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Micro-microbial electrochemical sensor equipped with combined bioanode and biocathode for water biotoxicity monitoring

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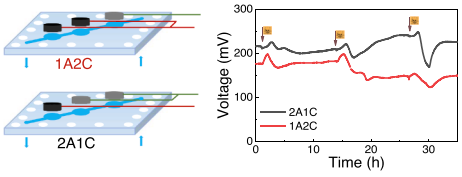
HIGHLIGHTS

- Novel micro-microbial electrochemical sensor is constructed.
- Sequential flowing membrane-free channel and bilateral passive O_2 supply are used.
- The ratio of number of bioanode to biocathode on performance of sensor is tested.
- Sensors make good response to the injection of formaldehyde into the inlet.
- A high microbial diversity of bioelectrodes was observed.

GRAPHICAL ABSTRACT



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ABSTRACT

The development of low-cost biosensors for water monitoring is expected to reduce potential risks from contamination accidents. This study reported a novel micro-microbial electrochemical sensor using combined bioanode and biocathode as the sensing element, characterized by a sequential flowing membrane-free channel and a bilateral passive oxygen supply. A decrease in the ratio of number of bioanode to biocathode resulted in a lower power generation, whereas, achieving a similar or even higher toxic response. The voltage was affected by both the flow rate and the acetate concentration. With the increased acetate concentration, a clear trade-off was observed between the electroactivity stimulation of bioanode vs. the electroactivity maintenance of biocathode. Biosensors made good response to the injection of formaldehyde (10 μL of 0.25%, and 100 μL of 0.025%) into the inlet. A high microbial diversity was observed. This work can lead to a revolutionizing way of water monitoring using self-powered micro-biosensors.

1. Introduction

The research of new technologies for water monitoring is expected to reduce the potential risks from contamination accidents (Do et al., 2020; Gupta et al., 2019; Hill et al., 2020). Physicochemical analysis methods, generally performed in centralized laboratories, can precisely detect the concentrations of specific pollutants, whereas, are incapable to assess their biological responses (Díaz-González and Fernández-Sánchez, 2019; Gupta et al., 2019). Biosensors possess an enormous potential for the environmental monitoring, which can be constructed with various bioreceptors, such as DNA, enzymes, and cells (Hao et al., 2020; Yi et al., 2020a). Among these biosensors, the self-powered microbial

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electrochemical sensors, for example microbial fuel cell (MFC) sensors, recently attract attention due to their potential application for on-line and long-term water biotoxicity monitoring (Khan et al., 2020; Qi et al., 2021). In microbial electrochemical sensors, the presence of pollutants could inhibit the extracellular electron transfer (EET) of electroactive microorganisms, to trigger a rather sharp change of electric signal (Chu et al., 2021; Xu et al., 2021a). In this regard, both bioanode and biocathode, with the enrichment of electroactive microorganisms, can be used in biosensing applications within these microbial electrochemical sensors (Jiang et al., 2017a).

Most studies on microbial electrochemical sensors are constructed with the bioanode as the sensing element, which is capable of outward EET by using organic matter as the electron donor and anode as the electron acceptor (Chu et al., 2020a; Jiang et al., 2015). However, the using of biocathode as the sensing element, which is capable of inward EET by using cathode as the electron donor and oxygen as the electron acceptor, is expected to increase the sensitivity of microbial electrochemical sensors (Jiang et al., 2018b; Yi et al., 2020b). The performance of biocathode sensing element for cheap and non-selective monitoring has been evaluated in the 3-electrode setup, which, however, is not working in a self-powered mode (Liao et al., 2018b; Prevoteau et al., 2019). In a self-powered microbial electrochemical sensor, the low redox potential of bioanode can drive oxygen reduction reaction (ORR) at the biocathode (Zhao et al., 2019). However, when using the outlet of anodic chambers as the inlet of cathodic chambers, the unconsumed organic matter could severely inhibit inward EET of biocathode (Jiang et al., 2017a). Air sparging with active oxygen supply is applied in a sequential flowing biosensor, to decrease the unconsumed organic matter in the cathodic chamber (Zhao et al., 2019). However, the air sparging might not be applicable for micro-biosensor, which could increase the complexity of device and might deprive its advantage as an energy self-sustaining device. A previous study has reported the construction of a gas diffusion biocathode sensing element for a unilateral passive oxygen supply in a two-chamber MFC sensor with a piece of membrane as the separator (Jiang et al., 2018a). The question is whether, and under what circumstances, a gas diffusion biocathode sensing element could be used to fabricate a membrane-free micro-microbial electrochemical sensor.

