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Novel alkaline-reduced cuprous oxide/graphene nanocomposites for non-enzymatic amperometric glucose sensor application

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ABSTRACT

This paper presents the fabrication of a highly sensitive and selective glucose sensor based on cuprous oxide/graphene nanocomposites-modified glassy carbon electrode (Cu_2O/graphene/GCE). The Cu_2O/graphene nanocomposites were synthesized based on a simple and straightforward chemical reduction process in alkaline aqueous solution using sodium carbonate as reductant. The size and shape of Cu_2O nanoparticles on graphene sheets can be controlled by changing the amount of graphene oxide added during reaction. The electrochemical properties of Cu_2O/graphene/GCE in 0.1 M phosphate buffer solution were investigated by cyclic voltammetry and electrochemical impedance spectroscopy. It was found that the pH, concentration of supporting electrolyte, and scan rate had very crucial effect on the sensitivity of prepared sensor towards glucose oxidation. At an applied potential of +0.50 V, the Cu_2O/graphene/GCE presented a high sensitivity of 1330.05 μA mM^{-1} cm^{-2} and fast response (within 3 s). The amperometric non-enzymatic glucose sensor developed had a linear relationship from 0.01 mM to 3.0 mM glucose and detection limit of 0.36 μM. In the presence of ascorbic acid, uric acid, dopamine, chloride and citrate ion and other carbohydrates, the interferences were negligible. The proposed sensor was successfully applied for the determination of glucose concentration in real human blood samples.

1. Introduction

Glucose is an essential nutrient for human life as the heat released by its oxidation is an important source of energy. Blood glucose level is also important to indicate the human health conditions and blood glucose level of the healthy people ranges from 3.9 to 6.0 mmol L^{-1} [1]. The electrochemical glucose sensor has been developed for application in various fields such as in clinical [2], food safety [3], environmental [4] and industrial [5]. In general, the sensors are reputed with good sensitivity and selectivity, low cost, robust and can be miniaturized for small sample's volumes [6,7]. Usually, a glucose sensor is synonymous with the glucose oxidase enzyme (GO_x) modified sensor. Although such sensors have high sensitivity their performance is restricted by the enzyme's immobilization process, activity and stability [8].

In order to combat the bottlenecks of enzymatic glucose sensors, extensive researches have been done to develop non-enzymatic sensors, which implement the direct electrocatalytic oxidation of glucose. Several materials such as platinum, gold, copper, silver, bismuth and mercury which can catalyze the oxidation of glucose were reported [9]. Out of all these materials, cuprous oxide (Cu_2O) based materials are shown to facilitate the good potential for the oxidation of glucose. Cu_2O is a semiconductor with good characteristics such as being non-toxic, possesses electrochemical activity, chemical stability, high electron communication features and involves low cost synthesis process. Li et al. [10] fabricated Cu_2O nanoparticles for glucose sensing application with a detection limit and sensitivity of 47.2 μM and 0.19 μA/mM/cm^2, respectively. Khan et al. [11] discovered that Cu_2O shuriken-like nanostructures exhibited an electrocatalytic performance for glucose detection with a sensitivity of 0.933 mA/mM cm^2. These findings however showed that Cu_2O particles alone are poorly conductive and not very sensitive towards glucose oxidation.

As time passed, it was proposed that fabricating metal oxide nanostructures onto graphene surface by suitable reaction could resolve the problems [12–14]. Liu et al. [15] demonstrated the excellent electrocatalytic activity of Cu_2O/graphene nanocomposites in comparison with graphene and Cu_2O as the electrode modifying material in the glucose oxidation, respectively. The nanocomposites exhibited typical current response to the oxidation of glucose sensor, while no obvious oxidation peak for glucose was found at the graphene and Cu_2O nanoparticles modified electrode. This finding is in excellent agreement with several earlier reports that have clearly indicated that metal oxide/graphene nanocomposites have displayed remarkable improvements in electrocatalytic activity and stability towards some crucial electrochemical reactions in amperometric glucose sensors.

