



Microbial fuel cell-based biosensor for toxic carbon monoxide monitoring

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ABSTRACT

This study presents an innovative microbial fuel cell-based biosensor for carbon monoxide (CO) monitoring. The hypothesis for the function of the biosensor is that CO inhibits bacterial activity in the anode and thereby reduces electricity production. A mature electrochemically active biofilm on the anode was exposed to CO gas at varied concentrations. A proportional linear relationship ($R^2 = 0.987$) between CO concentration and voltage drop (0.8 to 24 mV) in the range of 10% and 70% of CO concentration was observed. Notably, no further decrease of voltage output was observed by with further increasing CO concentration over 70%. Besides, the response time of the biosensor was 1 h. The compact design and simple operation of the biosensor makes it easy to be integrated in existing CO-based industrial facilities either as a forewarning sensor for CO toxicity or even as an individual on-line monitoring device.

1. Introduction

Carbon monoxide (CO) is considered as one of the most hazardous gasses for human health. Moreover, CO imposes negative environmental impact. Unfortunately, significant emissions of CO are generated as a byproduct of power plants and other human chemical activities [15,4]. Thus, timely gas leakage detection or forewarning is important for protecting human beings and work places from CO inroad. Over the last decades, a great deal of research has been dedicated in development of optical, semiconductor or electrochemical CO sensors. These sensors present high sensitivity and could detect CO at very low concentrations ranging from 1 ppm to hundreds ppm [13,18,22]. However, CO liberated from incomplete combustion or steel mill industry is usually at much higher levels [8]. So far, there is not any compact form of sensors focusing on the detection of high concentrations of CO. Besides, most of conventional sensors need noble metal fabrications which make them relatively expensive. Moreover, many conventional CO sensors operate at high temperatures (above 100 °C), and thus, are energy intensive. Thus, a simple, efficient and cost-effective sensor capable of CO monitoring at high concentrations, is needed.

Recently, various microbial fuel cell (MFC)-based biosensors have been presented. MFC are attractive as biosensors due to the sustainability, and also the easily detected signal. MFC biosensors have been extensively studied for various monitoring applications such as microbial activity, chemical/biochemical oxygen demand [20], volatile fatty

acids [10,11], nitric oxide reduction [23] and heavy metals (e.g. Cu^{2+}) [16,9]. MFC-based biosensors have also been employed to detect toxicity of specific pollutants (e.g. formaldehyde, Cu^{2+}) based on potential toxicity on exoelectrogens and reduction of electricity production [19].

Although CO could be utilized by Clostridium species as substrate [7], it is toxic to most of microorganisms. Thus, CO could also inhibit activity of bioanode in MFC and the drop of voltage could be an indicator of CO concentration. In this study, an MFC with mature anode biofilm was developed for the first time, to monitor CO concentration (termed as bioelectrical CO sensor). The objectives of this proof-of-concept study were (i) to demonstrate the feasibility of MFC for monitoring at high CO concentrations, (ii) to elucidate the correlation between voltage and CO concentration.

2. Materials and methods

2.1. Reactor configuration and startup

The MFC reactor used in this study consisted of two rectangular chambers made of nonconductive polycarbonate plates. The volume of anodic and cathodic chamber was of 70 and 35 mL, respectively. Two chambers were separated by a cation exchange membrane (CEM) (CMI 7000, Membrane international, NJ). The anode electrode was carbon brush (5.9 cm in diameter, 6.9 cm in length, Mill-Rose, USA) which was thermally pretreated at 450 °C for 10 min prior to use. Exoelectrogenic

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biofilm were enriched in the anode by inoculating with domestic wastewater which was taken from primary clarifier (Lyngby Wastewater Treatment Plant, Copenhagen, Denmark) and amended with 1 g/L sodium acetate (NaAc). Cathode electrode was made of Titanium mesh (4 cm × 3.5 cm) coated with Pt/C (0.5 mg (Pt)/cm²). The cathode chamber was filled by 50 mM phosphate buffer (PBS, pH 7.0) and constantly aerated by a pump through a sterile needle connected with 45 μm filter. Electric circuit between anode and cathode electrodes was connected via a 1000 Ω external resistance for voltage recording. After approximately 40 days operation, stable electrical energy output (620 ± 20 mV, set as baseline of sensing) was achieved in several successive batch assays, which indicated that electro-active biofilm was well established on the anode.

2.2. CO sensing tests

During batch tests, wastewater was replaced by nutrient solution (pH 7.2 ± 0.5) containing 50 mM PBS, 0.13 g/L KCL, 0.31 g/L NH₄Cl, 1 g/L NaAc, vitamin solution and trace mineral solution as previously described [21]. This nutrient solution was added to the anode chamber prior to each test to maintain the same conductivity and pH. Previously study indicated that effect of CO on conductivity and pH was negligible due to its poor solubility.

Before initiation of each batch test, the anode was flashed with nitrogen to remove oxygen. Subsequently, different concentrations of CO (from 0% to 100% at 10% interval, mixed with nitrogen) were fed into the anode through a peristaltic pump at 4 mL/min (OLE DICH, Instrumentmakers Aps, Denmark) (Fig. 1). A gas-diffuser was set on the bottom of anode chamber to cut CO into micro-bubbles upwards. After each test, the anolyte was refilled with fore-mentioned fresh nutrient solution to acquire fully recovery of electricity production. All the tests were conducted in replicate in water bath at 25 ± 0.5 °C.

