



# Non-enzymatic amperometric glucose biosensor based on nickel hexacyanoferrate nanoparticle film modified electrodes

Xiaoyan Wang<sup>a</sup>, Yun Zhang<sup>a,\*</sup>, Craig E. Banks<sup>b</sup>, Qiyuan Chen<sup>c</sup>, Xiaobo Ji<sup>c,\*</sup>

<sup>a</sup> College of Material Science and Engineering, Sichuan University, Chengdu, Sichuan 610064, China

<sup>b</sup> Faculty of Science and Engineering, School of Biology, Chemistry and Health Science, Division of Chemistry and Materials, Manchester Metropolitan University, Chester Street, Manchester M1 5GD, Lancashire, UK

<sup>c</sup> Institute of Metallurgy and Applied Physical Chemistry, College of Chemistry and Chemical Engineering, Central South University, Changsha 410083, China

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## ABSTRACT

A non-enzymatic amperometric glucose biosensor based on the modification of functional nickel hexacyanoferrate nanoparticles was prepared via electrochemical deposition. The electrochemical deposition of the nickel hexacyanoferrate nanoparticles was obtained by potential cycling in a solution containing nickel (II) and hexacyanoferrate (III) producing a modified surface with a high degree of uniformity. The modified electrode is exemplified towards the non-enzymatic sensing of glucose where using cyclic voltammetry and amperometry, low micro-molar up to milli-molar glucose concentrations are readily detectable. The non-enzymatic sensing of glucose also shows a modest selectivity over ascorbic acid. This platform offers a novel route for glucose sensors with wide analytical applications.

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## 1. Introduction

The development and fabrication of cost-effective, simple, accurate, portable and rapid sensors for glucose are socially important aiding diabetics globally which represents roughly 5% of the world's population [1]. Certain areas of medical, food and environmental analysis require rapid and inexpensive monitoring methods for glucose in blood and food [2,3]. The measurement of glucose concentration in blood is critical for diagnosis of diabetes mellitus in order to avoid some of the long-term diabetic syndromes such as retinal and kidney damage [4,5].

In responses to the needs for frequent or continuous monitoring of glucose in diabetes there is a need for the development of improved sensors [6]. Nowadays the pursuit of non-enzymatic glucose sensing with rapid response and precise measurement is a vigorous and competitive area of research. The direct oxidation of glucose by different electrodes in the absence of enzyme has been studied [7–12]. Various metal electrodes such as Pt, Au, and Ag have been proved to be highly electro-active in the anodic oxidation of glucose requiring a high overpotential. However, these can be problematic due to poisoning/fouling of the electrode surface, especially at gold and platinum electrodes. Additionally the cost of these pre-

cious metals needs to be considered. As a consequence, there are increasing interests on the fabrication of modified electrodes with low operating potential by enhancing electron transfer kinetics. In comparison, a few transition metal complexes, including the various transition metal hexacyanoferrate modified electrodes, have been shown to be efficient electro-catalysts for anodic oxidation of glucose, giving enhanced stability towards the target analyte, with low detection limits and wide analytical ranges achievable [13–18]. In this area problems from the instability of modified electrodes are evident and limit widespread implementation.

Consequently in this paper, we describe a simple route to the production of uniform functional nickel hexacyanoferrate nanoparticles using electrochemical deposition. The preparation method is simple and nickel hexacyanoferrate nanoparticles may be readily formed on a glassy carbon electrode surface constructing a simple, economical and accurate amperometric sensor for glucose. The non-enzymatic glucose biosensor based on nickel hexacyanoferrate (NiHCF) modified electrodes provides a prominent augmentation of response current toward glucose with a good stability and reproducibility.

## 2. Experimental

### 2.1. Chemical reagents

All chemicals were of analytical grade from Changzheng (Chengdu, China) and were used without further purification. All

\* Corresponding authors. Tel.: +86 28 8541 0272; fax: +86 28 8541 0272.