The fabrication of micro-biosensor could reduce the consumption of power and reagents (Gao et al., 2020; Uria et al., 2020), and enhance the performance for water monitoring (Banerjee et al., 2019; González-Pabón et al., 2021; Jiang et al., 2017b). For example, a microbial electrochemical sensor with a 75% smaller volume decreased the response time from 4 h to 40 min (Di Lorenzo et al., 2009). However, simply, and directly reducing the volume of conventional microbial electrochemical sensor is not practical, when equipping with both bioanode and biocathode as the sensing element. This is mainly because of that the unconsumed organic matter could severely suppress the inward EET of biocathode as discussed above. A sequential flowing membrane-free channel can be adopted in micro-microbial electrochemical sensor, as a certain proportion of organic matter in the inlet could be consumed by the bioanode firstly before it flows to the biocathode (Spurr et al., 2020). It is thus expected that the performance of micro-microbial electrochemical sensor could be affected by the ratio of number of bioanode to biocathode, the flow rate, and the loading of organic matter.

In this study, a novel micro-microbial electrochemical sensor was constructed using both bioanode and biocathode as the sensing element. The configuration was characterized by a sequential flowing membrane-free channel and a gas diffusion biocathode for a bilateral passive oxygen supply. The effect of acetate loading was evaluated by challenging with different flow rates and acetate concentrations. The performance of such biosensor for water monitoring was tested with the injection of formaldehyde into the inlet. The microbial community at bioanode as well as biocathode was also analyzed.

2. Materials and methods

2.1. Reactor construction and operation

The schematic of the micro-microbial electrochemical sensor is shown in Fig. 1. The ratio of number of bioanode to biocathode was 1:2 or 2:1, by using two bioanodes and one biocathode (2A1C), or using one bioanode and two biocathodes (1A2C) as sensing elements. The two bioanodes in 2A1C were parallelly connected, as well as the two biocathodes in 1A2C. The biocathodes were assembled with two pieces of waterproof breathable membrane (WBM) (Ningbo Shanquan Building Material Co., Ltd., China) for an enhanced bilateral passive oxygen supply. The WBM, commonly used in building construction, has been used in MFC for the fast fabrication of abiotic cathode and biocathode (Jiang et al., 2018a; Xing et al., 2015). The sequential flowing membrane-free channel was fabricated on an acrylic plate with 6 mm thickness. Three chambers opened on the acrylic plate with 16 mm in diameter were used for the equipment of sensing elements. The anode and cathode were made of 6 mm thick carbon felt. The carbon felt was pretreated with 1 M NaOH, 1 M HCl, and deionized water, to remove potential contaminations. The holes opened in gasket and end plate were used for passive oxygen supply to the biocathode sensing element.

The medium was prepared containing KH_2PO_4, 4.4 g L^{-1}; K_2HPO_4, 2.6 g L^{-1}; NH_4Cl, 0.31 g L^{-1}; Na_2SO_4, 0.05 g L^{-1}; $MgCl_2{\cdot}6H_2O$, 0.2 g L^{-1}; and yeast extract, 0.05 g L^{-1}, while the concentration of acetate was adjusted according to the experimental procedure. The micro-microbial electrochemical sensors were inoculated using the effluent from an MFC reactor fed with acetate (Jiang et al., 2019). During the process of polarization curve evaluation and internal resistance analysis, the concentration of acetate was 0.25 mM and the flow rate was maintained at 0.9 ± 0.1 mL h^{-1}. The anaerobic medium after sparging with nitrogen gas for 15 min was pumped into the biosensors using a multi-channel peristaltic pump (BT100-1L, Baoding Longer Precision Pump Co., Ltd., Baoding, China) in single-pass mode. To evaluate the effects of flow rate and acetate concentration, the flow rate was changed in a range from 0.9 ± 0.1 mL h^{-1} to 4.3 ± 0.2 mL h^{-1}, while the acetate concentration was prepared from 0.016 mM to 1 mM. The corresponding hydraulic retention time (HRT) was in a range from 7 to 32 min, considering a 40% porosity of carbon felt according to the manufacturer instructions. The external resistance was fixed at 5000 Ω, which was close to the internal resistance. A lower external resistance condition could enhance the toxic response (Yi et al., 2019), but also negatively increased the recovery time of the MFC sensors (Stein et al., 2012). In addition, a small external resistance was not applicable to measure the voltage signal accurately (Di Lorenzo et al., 2009). Previous studies have suggested that the output electronic quantity of MFC reactor was increased when the external resistance was closer to the internal resistance, considering a much lower variation of output electricity with time (Aelterman et al., 2008).

