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Encapsulating benzoquinone and glucose oxidase with a PEDOT film: Application to oxygen-independent glucose sensors and glucose/O_2 biofuel cells

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ABSTRACT

A modified electrode was proposed based on the sequential coating to immobilize both *p*-benzoquinone (BZQ) and glucose oxidase (GOD). Three electrodes, A, B, and C, were prepared separately by drop-coating the BZQ solution dissolved in different solvents on the stainless-steel/carbon (ssteel/C). Among those three electrodes, electrode B shows the best sensitivity of 2.21 mA M^{-1} cm^{-2}, a linear concentration range of 1.1–15 mM and a response time of 100 s at a sensing potential of 0.3 V. The responses of interferences, including ascorbic acid, dopamine, uric acid and acetaminophen, were ~0%, 1.4%, ~0% and 3%, respectively, taken the sensing current at 6.0 mM glucose as 100%. In a test of the human blood sample, an error of +3.6% was noticed for electrode B. Besides, for the biofuel cell application, maximum power densities reached 18.9 and 22.5 μW/cm^2 at 25 and 37 °C, respectively, with an all-solution-type biocathode.

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1. Introduction

Glucose is an attractive target, because it is not only an important biomarker for diabetes but also a kind of fuel for a biofuel cell. In other words, a glucose biosensor can work both for monitoring the glucose level and for the anode of the biofuel cell. An amperometric enzyme-based biosensor is the best choice for biochemical analysis due to their good selectivity, sensitivity, rapid response, miniature size, and reproducible results (Hamdi et al., 2006). The good selectivity is attributed to the specific catalysis by the enzyme. However, the electron transfer between the electrode and the redox center deeply embedded inside an enzyme is retarded, so mediators or oxygen must be used to pass electrons from the enzyme to the external circuit or to the solution. The mediator is required for oxygen-free system and the choice of the mediator is important. A proper mediator, such as ferrocene (Long et al., 2009), quercetin (Chen et al., 2009), BZQ (Lau et al., 2003) and its derivative (Babkina et al., 2006), can provide additional advantages of lowering the sensing potential and eliminating the interferences. Furthermore, the all-in-one electrodes, which encapsulated with the enzyme and the mediator, can show the advantages of convenient, reusable and workable with or without oxygen.

Generally speaking, the methods of enzyme immobilization include adsorption, entrapment, cross-linking and covalent bonding (Cunningham, 1998). For adsorption, the enzyme was adsorbed on the electrode by the attractive force of hydrogen bonds or opposite charges (Yang et al., 2004). The enzyme also can be entrapped in a matrix, such as sol–gel (Lin et al., 2007) and the conducting polymer of polypyrrole (Brahim et al., 2001; Singh et al., 2004). Besides, the enzyme and electrode can be directly linked by covalent bonds (Nien et al., 2009) or by cross-linking method (Tamiya et al., 1990). On the other hand, the immobilization of mediators is more difficult than that of enzymes, because mediators usually suffer from the leakage of small molecules and they are usually water-insoluble. In order to improve the stability of mediators on the electrode, the covalent method is a more effective one. For example, mediators were linked on a multi-wall carbon nanotube (Qiu et al., 2009), a polymer matrix (Himuro et al., 2009) or even an enzyme directly (Wu et al., 2008). Moreover, the mediator was linked both with the electrode and the redox center of enzyme for increasing the efficiency of electron transfer from the enzyme to the external circuit (Zayats et al., 2008). Qiu et al. (2007) proposed a method to link the small molecules, mediators, on the large Fe_3O_4@SiO_2 nanoparticles. Afterward, the enzyme and the nanoparticles were entrapped inside the matrix at the same time. Although linking of mediators with CNT, polymer matrix, or enzyme may provide an effective way of stabilization, it has some drawbacks such as leaching of mediators from CNT.

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In addition to the linking method, another easier way of immobilization of enzyme and mediators was reported using the co-adsorption method (Brunel et al., 2007; Habrioux et al., 2007). Encapsulating both the enzyme and the mediator can show the advantages of convenient, reusable, time saving, and workable with or without oxygen.

In the present investigation, we fabricated the mediator and enzyme encapsulated electrode without linking steps, based on sequential coating structure as shown in Fig. 1A. The first layer, the carbon paste, which was coated on the flexible substrate of stainless steel, was acted as an adsorbent layer for the mediator, BZQ, by the hydrophobic force. The BZQ and GOD were drop-coated on the electrode in order and the entrapped matrix, PEDOT, was electropolymerized finally on the outer layer to prevent the leakage of mediators and enzymes. The all-in-one electrode has the advantages of flexibility, easy to use in oxygen-free solution, low sensing potential and low interference effect. The advantage of the flexible sensor is that the structure is suitable for biomedical instrumentation because it does not cause discomfort (Iguchi et al., 2007; Chu et al., 2009). The performances for glucose monitoring and biofuel cell application were studied. Besides, the real sample of human whole blood was also monitored. The electron transfer illustration of biofuel cell is shown in Fig. 1B and the mechanisms of glucose sensing are shown in Eqs. (1)–(3) or the anodic compartment in Fig. 1B.