E-mail addresses: [y.zhang@scu.edu.cn](mailto:y.zhang@scu.edu.cn) (Y. Zhang), [xji.csu.edu@gmail.com](mailto:xji.csu.edu@gmail.com), [xiaobo.ji@gmail.com](mailto:xiaobo.ji@gmail.com) (X. Ji).

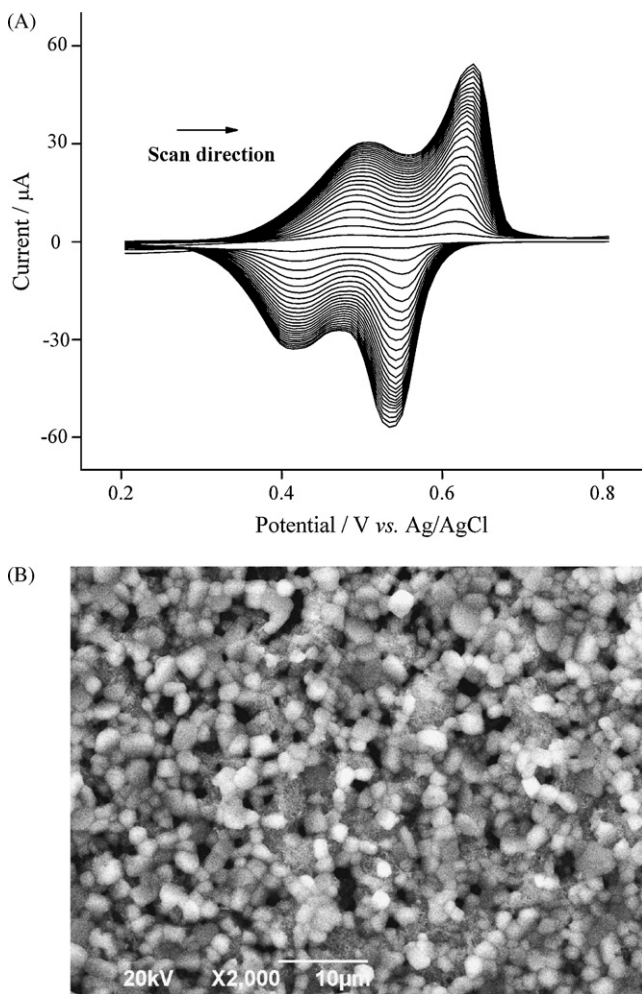
solutions were prepared with double distilled water, and degassed with oxygen free nitrogen. All experiments were carried out at a temperature of  $295 \pm 3$  K.

