DOCTORAL THESIS

Development of amperometric biosensors with carbon nanotube composite materials

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Date of Award:
2008

Development of Amperometric Biosensors with Carbon Nanotube Composite Materials

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A thesis submitted in partial fulfillment of the requirements

for the degree of

Doctor of Philosophy

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Hong Kong Baptist University

May 2008

Abstract

Carbon nanotubes (CNTs) have attracted much attention since their discovery. CNTs offer unique mechanical, chemical, physical and electronic properties. Many investigations have demonstrated that CNTs possess good electrocatalytic activities. This thesis work mainly focuses on the development of amperometric glucose biosensors utilizing carbon nanotubes and CNT composite materials. The electrochemical behaviors of the CNT composite materials have been investigated and the biosensor performance was explored.

CNT-modified electrodes with different types of CNT materials have been prepared. Electrochemical reactions involving ferricyanide and hydrogen peroxide were examined at the CNT modified electrodes. Glucose biosensors have been prepared with different types of CNT materials and the amperometric behaviors for glucose determination have been examined. Single walled carbon nanotube biosensor usually exhibited a wider dynamic range and higher sensitivity for glucose determination.

Carbon nanotubes have been combined with different materials, including Prussian blue inorganic nanoparticles, copper nanoparticles and gold colloid, to prepare different CNT-based electrode substrates. Different polymeric materials were utilized as enzyme entrapment agents. Chitosan, PDDA and poly(toluidine blue O) films have been employed. Amperometric glucose biosensors can be prepared by using different CNT composite materials and the biosensor performance was investigated.

A highly sensitive glucose biosensor has been fabricated by electrochemical deposition of chitosan (CHIT) and glucose oxidase (GOD) on single-walled carbon nanotube (SWCNT) modified glassy carbon electrode with copper nanoparticles. The presence of SWCNT and copper nanoparticles offered improvement in electron transfer at the electrode

surface. Electrodeposited chitosan film provided biocompatible matrix for the immobilization of enzymes. The biosensor displayed linear response to glucose from 0.02 to 5 mM with a sensitivity of 22.4 mA M^{-1} cm^{-2} at an applied potential of +0.65 V (*vs.* Ag|AgCl). On the other hand, Au colloid was prepared and assembled on the CNT surface by a positively charged polymer PDDA. The direct electrochemistry of GOD on the GC/CNT+Au/PDDA modified electrode was observed.

A mediator glucose biosensor has been constructed by immobilizing glucose oxidase at electropolymerized poly(toluidine blue O) film on carbon nanotube modified glass carbon electrode. Toluidine blue O moieties served as mediators for direct oxidation of glucose oxidase and as polymeric network to maintain the biosensor activity. The detection potential could be decreased to –0.1 V (vs. Ag|AgCl), where common interferences such as ascorbic acid, uric acid and Acetaminophen were not oxidized to cause interferences. The amperometric glucose biosensor offered a sensitivity of 5.09 $mAM^{-1}cm^{-2}$ and a wider linear range of 1-18 mM.

Utilizing the unique properties of multi-walled carbon nanotubes, an amperometric biosensor based on horseradish peroxidase (HRP) was fabricated. Experimental results indicated that there was direct electron transfer between HRP enzyme and CNT-modified glassy carbon electrode. Electrocatalytic reduction of hydrogen peroxide at the GC/CNT/HRP/Nafion electrode was investigated. Additionally, a mediator-free bienzyme glucose biosensor based on horseradish peroxidase and glucose oxidase was constructed. A zero applied potential offered an improvement in the selectivity of the biosensor. Under the optimized conditions, the bienzyme biosensor exhibited a high sensitivity for glucose detection.

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