,2]. HCT is a calcium-sparing diuretic; it can help rid the body of excess water, but retain calcium. HCT is frequently used for the treatment of hypertension, congestive heart failure, symptomatic edema, diabetes insipidus, renal tubular acidosis, and the prevention of kidney stones [3]. It is also sometimes used for hypercalciuria, Dent's disease, and Ménière's disease.

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Hydrochlorothiazide is named 6-chloro-1,1-dioxo-3,4-dihydro-2H-1,2,4-benzothiadiazine-7-sulfonamide, as recorded in the IUPAC system. The chemical formula of HCT is $C_7H_8ClN_3O_4S_2$.

Anodic stripping voltammetry (ASV) is the most widely used form of stripping analysis. It is considered a voltammetric method for quantitative determination of trace concentrations for heavy metals, dyes, drugs, and so on. The analyte of interest is electroplated on the working electrode surface during a deposition step, and oxidized from the electrode during the stripping step, so the current is measured during the stripping step. The oxidation of species is recorded as a peak in the current signal at the potential at which the species begins to be oxidized. Thereafter, the stripping step can be either linear, staircase, square-wave, or pulse. Many reviews have been published which illustrate the wide spectrum and scope of ASV applications in the analysis of toxic metals [4], pharmaceutical compounds [5–12], and in forensic science [13].

Cyclic voltammetry (CV) is the most widely used technique for acquiring qualitative information about electrochemical reactions. The power of CV results from its ability to rapidly provide considerable information on the thermodynamics of redox processes and the kinetics of heterogeneous electron transfer reactions and on coupled chemical reactions or absorption processes. CV is often the first experiment performed in an electrochemical study. In particular, it offers a rapid location of redox potentials of the electroactive species, and convenient evaluation of the effect of media on the redox process [14].

ASV is used as an analytical quantitative method capable of monitoring the signal of HCT at low concentration levels. Hence, a wide variety of analytical methods were found to determine trace concentrations of many drugs such as HCT in pharmaceutical formulations and biological fluids [5–12]. Instrumental techniques of chemical analysis were successively used to determine HCT that included spectrophotometry [15–17], chromatography [17,18], liquid chromatography-UV [19], HPLC [20–23], and other electrochemical methods [17,24,25].

The glassy carbon electrode has been used to determine some chemical compounds in different literatures, as modified electrode to enhance electrochemical study [26–30]. An electrochemical detection used to analysis most compounds in the previous studies [31–33].

ASV is considered a powerful electrochemical procedure that enhances the sensitivity of HCT pharmaceutical content in some pharmaceutical formulations. No published article for analysis of HCT using ASV has been reported in the literature. Therefore, this research was carried out to study the ASV behavior of HCT in order to develop an effective and sensitive electrochemical method for determination it in pharmaceutical formulations and biological fluids.

2. Methods

2.1. Apparatus

ASV and CV measurements were carried out using 797 VA Computrace (Metrohm, Herisau, Switzerland) controlled by (VA Computrace 2.0) control software. Anodic stripping and cyclic voltammograms were printed via a HP color laserjet CP1215 printer (China). A three electrode system was used, including a glassy carbon electrode as the working electrode. pH values were studied using a Hanna instrument pH211 (Romania). An Oxford adjustable micropipette (Huawei, Ireland) was used to measure microliter volumes of the drug standard solutions. The Labofuge 200 instrument, Heraeus Sepatech (Germany) was used to centrifuge the urine and plasma samples, which were then suitable for voltammetric analysis.

2.2. Reagents

The chemicals used were of analytical reagent grade and used without further purification. HCT stock solution of 1×10^{-2} M was prepared by dissolving the appropriate amount of HCT in distilled water in a 25 mL volumetric flask. The drug stock solution was stored in a dark place. The standard solutions of HCT with lower concentrations were prepared by diluting the stock solution with distilled water daily. Britton–Robinson (B–R), carbonate, phosphate, and acetate supporting buffers were prepared for the voltammetric analysis of HCT [34].

2.3. Procedure

The general procedure which applied for getting all voltammograms was as follows: 10 mL of B–R buffer pH 3 was injected in to a dry and clean voltammetric cell and then a required standard solution of HCT was added. All buffer solutions were purged with a nitrogen gas for nearly 3 minutes initially, while these solutions were stirred. The deposition potential of 0.0 V versus an Ag/AgCl reference electrode was applied to a glassy carbon electrode while the solution was stirred for 20 seconds. Anodic scans were carried out over the range 800–1700 mV. The voltammetric measurements were performed at room temperature (25°C).

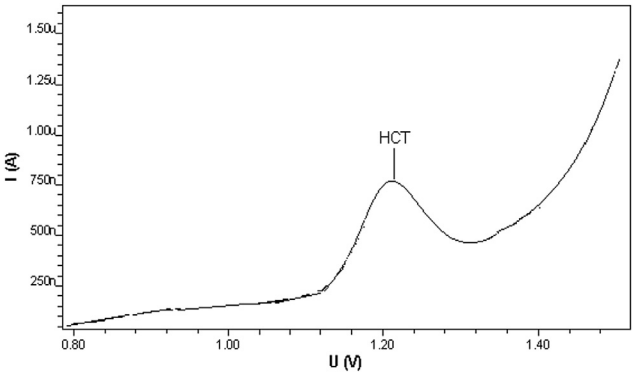

Fig. 1 – Anodic stripping voltammogram of hydrochlorothiazide (HCT) in Britton–Robinson buffer at pH 3, experimental conditions: t_{acc} = 20 seconds, E_{acc} = 0.0 V, scan rate = 150 mV s^{-1}, drug concentration = 6×10^{-6} M.