## 2.2. Instrumental

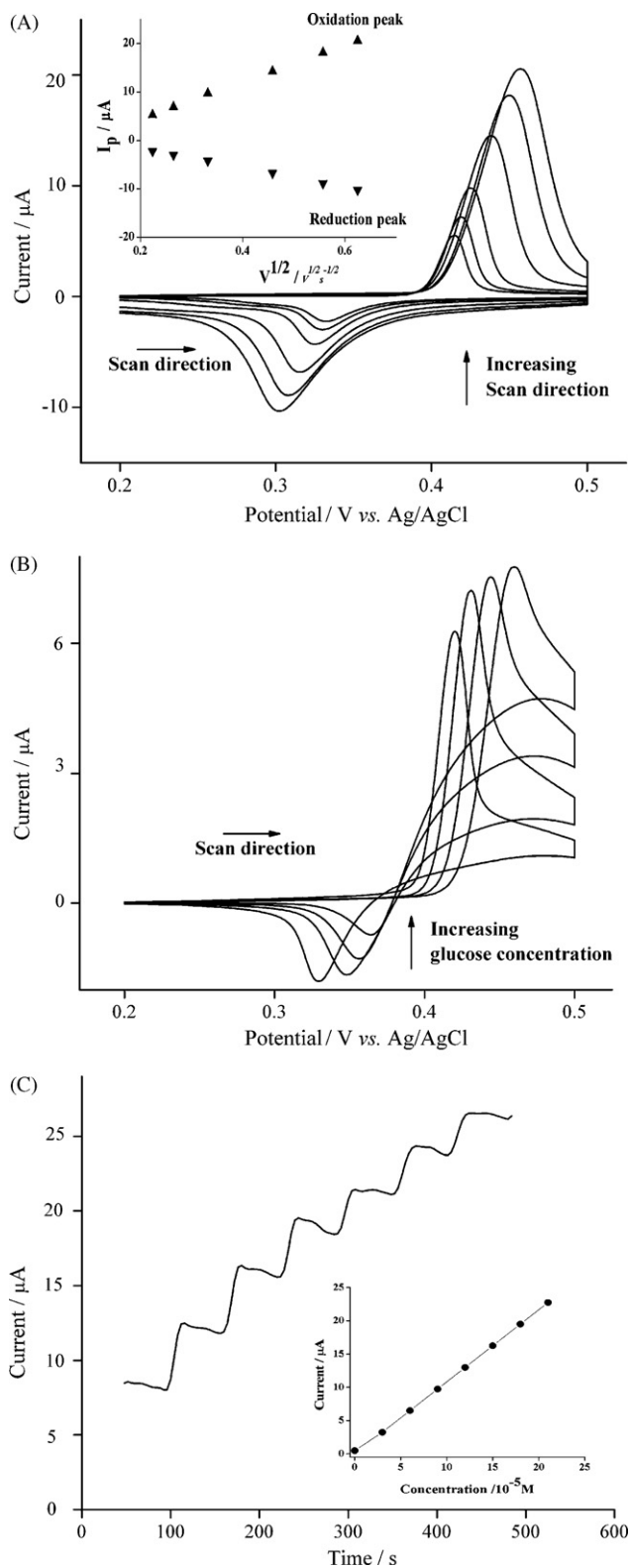
Electrochemical measurements were performed using a LK9805 electrochemical analyzer (Tianjin, China). A glassy carbon electrode (GCE) was used as working electrode with a bright platinum foil as a counter electrode and a Ag/AgCl reference electrode completing the circuit. Scanning electron micrographs (SEM) were obtained with a scanning electron microscope (SEM, Hitachi S-800, 20 kV).

## 2.3. Preparation of the modified electrode

Electro-deposition was performed using solutions containing 0.05 M KCl, 0.5 mM  $K_3[Fe(CN)_6]$  and 1.2 mM  $NiCl_2$ . Unless otherwise stated, the procedure involved 25 full voltammetric cycles at  $50 \text{ mV s}^{-1}$  in a potential range from  $-0.1$  to  $1.0$  V. NiHCF film was fabricated on GCE substrates with a diameter 5 mm. Prior to the film deposition, the GCE was polished on alumina lapping compounds (BDH) of decreasing sizes ( $5\text{--}0.1 \mu\text{m}$ ) on soft lapping pads.



**Fig. 1.** (A) Cyclic voltammograms (25 cycles) of NiHCF films at the glassy carbon electrode recorded in  $0.5 \text{ mM } K_3[Fe(CN)_6] + 0.05 \text{ M KCl} + 1.2 \text{ mM } NiCl_2$  solutions with Ag/AgCl reference electrode. Scan rate:  $50 \text{ mV s}^{-1}$ . (B) SEM images of the NiHCF film.



**Fig. 2.** (A) Cyclic voltammetric responses of NiHCF modified electrodes at various scan rates from  $50$  to  $500 \text{ mV s}^{-1}$  in  $0.5 \text{ M NaOH}$  solution. Inset: The plot of the oxidation peak currents and the reduction currents for NiHCF modified electrodes against the square root of scan rates. (B) Electrochemical responses of NiHCF modified electrodes to glucose in  $0.5 \text{ M NaOH}$  solution: (a) without glucose (b)  $2 \text{ mM}$  glucose (c)  $4 \text{ mM}$  glucose (d)  $8 \text{ mM}$  glucose. Scan rate:  $50 \text{ mV s}^{-1}$ . (C) Amperometric responses of NiHCF modified GCE to increasing the glucose concentration with applied potential  $0.5 \text{ V vs. Ag/AgCl}$ ; Inset: Corresponding calibration curve at NiHCF modified glucose biosensor.

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