DOCTORAL THESIS

Development of reduced graphene oxide based nanocomposities for electrochemical biosensing applications

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Abstract

The modification of electrodes is always an important task in electrochemical detection of electroactive and biological molecules. Chemically modified electrodes can offer improved selectivity and sensitivity for the target analyte, which greatly enhance the electrode performance. Various materials such as conducting polymers, metal nanoparticles and carbon nanomaterials have been exploited and widely used for the modification of electrodes. Electrochemical or spontaneous deposition, electrostatic adsorption, layer-by-layer self assembly and covalent binding have also been developed for electrode modification and offer improved performance.

Both Prussian blue (PB) and toluidine blue O (TBO) are excellent redox mediators and very popular in electrode modification. PB has shown strong catalytic property for the reduction of hydrogen peroxide, but the application in biosensor fabrication is limited for its instability at neutral pH. Graphene, as a single-atom-thick carbon material, is considered an ideal platform for designing composite nanomaterials for high-performance electrochemical or electrocatalytic devices. The combination of PB with reduced graphene oxide (RGO) and poly(toluidine blue O) (PTBO) will greatly improve the stability of PB. An amperometric biosensor based on glassy carbon (GC) electrode modified with reduced graphene oxide, PB and poly(toluidine blue O) was developed. Experimental results showed that the GC/RGO/PB/PTBO modified electrode offered an excellent electrocatalytic activity toward the reduction of hydrogen peroxide due to the possible synergistic effects of the PB-PTBO composite material. After codeposition of glucose oxidase (GOD) and chitosan (CHIT)

coating, the resulting GC/RGO/PB/ PTBO/CHIT-GOD electrode exhibited excellent response to glucose with a sensitivity of 59 mA M^{-1} cm^{-2}, a low detection limit of 8.4 μM and a linear range from 0.02 to 1.09 mM at a detection potential of +0.2 V vs. Ag|AgCl reference.

Reduced graphene oxide – gold nanoparticles composites were prepared by depositing gold nanoparticles (AuNPs) on the surface of reduced graphene oxide with different RGO-to-AuNPs weight ratios. The resulting composite materials were characterized morphologically and optically by scanning electron microscopy and UV-Visible absorption spectroscopy. Electrocatalytic effect of different composites toward the reduction of hydrogen peroxide has been investigated. The results demonstrated that RGO-AuNPs composites displayed high stability and catalytic effect for the analysis of hydrogen peroxide compared with that of RGO and AuNPs alone, demonstrating the possible synergistic effects of the RGO-AuNPs composite materials. Additionally, direct electron transfer of glucose oxidase was achieved after codeposition of GOD and chitosan. The glassy carbon electrode modified with RGO-AuNPs/CHIT-GOD material exhibited an excellent catalytic effect for glucose detection with a sensitivity of 34 mA M^{-1} cm^{-2} at a detection potential of −0.3 V vs. Ag|AgCl.

A reduced graphene oxide - gold nanoparticles - Prussian blue (RGO-AuNPs-PB) composite was prepared by spontaneous deposition of PB on RGO-AuNPs surface. The GC/RGO-AuNPs-PB modified electrode offered good electrocatalytic activity toward the reduction of hydrogen peroxide, indicating the possible synergistic effects of the RGO-AuNPs-PB composite material. Direct electron transfer of glucose oxidase can be realized after codeposition of GOD and chitosan coating on GC/RGO-AuNPs-PB electrode. The resulting

GC/RGO-AuNPs-PB/GOD-CHIT biosensor exhibited an excellent amperometric response to glucose with a sensitivity of 84 mA M^{-1} cm^{-2}, and a linear range from 13 μM to 2.0 mM at +0.1 V vs. Ag|AgCl.

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