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Determination of serum glucose using flow injection analysis and highly selective glucose sensor based on composite films

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ABSTRACT

A novel flow injection analysis (FIA) system suitable for measurements of glucose in blood serum is developed. In the proposed FIA system, a new kind of glucose sensor based on composite polymer films and well-immobilized enzyme was fabricated. An electrochemical technique of scanning electrochemical microscopy (SECM), and electrochemical impedance spectroscopy (EIS) were used for the characterization of the newly fabricated biosensor. A wide linear range of 0.1–50 mM for glucose detection was reported in virtue of the new configuration of the sensor and the developed FIA system. The reproducibility of signals was quite good with relative standard deviation (RSD) values for $n = 4$ injections (typically 5.7%). Animal blood serum was directly injected and assayed in this simulative physiological system. Good analytical recovery of glucose spiked into serum samples, with recoveries in the range of 96.7–105.0%, was exhibited. Under optimized conditions, detection of serum glucose for normal people and diabetics using our proposed method is possible.

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1. Introduction

Diabetes mellitus is a group of dysmetabolic syndrome resulted from genetic cause, metabolic diseases, autoimmune disorders, microorganism infection, toxins, etc. Diabetic emergency such as hyperglycemia and hypoglycemia needs to be avoided, and it is also quite necessary to confirm the effectiveness of clinical treatment. Therefore, testing of physiological glucose levels for normal people and diabetics is critical. Glucose sensors, as one of the most popular electrochemical biosensors, have been extensively developed and applied in clinical testing of diabetes, biological and chemical analysis and food industry [1–3].

FIA technique was first introduced in 1975, presenting quantitative information of samples reproducibly in flowing streams, and has been widely applied in many fields including biochemistry, environmental science and medical science. A certain volume of samples was injected into a reagent stream continuously flowing through a tube, forming a sample area, and further mixed with the reagent(s) in streams, and finally reached the detector that typically quantitates the product of the chemical reaction. Meyerhoff et al. have reported the measurement of *S*-nitrosothiols (RSNOs) in animal blood plasma using FIA method, in which a differential experiment step and an amperometric nitric oxide (NO) sensor were applied [4]. Kolev et al. successfully developed a novel FIA system that used a polymer inclusion membrane (PIM) for the on-line extractive separation and determination of Zn(II) in the presence of a range of other metal ions [5]. The FIA method has been used by some groups for glucose measurements [6,7] as well, however, problems such as poor ability of anti-interference and narrow linear range are difficult to resolve, producing complicated process. In the present work, a kind of glucose sensor with new configuration was fabricated and further applied as the detector in the FIA system, resulting in good ability of anti-interference and wide linear range for sample detection, and the process for glucose measurements was very simple. Animal serum samples containing glucose were directly injected into the system and further carried by the flowing streams to the position of detector where samples reacted with oxygen and enzyme. Consequently, hydrogen peroxide generated by the enzymatic reaction would be detected and measured by the biosensor in proportion to the amount of glucose in samples [8]. A typical recording of output has the form of a current peak, and the height of the peak is related to the concentration of the analyte according to a standard calibration curve obtained with glucose standards prepared [9].

Polymer films have been widely employed in biosensors to increase permselectivity, to prevent the electrode surface from fouling, to entrap or immobilize a mediator, to extend the linear range of biosensors, and to improve the biocompatibility of biosensors [10–12]. Polymer films, categorized as conducting, nonconducting and composite, are mostly fabricated using solvent

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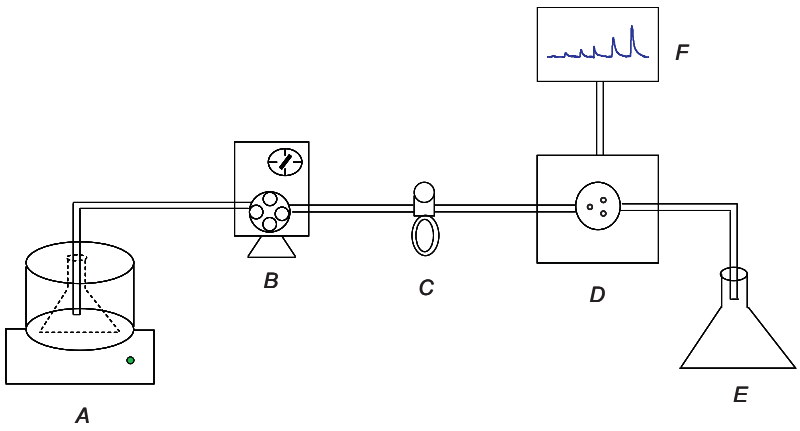

Fig. 1. Schematic diagram of flow injection analysis system. (A) Temperature controlled carrier stream (PBS buffer); (B) peristaltic pump; (C) six-port rotary injection valve; (D) temperature controlled flow-through cell equipped with detector (amperometric glucose sensor); (E) waste; (F) recorder.