$$\text{Glucose} + \text{GOD}_{ox} \rightarrow \text{Glucolactone} + \text{GOD}_{red} \quad (1)$$

$$\text{GOD}_{red} + 2\,\text{BZQ}_{ox} \rightarrow \text{GOD}_{ox} + 2\,\text{BZQ}_{red} \quad (2)$$

$$2\,\text{BZQ}_{red} \rightarrow 2\,\text{BZQ}_{ox} + 2\,\text{H}^+ + 2\,\text{e}^- \quad (3)$$

2. Experimental

2.1. Chemicals and instruments

The target (or fuel), D-(+)-glucose, and the interferences, ascorbic acid (AA) (>99%), uric acid (UA) (>99%), dopamine hydrochloride (DA) and acetaminophen (AP) (>99%), were purchased from Sigma. Sodium dihydrogen phosphate (NaH_2PO_4) and disodium hydrogen phosphate (Na_2HPO_4) were obtained from Sigma and the same were used for the preparation of 0.1 M PBS solution of pH 7 or 5.5. For the anode, the enzyme, glucose oxidase (GOD) (EC 1, 1, 3, 4) type VII-S from *Aspergillus niger*, and the mediator, *p*-benzoquinone (~98%, reagent grade) were purchased from Sigma and Aldrich, respectively. For the cathode, the enzyme, laccase (Lac) (EC 1, 10, 3, 2) from *Trametes versicolor*, and the mediator, 2,2-azino-bis(3-ethylbenzothiazoline- 6-sulfonic acid) diammonium (ABTS) salt were received from Fluka and Sigma, respectively. The monomer, 3,4-ethylenedioxythiophene (EDOT), surfactant, polyethylene glycol (PEG, MW = 20,000) and bacteriostat, sodium azide (>99.5%) were purchased from Aldrich, Merck and Sigma, respectively. Besides, the flexible substrate was stainless steel SUS 301 and the membrane for biofuel cell was Nafion® 117 (thickness is 0.007 in). The de-ionized water (DIW) was used throughout the experiments. Thermal curable conductive carbon for printed circuit boards was purchased from Ishen Chemical Supply, Taipei, Taiwan. CNT used in this work was multi-wall CNT (MWCNT), which was purchased from Nanotech Port Co. (Taiwan); these MWCNTs were produced via the chemical vapor deposition (CVD, or sometimes called the catalytic pyrolysis). All electrochemical

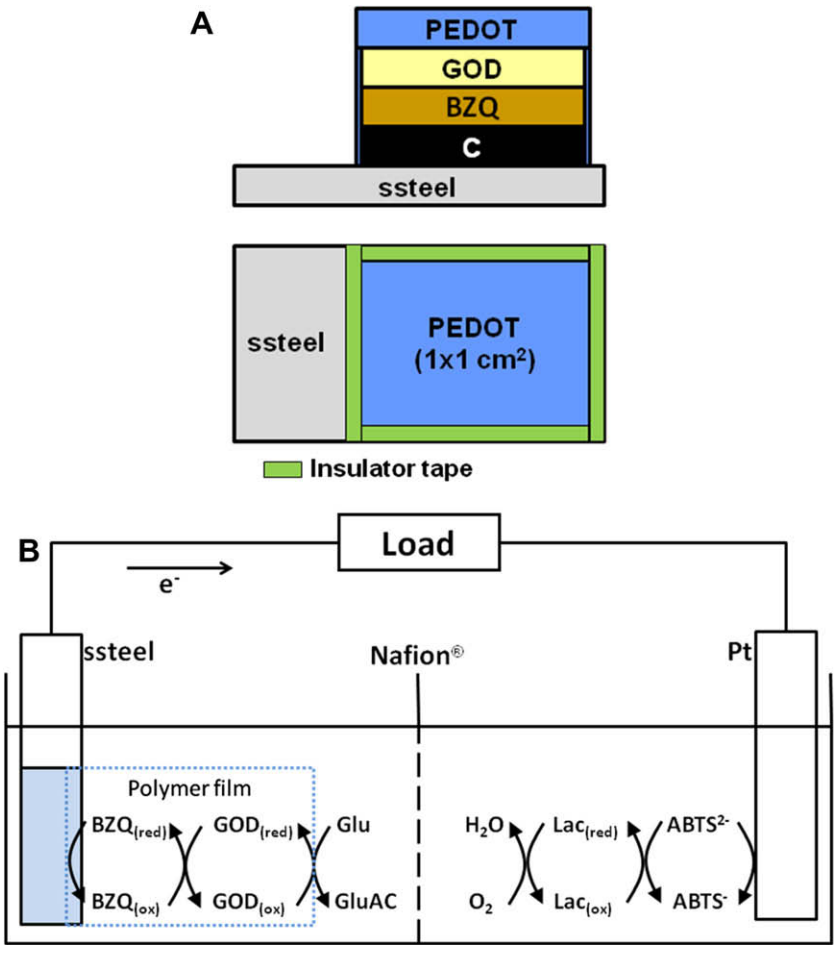

Fig. 1. (A) The deposition sequences of the modified electrode in side and top views and (B) the electron transfer path in the biofuel cell.

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