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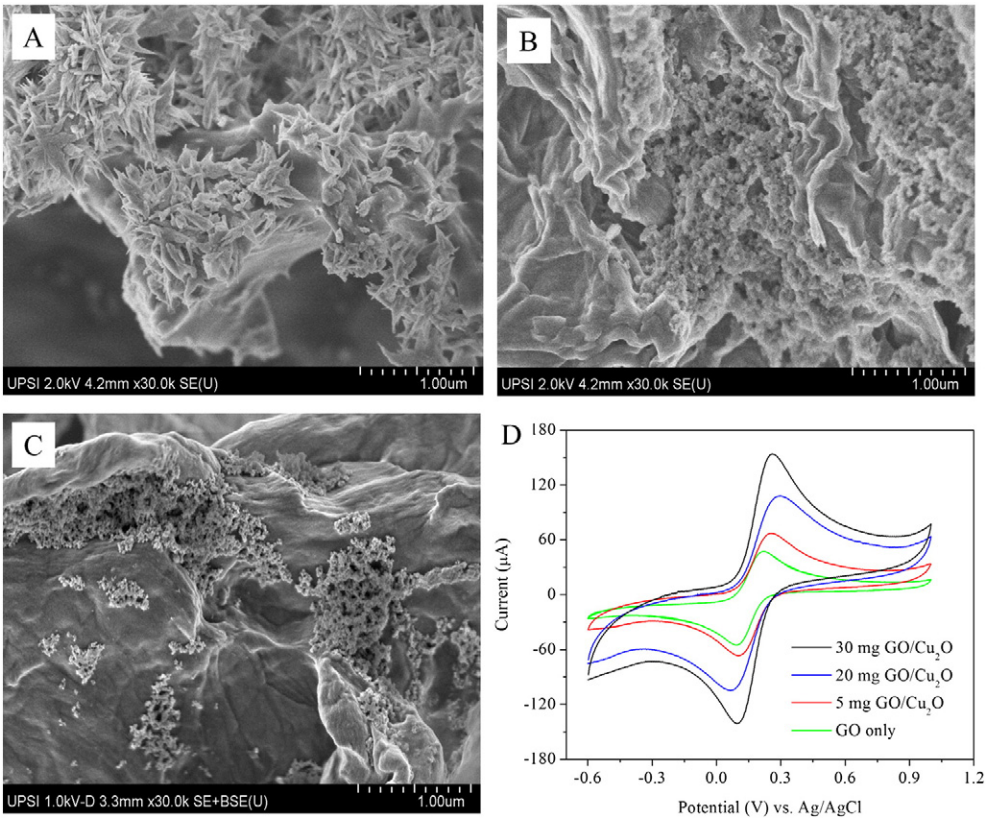

Fig. 1. FESEM images of Cu_2O/graphene nanocomposites at different amounts of GO introduced to the Cu_2O, where (A) 5 mg, (B) 20 mg, and (C) 30 mg GO, and (D) CV curves of the Cu_2O/graphene nanocomposites at different amounts of GO introduced to the Cu_2O.

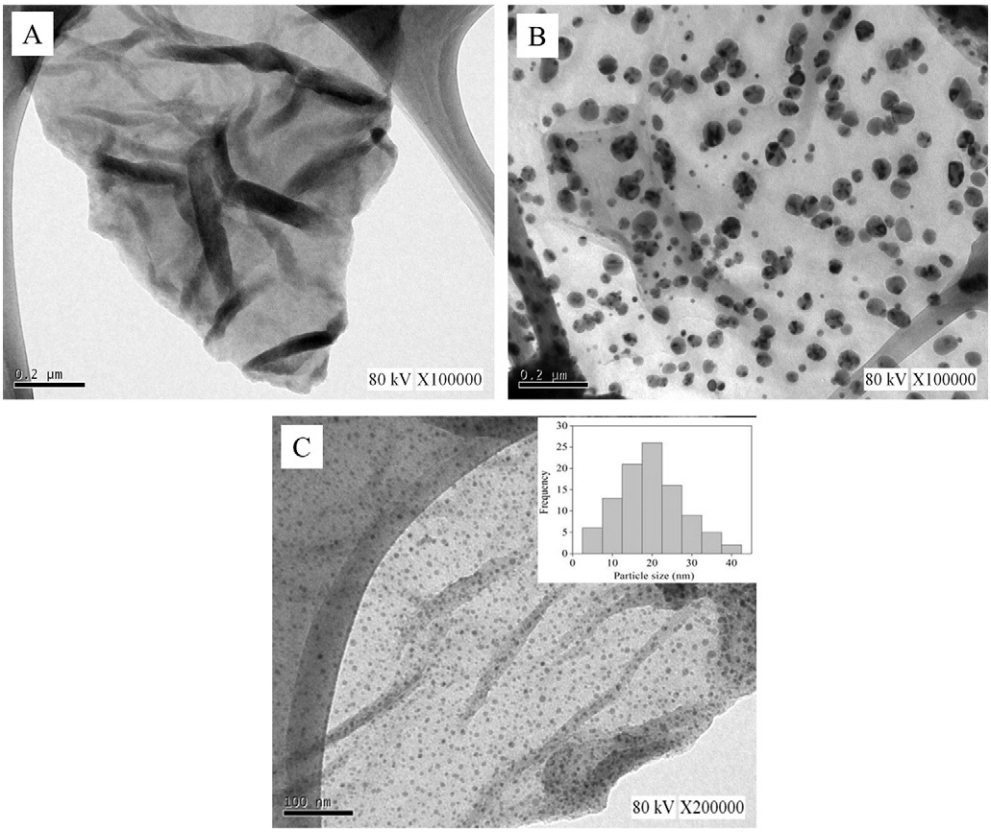

Fig. 2. TEM image of (A) graphene, (B) Cu_2O nanoparticles, and (C) Cu_2O/graphene nanocomposites.