



Efficient electrocatalytic oxidation and determination of isoniazid on carbon ionic liquid electrode modified with electrodeposited palladium nanoparticles



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ABSTRACT

Palladium nanoparticles (PdNPs) were electrodeposited on the carbon ionic liquid electrode (CILE) for evaluating the assemblage for electrochemical determination of isoniazid (INH). The surface morphological investigation of the fabricated electrode by using scanning electron microscopy indicated efficient electrodeposition of PdNPs with the particle diameter of 30–100 nm when a constant potential of -0.2 V for duration of 60 s was applied. Electrocatalytic oxidation of isoniazid at this electrode was investigated in phosphate buffer solution (pH 7.0) using cyclic voltammetry. A highly reproducible and well-defined peak with a high electrocatalytic peak current was obtained for INH at a low overpotential of $+0.34$ V versus Ag/AgCl. Under optimized experimental conditions, linear relationship between anodic peak current and INH concentration in the ranges of 5.0×10^{-6} – 1.0×10^{-4} and 1.5×10^{-4} – 2.6×10^{-3} mol L⁻¹ INH with a detection limit of 4.7×10^{-7} mol L⁻¹ was obtained. The procedure was successfully applied for analysis of INH in human blood serum and pharmaceutical samples.

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1. Introduction

Pyridine-4-carboxylic acid hydrazide (isonicotinic acid hydrazide, INH) commercially known as isoniazid (Fig. 1), is widely used for treatment and also prevention of tuberculosis, the second-most common cause of death from infectious disease [1,2]. Isoniazid is used along with rifampicin and streptomycin for the chemotherapy of the disease [3]. From the World Health Organization (WHO) recommendation, a daily dose should be 4–6 mg/kg of the body weight [4]. However, a high concentration of isoniazid in the human body can lead to epilepsy, liver function failure, and even death [5]. Therefore, the control of isoniazid level in human body fluids is very important in clinical chemistry. On the other hand, small concentration difference between effectively therapeutic and toxic dosages makes the development of a simple, rapid, selective and highly sensitive method vital for determination of INH level in body fluids. It is also essential for pharmaceutical industries to determine the INH level in their products for quality control purposes.

Many analytical techniques have been reported for determination of isoniazid such as: ultraviolet–visible spectrophotometry [6,7], fluorimetry [8,9], high-performance liquid chromatography [10,11], gas chromatography [12], capillary electrophoresis [13], chemiluminescence spectroscopy [14,15], and electroanalytical methods [16–21]. The chromatographic methods commonly use expensive instrumentation

and high volume of organic solvents, while the spectroscopic methods have their own difficulties where they usually need additional reagents and suffer from complicated procedures. Electroanalytical techniques are of particular advantage because of their practicality, simplicity, low-cost, good sensitivity, precision, and rapidity for real-time monitoring of drugs and biological compounds. However, electrochemical oxidation of INH at a bare glassy carbon (GCE) electrode and traditionally carbon paste electrodes (CPE) suffers from electrode fouling, sluggish electron transfer kinetics and large oxidation overpotential [22]. The usual approach in overcoming these limitations is modification of the electrode surface with chemical modifiers such as polymers [23], metal nanoparticles [24], carbon nanotube [25] and functionalized graphene [26].

In general, a nanoparticle-modified electrode can provide high effective surface area and mass transport. It also can catalyze and control the local microenvironment in comparison with macroelectrodes [27]. However, the electrode performance is governed by fabrication route, substrate materials as well as the nature of nanoparticle itself. The modification can be attained either by incorporation of chemically-synthesized nanoparticles into the electrode paste or electrodeposition of metal nanoparticles on the electrode surface. The electrodeposition is preferred, because the particles with higher purity, more controllable and lower particle size distribution can be achieved in shorter time scales [28,29]. Nobel metal nanoparticles have good mechanical and electrical properties, high stability and low aggregation. These properties improve their usage as catalysts for electrochemical application. Palladium (Pd) nanoparticles possess unique property and have been used as electrode modifier for electrocatalytic purposes [30].

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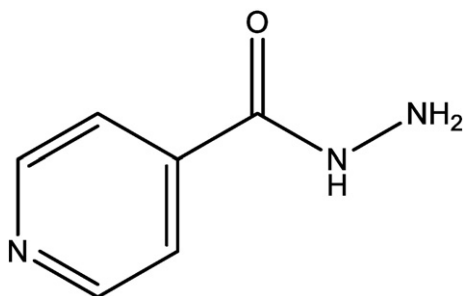


Fig. 1. Chemical structure of the INH.

The nature of substrate can immensely affect the morphologies of nanoparticles as well as their electrocatalytic properties [31]. From the first introduction of carbon ionic liquid electrode (CILE) by Safavi et al. [32], it has been widely used for electrochemical determination of various pharmaceutical and biological compounds owing to its high electron transfer rate, high conductivity and antifouling effect [33]. Moreover, it has been recognized to be a versatile substrate to be modified with different metal and metal oxide nanoparticles for electrocatalytic oxidation of many compounds [34–36]. For example, the electrocatalytic effect of palladium nanoparticles, electrodeposited on the carbon ionic liquid electrode (PdNPs/CILE), has been studied for oxygen reduction [30], formaldehyde oxidation [37] and hydrazine oxidation [38].