casting, electropolymerization, and adsorption. Belanger et al. electropolymerized a conducting glucose oxidase/polypyrrole film onto the Pt disk of a rotating ring-disk electrode to fabricate a glucose sensor [13]. Yacynych et al. evaluated the effectiveness of various electropolymerized films and reported that poly(1,3-phenylenediamine/resorcinol) film covered electrode was not fouled by serum solution. In that work, glucose oxidase (GOD) was immobilized onto the electrode surface by crosslinking with glutaraldehyde followed by an electropolymerized film, and the resulted sensor had a linear response (2.5–10 mM) using FIA method [14]. In the present work, we simultaneously immobilized GOD into the interspace of poly(1,3-phenylenediamine/resorcinol) molecules during electropolymerization followed by modification of Nafion perfluorinated ion exchange resin (5 wt.% solution in lower aliphatic alcohols/H_2O mix containing 45% water) film. A wider linear range of 0.1–50 mM for serum glucose detection was achieved in FIA system, and such configuration of glucose sensor and so wide linear range for detection have rarely been reported by now.

2. Experimental

2.1. Materials and reagents

Glass carbon working electrode (1.5 mm in radius), platinum wire counter electrode (0.5 mm in radius) and Ag/AgCl reference electrode were purchased from CH Instrument (Chenhua Instrument Co., Ltd., Shanghai, China). All potentials were reported against the Ag/AgCl reference electrode.

All chemicals were of analytical grade or better and used as received without further purification. D-(+)-Glucose and glucose oxidase (GOD) were obtained from Sigma–Aldrich. Potassium ferricyanide, ethylenediaminetetraacetic acid (EDTA), 1,3-phenylenediamine (m-PD) and resorcinol were purchased from a chemical supplier (J&K Scientific Co., Ltd., Beijing, China). Alumina polishing powder (1.0 μm, 0.3 μm, 0.05 μm) was obtained from CH Instrument (Chenhua Instrument Co., Ltd., Shanghai, China). Potassium chloroplatinate was obtained from a commercial chemical supplier (Boyuan Chemicals Co., Ltd., Jinan, China). Nafion perfluorinated ion exchange resin was purchased from Dupont Company (DE, USA). 0.01 M phosphate buffer solution (PBS) was prepared freshly from appropriate reagents. All aqueous solutions were prepared with 18.2 MΩ cm ultrapure water using a Milli-Q filter (Research UV, Hetai Instrument Co., Ltd., Shanghai, China).

2.2. Apparatus and method

Electrochemical experiments were carried out using a developed FIA system, the diagram of which is shown in Fig. 1. A peristaltic pump (BT01-Y21515, Tianjin Xieda electron. Co., Ltd., Tianjin, China) was applied to pump the carrier stream (PBS), and the thermostat (85-2, Siyue Instrument, Co., Shanghai, China) was used to keep the required temperature. The sample loop and polyetheretherketone (PEEK) tubing used for carrier stream were purchased from a commercial supplier (RUSH Science & Technology Co., Ltd., Hangzhou, China). All current measurements from the glucose sensor were performed using a potentiostat (CHI 920C, Chenhua Instrument Co., Ltd., Shanghai, China). Samples were injected into the carrier stream through the six-port rotary valve (Rhenodyne 7725i) and loaded into the fitted 200 μL sample loop. Immediately, samples would be carried to the thermostatic flow-through cell equipped with detector in the oxygen atmosphere, and glucose contained in samples would be reacted to generate hydrogen peroxide as a product. Glucose sensor, as the detector, would respond to this hydrogen peroxide, and a current peak related to glucose concentration was displayed by the recorder. Calibration curves were obtained by injecting freshly prepared standard glucose solutions into the FIA system. All samples and all components of the FIA system including the carrier stream, tubing and electrochemical cell were prevented from light with aluminum foil. Furthermore, EDTA was added to blood samples as an anticoagulant. Construction procedure of polymer films was analysed by means of the technique of scanning electrochemical microscopy (SECM, Chenhua Instrument Co., Ltd., Shanghai, China) and electrochemical impedance spectroscopy (EIS, CHI 920C, Chenhua Instrument Co., Ltd., Shanghai, China).

2.3. Fabrication of glucose sensors

The glass carbon working electrode was mechanically polished with successively finer grades of deagglomerated alumina slurries down to 0.05 μm in particle size. An ultrasonic cleaner was used to remove residual alumina loosely bound to the electrode surface. The polishing procedure was repeated until the working electrode was electrochemically clean for use. The ensuing working electrode was platinized in a saturated potassium hexachloroplatinate solution by cycling the potential from +0.70 to −0.35 V (vs. Ag/AgCl) at a scan rate of 20 mV/s for 8 min using the potentiostat. Poly(m-PD/resorcinol) films were electrochemically grown from a fresh solution containing 1.5 mM m-PD and 1.5 mM resorcinol

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