



A novel paperfluidic closed bipolar electrode-electrochemiluminescence sensing platform: Potential for multiplex detection at crossing-channel closed bipolar electrodes

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ABSTRACT

In this work, we for the first time reported a paperfluidic crossing-channel closed-BPE based electrochemiluminescence (P-3C-BPE-ECL) platform where multiple “band”-shaped C-BPEs are situated perpendicular to two parallel channels, and multiplex detection can be achieved in the reporting channel. The paperfluidic devices are easily fabricated by wax and carbon ink-based screen-printing processes. Under optimized conditions, the P-3C-BPE-ECL is applied to the quantitative analysis of hydrogen peroxide (H_2O_2) and glucose, with corresponding linear ranges of 0.075–10 mM and 0.08–5 mM, and corresponding detection limits of 0.041 mM and 0.03 mM. To our knowledge, this is the first demonstration of paperfluidic C-BPE-ECL method for glucose determination. Based on its acceptable selectivity, the P-3C-BPE-ECL method is used for measurements of glucose in four complex samples (human serum and urine, wine, and glucose injection) and is compared with the traditional methods. The results indicate a good agreement and prove the reliability and accuracy of the proposed platform. Importantly, the proposed method is demonstrated to have the potential for duplex detection of glucose and uric acid, which intensively matches the requirement of those patients simultaneously suffering from diabetes and gout. Therefore, we believe that the P-3C-BPE-ECL could provide a new platform for wide biochemical applications.

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1. Introduction

Bipolar electrochemistry has gradually become a well-established technique since the original concept of bipolar fluidic bed electrodes was introduced in the late 1960s [1]. It exploits the wireless nature of bipolar electrodes (BPEs) for applications that are either impossible or inconvenient to achieve using conventional wired electrodes. A BPE is an electrically conductive object that can promote both reduction and oxidation reactions even though it is not directly connected to an external power supply. In principle, when a potential gradient is provided between two driving electrodes, the redox reactions preferentially occur at the ends of BPE where the interfacial potential difference between the electrolyte solution and BPE is the highest [2]. Relative to traditional

three-electrode electrochemical systems, BPE systems can offer distinctive advantages due to their wireless nature, including great potential for simultaneous control of multiple working electrodes under a single driving electrode, and requirement of only simple instrumentation that can be easily handled, even by inexperienced persons [3].

According to the current flow path, BPE can be classified as open and closed BPE (O-BPE and C-BPE). For the former, both cathode and anode are placed in the same fluid, typically inside a microchannel, and the BPE transfers current by electrons and ions between the two poles. So far, a variety of O-BPE systems have been developed for sensing, screening, focusing and micro-swimmers applications [2]. However, all O-BPE systems exhibit quite a low current efficiency (η_c , $\eta_c = \text{the current flow through the BPE} / \text{the current flow through the driving electrode}$), because a significant portion of the total current between the driving electrodes is carried by an ionic current in the microchannel, which may greatly limit their practical applications. Different from conventional O-BPE systems, C-BPE systems place the anode and cathode in different solutions, such that they are chemically and fluidically isolated [4]. Relative to O-

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BPE, C-BPE has several advantages: (1) the two poles are separated into different electrolytic cells and the current only passes through the BPE, so it can provide 100% current efficiency in theory; (2) two distinct redox systems in different solution environments are independent from each other, thus minimizing cross-talk or interference between the two systems; (3) the driving voltage is dropped almost entirely at the solution interface adjacent to the two extremities of the C-BPE if two idea nonpolarizable electrodes are applied, thereby a relatively low voltage is usually sufficient to drive both redox reactions on the C-BPE; and (4) because of the lack of the microchannel and ionic current path, the electrochemical current on the C-BPE can be directly measured, and the rates of the faradic processes on the BPE can be reported. Possibly due to these attractive features, C-BPE has drawn more interest, and has been used as a novel approach for various applications covering sensing, screening, functionalization of micro/nanowires, keypad lock, and so on [5].

Over the past years, several methods have been explored for signal readout in C-BPE systems, including cyclic voltammetry [4], fluorescence [6], luminescence from light emitting diode [7], electrochromism [8], and electrochemiluminescence (ECL) [9,10]. Among these methods, the optical readouts based on ECL and fluorescence may be attractive, because the light emissions can be remotely monitored for indirect measurement of current flowing through the C-BPE (i.e. the Faradaic reaction at C-BPE poles). Moreover, these two methods are easily used to visualize electrochemical signals on large arrays of C-BPEs simultaneously. Generally, the fluorescence method is highly sensitive and selective, and can be developed for many applications such as cell imaging, bisulfite detection and lead(II) measurement [11–15]. However, the fluorescence analysis using C-BPE is relatively rare, possibly because a relatively complex and expensive optical detection system is often required. On the contrary, the use of ECL as a reporting mechanism is very popular in C-BPE systems [10], since ECL is usually characterized by no need for a light source, low background interference, low cost of instrument, and simple operation process. Up to now, some C-BPE-ECL microfluidic chips have been developed for detection of various targets, including cancer biomarkers (adenosine triphosphate [16], prostate-specific antigen [16–19], α -fetoprotein [16], carcino-embryonic antigen [20] or thrombin [16]), cancer cells [21–23], tripropylamine (TPA) [24–26], hydrogen peroxide (H_2O_2) [20,24,25,27,28], dopamine [24], ascorbic acid [25] and $Fe(CN)_6^{3-}$ [9,24].

