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Amperometric biosensors based on carbon nanotubes with different polymer coatings

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Abstract

Carbon nanotubes (CNTs) have emerged as a new class of nanomaterials that exhibit a unique combination of excellent mechanical, electrical and electrochemical properties, which have stimulated increasing interest in the application of CNTs as important components in biosensor developments.

Multi-walled CNT (MWNT)-modified glassy carbon electrode (GCE) with different types of MWNT materials, including regular MWNTs, MWNTs with carboxyl (–COOH) and hydroxyl (–OH) functionalities under both purified (acid-treated) and unpurified conditions, have been prepared. Electrochemical reactions involving ferricyanide and hydrogen peroxide were evaluated at the MWNT-modified electrodes. Glucose biosensors have also been fabricated with different types of MWNT materials and the amperometric behaviors of the modified electrodes for glucose determination have been examined. Experimental results revealed that the purified MWNT materials offered better electron transfer properties and electrocatalytic activities to hydrogen peroxide and glucose than the corresponding unpurified MWNTs. Moreover, electrodes modified with the purified MWNT and MWNT-OH materials exhibited slightly improved electrochemical performances when compared with the purified MWNT-COOH material for the substrates (H_2O_2 and glucose) evaluated.

The utilization of CNT-based biosensors for the amperometric detection of glucose by casting the MWNT-modified GCE with different polymeric enzyme entrapment materials has been investigated. The polymeric materials examined in this thesis work include the non-conducting poly(*o*-phenylenediamine) (P*o*PD) film, as well as the conducting poly(toluidine blue O) (PTBO), polypyrrole (PPy) and Nafion films. Glucose oxidase (GOx) was co-immobilized on electrode

either by electrochemical polymerization or enzyme entrapment. The electrochemical behaviors of different biosensors were explored and compared in terms of sensitivity and linear range for glucose detection under three particularly selected potentials of +0.70 V, –0.10 V and –0.30 V (*vs.* Ag|AgCl reference), respectively.

The PTBO-based enzymatic system was then selected to construct an amperometric hydrogen peroxide biosensor utilizing horseradish peroxidase (HRP) at a low detection potential of –0.10 V. Furthermore, a bienzyme glucose biosensor by co-immobilizing both HRP and GOx together onto a MWNT-modified GCE in an electropolymerized PTBO film has been developed. In this design, the HRP layer could be used to analyze hydrogen peroxide with TBO mediators, while the bienzyme system (HRP + GOx) could be utilized for glucose determination. An unusual amperometric response for glucose detection was observed due to the competition between oxygen and an oxidized mediator for the regeneration of the oxidase enzyme. The reaction schemes for glucose detection at the PTBO coated bienzyme electrodes have been examined in detail. The bienzyme electrode offered high sensitivity for amperometric determination of glucose at low potential, displaying Michaelis-Menten kinetics. The bienzyme glucose biosensor offered a linear response up to 1.2 mM with a sensitivity of 113 mA M^{-1} cm^{-2} at an applied potential of –0.10 V in air-saturated electrolytes. The reaction schemes for glucose detection at the PTBO coated bienzyme electrodes have been examined in detail.

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