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## Low Cost Hydrogen Peroxide Sensor from Manganese Oxides Modified Pencil Graphite Electrode

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### Abstract

Electrodeposition of manganese oxides film onto the cheap pencil graphite electrode using potassium permanganate precursor provides the good alternative method of fabrication the low cost hydrogen peroxide sensor. Effect of deposition potential, deposition time and concentration of potassium permanganate were investigated. The modified electrode displayed electrocatalytic activity towards the oxidation of hydrogen peroxide in alkaline medium. Amperometric detection of hydrogen peroxide in ammonium buffer pH 9.0 is possible at the operation potential of +0.50 V vs Ag/AgCl instead of over +0.80V vs Ag/AgCl with unmodified electrode. Linear concentration range between 0.50-138 ppm of hydrogen peroxide was obtained with a detection limit of 0.28 ppm.

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**Keywords:** Hydrogen peroxide sensor; pencil graphite electrode; manganese oxides

### 1. Introduction

H<sub>2</sub>O<sub>2</sub> is an important species which has various applications as oxidizing agent, antimicrobial agent and sterilizing agent in industrial, clinical and pharmaceutical processes. It is also a by-product of oxidases enzymatic reactions and has been widely employed in fabrication of many biosensors. Electrochemical detection by direct oxidation of H<sub>2</sub>O<sub>2</sub> on common working electrode materials such as glassy carbon or noble metals requires highly positive potentials which are prone to many oxidizable components. Therefore, many types of mediator were introduced as modifier for electrode surface modification in order to enhance the analytical signal and/or lower the reaction overvoltage. Manganese dioxide and mixed valences of manganese oxides have been used as modifiers with carbon based electrodes such as carbon paste electrode and screen-printed carbon electrode and successfully employed as H<sub>2</sub>O<sub>2</sub> and glucose sensors [1-5]. In alkaline medium, the manganese dioxide behaves as mediator by

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turning  $\text{H}_2\text{O}_2$  to  $\text{O}_2$  and  $\text{H}_2\text{O}$  while it is also reduced to lower oxidation state. Then  $\text{MnO}_2$  can be regenerated electrochemically yielding the catalytic current as the sensor signaling [3].

The aim of this work is to develop a simple and low cost  $\text{H}_2\text{O}_2$  sensor based on pencil graphite electrode (PE) modified with manganese oxides which are simply cathodic deposited from the cheap and easy available potassium permanganate solution.

## 2. Materials and Methods

### 2.1. Materials

Aqueous solution of  $\text{H}_2\text{O}_2$  was prepared freshly each day from stock 30 % (w/w)  $\text{H}_2\text{O}_2$  (Merck-Schuchardt OHG, Hohenbrunn, Germany). Other reagents were from Sigma-Aldrich (Dublin, Ireland). Deionized water was used throughout the experimental work. Ammonia /ammonium chloride buffers pH 8-10 were prepared from 0.2 M aqueous  $\text{NH}_3$  solution and 0.2 M  $\text{NH}_4\text{Cl}$ . Phosphate buffer pH 7.4 was prepared from proper volumes of 0.2 M  $\text{NaH}_2\text{PO}_4$  and 0.2 M  $\text{Na}_2\text{HPO}_4$ . Potassium permanganate (Ajax Finechem Pty Ltd., Australia), prepared as solutions of the concentration range 0.001-0.05M was used for manganese oxides film coating on graphite pencil lead (2B, STEADLER).

### 2.2 Fabrication of the manganese oxides modified PE

The graphite pencil lead of 35 mm length was wiring to a copper rod (0.5 mm o.d., 45 mm length) with a thin copper wire and applied silver paint (SPI supplies) for electrical connection. A micropipette tip was used as electrode body, sealed with PVC glue and the exposed pencil rod was 10 mm length. Geometric area of the exposed part of PE was about 16 mm<sup>2</sup>. The PE was soaked for 12 h in 50%  $\text{HNO}_3$  before 2 h in ethanol and rinsed thoroughly with deionized water. The pretreated PE was then coated with manganese oxides film by cathodic deposition from potassium permanganate solution at a constant stirring rate of 1000 rpm and a fix interval time. The coated PE was washed with deionised water to remove potassium permanganate residue and kept air dried before use. Controlled potential electrolysis, voltammetric and amperometric measurements were performed by the AUTOLAB PGSTAT101 (Ecochemie, The Netherlands) with NOVA 1.8 software.

## 3. Result and Discussion

### 3.1 Voltammetric behavior of the modified PE

Cyclic voltammograms of the coated PE in  $\text{NH}_3/\text{NH}_4\text{Cl}$  buffer pH 9.0 and the bared PE recorded in  $\text{KMnO}_4$  solution (Fig.1a) imply to the similar redox activity of electrode reaction. The reduction of permanganate ion to lower oxidation states of manganese on the PE started from 0.40 V vs Ag/AgCl. At the successive second scan in wider potential range shows the multistep electrode reaction. For the coated PE (from controlled potential electrolysis at +0.10 V) revealed a good redox couple of the coated manganese oxides at  $E_{pc} = 0.02$  V and  $E_{pa} = 0.25$  V which could be employed for catalytic reaction of  $\text{H}_2\text{O}_2$  while the bare PE gave negligible response. From linear sweep voltammograms (Fig.1b), the oxidation current of the coated PE arises from 0.10 V vs Ag/AgCl and it was increased obviously in the presence of  $\text{H}_2\text{O}_2$  (6.5 mM) comparing to the unmodified PE. Thus, oxidation of  $\text{H}_2\text{O}_2$  can be detected at less positive potential. Different peak position between each modified PE might due to different structure orientation and mixed valences of manganese oxides deposited on the PE. Hence, the operating potential at 0.50 V vs Ag/AgCl was chosen for sensing  $\text{H}_2\text{O}_2$  with the modified PE.

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