Although the mentioned-above C-BPE-ECL chips have exhibited an acceptable analytical performance, they still have some problems for practical applications. Firstly, tedious, complicated and time-consuming microfluidic technologies are required. Secondly, expensive chip materials are applied. Thirdly, the indium tin oxide (ITO) coated glass is used as the bipolar substrate for nearly all C-BPE-ECL chips. Although the ITO C-BPE has excellent conductivity and superior transparency, SnO_2 at ITO is easily reduced when the applied driving voltage is relatively high, causing the damage of ITO and then the instability of the out signals. To eliminate the self-reduction of ITO, the electrodeposition of Au [21] and Pt [25] noble metals onto the BPE cathodes has been applied, which increases the complexity of the fabrication process. Fourthly, the previous C-BPE-ECL chips are very difficult to perform multiplex detection in a single electrolytic cell (reaction microchannel), possibly because the width of two microchannels is usually difficult to accommodate multiple BPE bands. To partly solve these problems, we and other groups have introduced simple and low-cost paper- or cloth-based fluidic chips to the C-BPE-ECL field [29–33]. Nevertheless, these fiber material-based C-BPE-ECL chips still do not solve the above fourth problem. To our knowledge, there have been no reports on multiplex C-BPE-ECL in a single on-chip microchamber where each “band”-shaped

BPE can exhibit a similar ECL performance under the same conditions.

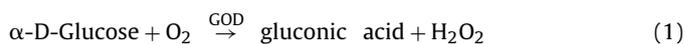
In this paper, a novel paperfluidic C-BPE-ECL sensing platform has been developed, where one or more “band”-shaped C-BPEs are placed perpendicular to two parallel paper channels, and the cathode and anode of each C-BPE are connected onto the supporting channel and reporting channel. Such a paperfluidic crossing-channel C-BPE based ECL (P-3C-BPE-ECL) platform provides several attractive features: (1) the paperfluidic devices are simple in construction, low in cost, and do not require any experienced persons; (2) the ECL intensities at C-BPEs are almost identical when the single target is analyzed; and (3) the multiplex assays in the reporting channel can be performed for detection of various targets. Based on the commonly-used luminol/ H_2O_2 system, the P-3C-BPE-ECL platform can be used to quantitatively determine H_2O_2 and glucose, with acceptable detection sensitivity, wide dynamic range, and high selectivity. Additionally, the glucose levels in various complex samples are measured on the proposed platform, with acceptable similarity to those obtained by the conventional methods. As is well-known, the determination of glucose has attracted increasing attention in many fields such as food analysis, bioreactor detecting, and clinical diagnosis.

2. Experimental section

2.1. Device design and sensing principle of the P-3C-BPE-ECL platform

Fig. 1A schematically illustrates the design of the P-3C-BPE-ECL device. It is made of the paper substrate, and its whole structure is symmetrical about the center of the paperfluidic chip. On the chip, two parallel hydrophilic channels (i.e. supporting channel and reporting channel) are surrounded by a hydrophobic barrier. On one hand, one or more “band”-shaped C-BPEs perpendicularly stride across two paper channels, and the cathode and anode of each C-BPE are respectively located inside the supporting channel and the reporting channel. On the other hand, the anodic and cathodic driving electrodes are situated on the left end of the supporting channel and on the right end of the reporting channel, respectively.

The potential profile across the two paper channels is shown in Fig. 1B. As is well-known, it is the interfacial potential difference ($\Delta\varphi_a$) that drives the faradic reactions, no matter at the working electrode in a conventional three-electrode system or the poles of C-BPE [19]. For the left C-BPE (C-BPE_L) (or right C-BPE, C-BPE_R), the applied potential equals the sum of the four interfacial potential differences ($\Delta\varphi'_a$, $\Delta\varphi_{c1}$ (or $\Delta\varphi_{c2}$), $\Delta\varphi_{a1}$ (or $\Delta\varphi_{a2}$), and $\Delta\varphi'_c$) and potential dropped on the solutions and C-BPE_L (or C-BPE_R). At the anodic pole of the C-BPE in the reporting channel, the anodic interfacial potential difference ($\Delta\varphi_{a1}$ or $\Delta\varphi_{a2}$) drives the ECL reactions. Under the presented conditions, for the left and right C-BPEs, the total potential drops across the solutions and C-BPE are the same, and thus the value of $\Delta\varphi_{a1}$ is equal to that of $\Delta\varphi_{a2}$. As a result, the ECL intensities at the C-BPEs should be almost equal under similar conditions. This feature is important for multiplex analytical applications. If the applied potential is sufficiently high, faradaic reactions occur at both poles of the C-BPE, one for oxidation at the anode and the other for simultaneous reduction at the cathode. Fig. 1C shows the sensing principle at the C-BPE based on the proposed luminol/ H_2O_2 reaction system. Obviously, the reaction system can be used to quantitatively detect H_2O_2 . When glucose is detected, the sensing assay is based on the enzymatic conversion of glucose by glucose oxidase (GOD) to form H_2O_2 . And, the possible reaction mechanism is described as follows [34]:



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