2.3. Chemicals and analyses

All chemicals were analytical pure without further purification. CO was purchased from AGA Industrial Gases (Denmark). Voltage across the resistance was recorded at 5 min intervals throughout the test using a potentiostat (CT-4008W, Neware Battery Testing System, China).

3. Results and discussion

The bioelectrical CO sensor was tested with different CO concentrations (as percentage). Fig. 2 illustrates the correlation of voltage drop which was determined as the voltage difference before and 1 h after adding CO in the anode. It can be seen that voltage drop was

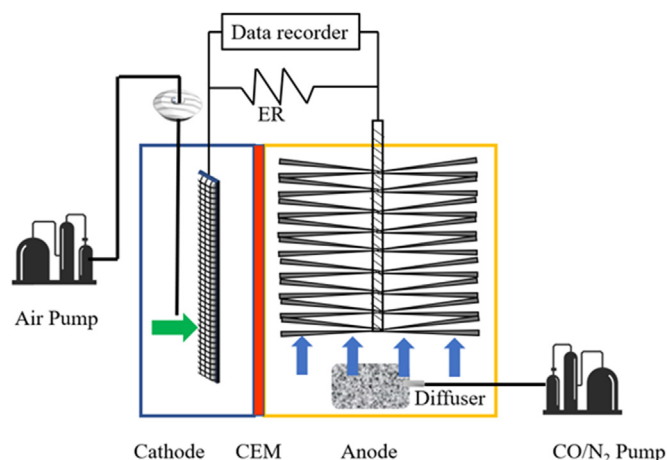


Fig. 1. Schematic illustration of bioelectrical CO sensor.

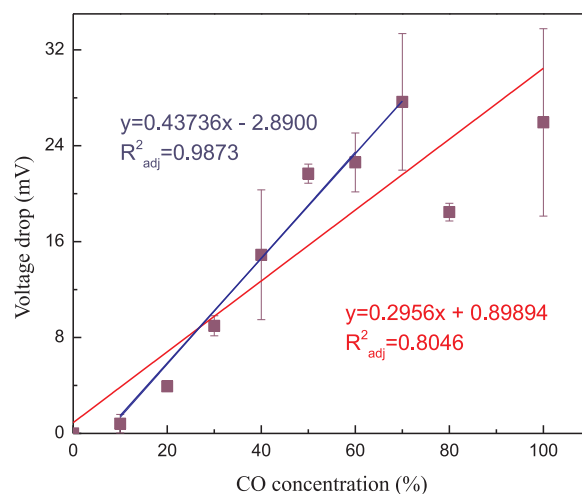


Fig. 2. The relationship between voltage drop (Δ mV) and CO concentrations after 1 h operation and linear correlation in the CO concentration ranges of 10–70% (Blue line) and 0–100% (Orange line), respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

increased with the increasing of CO concentrations. Detectable inhibition (~ 1 mV drop) firstly appeared when 10% CO (mixed with 90% of N₂) was pumped into the anode chamber. Approximately 24 mV of voltage drop was observed when CO concentration was up to 70% and then leveled out in the range of 22–34 mV for higher CO concentrations. Overall, the regression coefficient (R_{adj}^2) was 0.987 which indicated that the biosensor could be used to monitor the CO concentrations in the range of 10–70%. As anolyte was amended with sufficient sodium acetate which provided sufficient substrate for continuous electricity generation, the voltage decrease could only be explained by the slight inhibition effect of CO on the exoelectrogens, and thereby reducing the amounts of electrons generated from microbial metabolism. When the CO concentration was below 10%, toxicity was hardly detected. However, when CO was higher than 70%, the voltage did not decrease further probably due to the limited solubility of CO in the anolyte. These results indicate that MFC can be an effective biosensor for CO monitoring. CO could affect the current generation of MFC biosensor in two different ways. Firstly, similar to human CO poisoning, carbon monoxide might chelate with enzymes, which are involved in extracellular electron transfer or cell growth [6]. Secondly, CO may influence the metabolic processes.

The response time (RT), defined as the minimum time needed to acquire a reliable voltage measurement correlating to CO concentration was assessed. The biosensor became more sensitive (shorter RT) at higher CO concentrations (Fig. 3a). Accordingly, circuit current needed more time to recover to the former level after CO exposure. To be more specific, no lag phase of recovery was needed when CO inlet concentration was lower than 10% while it would take more than an hour to fully recover when 70–100% CO was tested. Actually, the voltage output would not immediately stop decreasing after the exposure at high CO levels. Instead, the voltage continued decreasing for approximately 20 min after CO was replaced by nitrogen. It could be deduced that the toxicity of CO might have negatively affected the microbial metabolism rather than thermodynamic electron-transfer reactions. Fig. 3b presents the different regression coefficients (R^2) between current and CO concentrations at different operating-time. Apparently, the correlation became more linear when the reaction time increased. The results indicated that in order to acquire an accurate monitoring of CO concentration, at least one-hour response time (R^2 of 0.987) should be applied (Fig. 3b).

Benefit from better characteristics of chemical catalysts, chemical based CO sensors (Table 1) generally presented higher selectivity and

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