Herein, CILE, having N-octylpyridinium hexafluorophosphate as binder was used to be modified with Pd nanoparticles which revealed high electrocatalytic activity towards the oxidation of INH. Under optimized experimental conditions, INH showed a well-defined oxidation peak with high current at the modified electrode. The modified electrode provided a low overpotential in comparison with traditional electrodes. High sensitivity towards INH, allowed successful using of the electrode for determination of this drug at low concentration level in both biological and pharmaceutical samples.

2. Experimental

2.1. Reagents

Isoniazid, 1-iodooctane, pyridine, ammonium hexafluorophosphate, and graphite powder (mesh size: <math><100\ \mu\text{m}</math>) were purchased from Merck. Palladium(II) chloride was obtained from Uni-Chem and sulfuric acid (98%, AR grade) from Aldrich. All chemicals were of the reagent

grade. Reagent solutions were prepared freshly before using. Phosphate buffer solution (PBS pH 7.0) was utilized as supporting electrolyte.

2.2. Electrode preparation

N-octylpyridinium iodide ($[\text{C}_8\text{Py}] [\text{I}]$) was synthesized and purified using a previously reported procedure [39]. Carbon ionic liquid electrode was prepared by hand-mixing in a mortar. The graphite powder and IL ($[\text{C}_8\text{Py}][\text{PF}_6]$) were in a 50:50 weight ratio [40]. A portion of the resulting paste was packed firmly into the cavity (1.6 mm diameter) of a Teflon holder. The electrode was then heated in an oven at $80\ ^\circ\text{C}$ for 2 min. After heating, the electrode was left to cool to room temperature. The electrical contact was established via a stainless steel handle. A new surface was obtained by polishing the electrode surface by rubbing it against a smoothed-surface paper. Palladium nanoparticles were electrodeposited on the CILE (PdNPs/CILE) at the optimum potential of $-0.2\ \text{V}$ applied for 60 s in 10.0 mL of $1.0\ \text{mmol L}^{-1}$ PdCl_2 (in $1.0\ \text{mol L}^{-1}$ H_2SO_4 solution). The electrode surface was washed with distilled water afterward.

2.3. Apparatus

Voltammetric measurements were performed with an electrochemical analyzer PGSTAT302N (Metrohm Autolab B.V., Utrecht, The Netherlands) equipped with NOVA 1.7 software. The electrochemical cell was assembled with a conventional three-electrode system; an Ag/AgCl/KCl (sat'd) as the reference electrode (Metrohm), a platinum wire as the counter electrode and a palladium modified carbon ionic liquid electrode with 1.6 mm diameter as the working electrode. A Metrohm 780 pH meter was used for monitoring the pH values. The surface morphological characterization of this electrode was studied with a KYKY-EM3200 SEM operating at 25.0 kV. The digital ultrasonic cleaner model CD-4820 (170 W, 50 Hz) was used to sonicate the samples.

2.4. Sample preparation

Five isoniazid tablets (300 mg per tablet, Darou Pakhsh Co., Tehran, Iran) were weighed and micronized in an agate mortar. A weight of 150 mg of the ground material was transferred into 10 mL volumetric flask and filled with $0.1\ \text{mol L}^{-1}$ phosphate buffer solution (pH 7.0;) then was sonicated in ultrasonic bath for 15 min before being filtered through a filter paper. The obtained solution was directly analyzed without doing any pretreatment or extraction step except diluting with the supporting electrolyte.

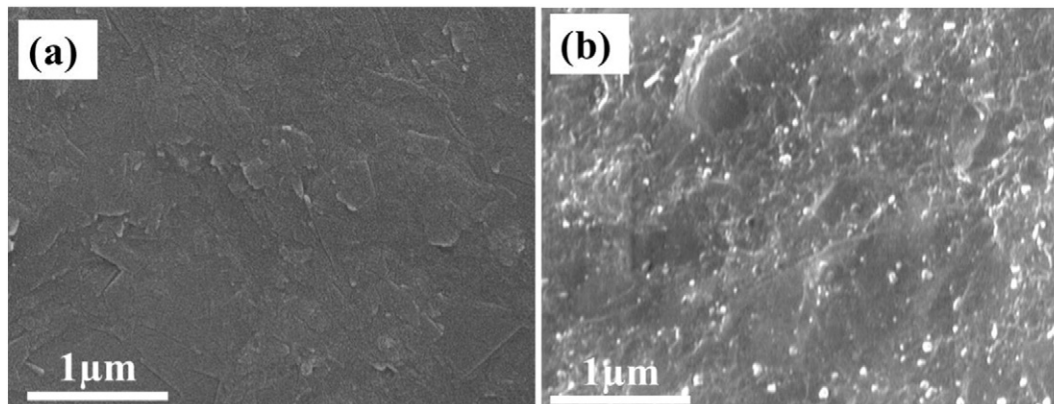


Fig. 2. The SEM image of (a), CILE; (b), PdNPs/CILE.

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