

RESEARCH PAPER

Cite this article as: Chin J Anal Chem, 2014, 42(8), 1220-1224.

characteristics of the closed bipolar electrode were studied by Zhang group^[16,25]. They successfully developed the closed bipolar nano and micro electrodes^[16], studied the

electrochemical sensing and superfine space sensing

characteristics, and greatly expanded the closed bipolar

electrode space for development and application. However,

the closed bipolar electrode electrochemical luminescence

Herein, a novel closed bipolar electrode electrochemilumin-

escence (ECL)-based device was designed and used to

investigate the ECL behaviors of luminol. This method had

the following advantages. Firstly, compared to microfluidic

BPE electrochemical luminescence device, this device was

simple, cheap and easy processing. Second, the anode and

cathode of BPE were separated in two poles by which the

electrochemical luminescence sensor reaction medium were

conveniently chosen in terms of their respective requirements,

avoiding the interference with each other. Therefore, "the

disturbance effect" in classical microfluidic liquid, such as

electroosmotic flow in BPE system could be effectively

and

the

concentration, separation medium

properties were not widely explored.

analyte

Electrochemiluminescence Behavior of Luminol at Closed Bipolar Electrode and Its Analytical Application

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Abstract: A new closed bipolar electrode electrochemiluminescence (ECL)-based device was designed and used to investigate the ECL behaviors of luminol in this device. The results showed that while a suitable voltage was applied to the two poles of the closed bipolar electrode, both the positive charge ions and luminol-based anionic ions could be enriched on the two poles of the closed bipolar electrode, respectively. More importantly, the ECL signals, generated from the electro-oxidation of luminol on anodic pole, was found to be related to the total amount of positive charged ions on the cathodic pole of the closed bipolar electrode. Under the optimum conditions, the ECL response was linearly to the concentration of analyte in the range of 1.0×10^{-9} – 1.0×10^{-8} M with a detection limit of 1.1×10^{-10} M. Based on this finding, a new ECL method for sensing the solution conductance was firstly developed.

Key Words: Electrochemiluminesence; Closed bipolar electrode; Luminol; Solution conductance

1 Introduction

In recent years, bipolar electrode (BPE), due to its simple structure, easy to be fabricated and processed, and make a electrochemical oxidation-reduction reaction without any direct electrical connection with the power supply, was further applied in synthesis of electrochemical^[1–6], manipulation of materials^[7], electrochemical sensing and electrochemiluminesence (ECL) sensing^[8–14]. It has been become a hot subject in analytical chemistry^[15].

At present, there are open and closed bipolar electrodes in terms of the design of the bipolar electrode structure^[15,16]. The theory research and application of open bipolar electrode was special widely^[15]. Crooks group developed a bipolar electrode by combining an open bipolar electrode with a microfluidic channel. The developed bipolar electrode was widely used in electrochemical luminescence chemical^[8–14], biological sensing^[17], analyte separation and enrichment of research field^[18–24], greatly broadening the bipolar electrode analysis of theoretical research and application^[15].

Furthermore, the electrochemical theory and analytical

Received 14 December 2013; accepted 2 April 2014

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This work was supported by the National Nature Science Foundation of China (No. 21375085).

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reduced or eliminated, and the signal stability and reproducibility of the system were obviously improved.

2 Experimental

2.1 Instruments and reagents

MPI-A electrophoresis electrochemical luminescence detector was purchased from Ruimai Companies in Xian, China. DDS-11A digital conductivity meter was purchased from Shanghai Ray Magnetic Productive instruments Co., LTD, China. Two platinum wires, 1.5-milliliter centrifuge tube, 0.5 mm diameter polytetrafluoroethylene tube and weighing bottle (25×25) were used in this experiment.

Phosphate buffer solution (pH 7.4) was prepared with ultrapure water (Sinopharm Chemical reagent Co., LTD, China). Sodium hydroxide (Sinopharm Chemical reagent Co., LTD, China), luminal (Sigma-Aldrich Co., Llc.) and ultrapure water were used.

2.2 Fabrication of closed bipolar electrode

An isolated platinum wire (0.5 mm diameter, 10 mm length) was placed into a glass tube (40 mm length), and sealed with alcohol lamp.

2.3 Experimental methods

As shown in Fig.1, a power could provide high pressure of 0–20 kV, tow platinum electrodes were connected to their two ends, and submerged in the electrolytic cell, respectively. The closed bipolar electrode was inserted in the ECL pool to detect the ECL behavior of this device. The Teflon tube was connected on both ends to make the whole circuit to be unblocked.

The cathodic segment of the BPE was filled with analyte solution, and the anodic segment of the BPE was filled with 1.0×10^{-4} M luminol solution. By applying pulse voltage to drive electrodes, the sample rate was 10 times per second, the high pressure of photomultiplier was set at 700 V, and the ECL signals were recorded.

3 Results and discussion

3.1 Design of closed bipolar electrode electrochemiluminescence (ECL)-based device

As shown in Fig.1, the anode reservoir and connecting pipe a were filled with analyte solution, the cathodic reservoir, connecting pipe b and the ECL pool were filled with 1.0×10^{-4} M luminol solution. The closed bipolar electrode was inserted into the ECL pool. An electric field gradient was generated in the vicinity of a bipolar electrode by applying a driving voltage, thereby lead to concentrate charge analyte at the poles of BPE^[26]. If driving voltage was sufficiently high, the faradaic electrochemical reactions would occur at the poles of the BPE. The luminol that concentrated on the anodic segment of the BPE could produce ECL signal, and then sensing the cation that concentrated on the cathodic segment of the BPE. Therefore, according to the ECL signals, the total amount of impurity ions in the solution was sensed.

3.2 Investigation of electrochemiluminescence characteristic of luminol at closed bipolar electrode

To test function of the device, the ECL sensing properties of the device using the luminol as the ECL reagent and phosphate buffer solution (PB solution) with low concentration and complex components as the analyte solution were investigated. When the pulse voltage was high enough to drive electrodes, the anodic segment of the BPE would produce strong ECL signals. When the applying pulse voltage time was set at 3 s, the recording the ECL signal time was 40 s and the good ECL signals could be observed. The ECL dynamic curve is shown in Fig.2. More importantly, as the pulse voltage was continuously applied, the ECL signals would continue to increase. Thus, if there was not electrolyte in the cell, only the weak ECL signals could be detected. Therefore, this ECL signals could be used for sensing the amount of electrolytes in the solution.



Fig.1 Scheme for the closed bipolar electrode and ECL device



Fig.2 ECL profile of luminol $(1.0 \times 10^{-4} \text{ M})$ on bipolar electrode a: $1.0 \times 10^{-5} \text{ M PBS}$ (pH 7.4), b: Ultrapure water

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