The performance of micro-microbial electrochemical sensors for water monitoring was tested with the injection of 10 μL or 100 μL of formaldehyde with known concentration into the inlet. High concentration of formaldehyde was tested, considering that during toxic "shocks" the concentration of unwanted release of toxicants could be orders of magnitude higher than its regular conditions (Lazzarini Behrmann et al., 2020). During water monitoring, the external resistance was not connected, and thus these sensors were under the open circuit mode. The open circuit mode has been successfully applied in microbial electrochemical sensors for nitrate monitoring in aquatic environment (Wang et al., 2018). All biosensors were operated in duplicate and in room temperature (28 ± 2 °C).

2.2. Analyses and calculations

The cell voltages were recorded using a battery testing system (CT-4008, Neware, Shenzhen, China). The electrochemical impedance

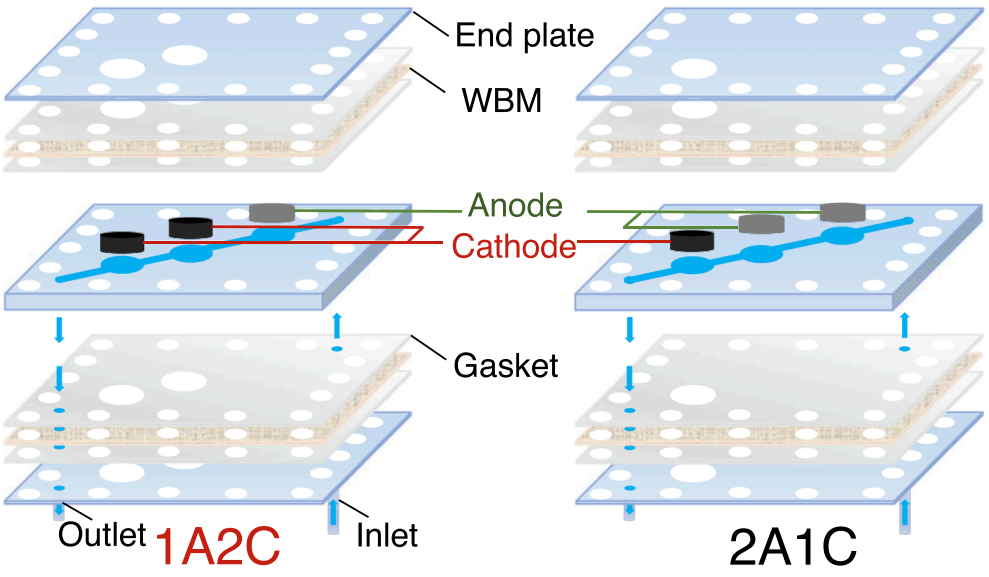

Fig. 1. Schematic of micro-microbial electrochemical sensors using both bioanode and biocathode as the sensing element. The ratio of number of bioanode to biocathode was 1:2 (1A2C) or 2:1 (2A1C). WBM: waterproof breathable membrane.

spectroscopy (EIS) was conducted with an electrochemical workstation in the two-electrode mode (CHI 660E, Chenhua Instrument Co., Shanghai, China), by using cathode as the working electrode while anode as the counter and reference electrode (Guo et al., 2020). The EIS were evaluated with a frequency range changed from 100,000 to 0.010 Hz, while the voltage amplitude was fixed at 10 mV. The measured EIS data was fitted with ZView software (version 3.1, Scribner Associates, Inc.) using an equivalent circuit model. The polarization curve was obtained by conducting the linear sweep voltammetry (LSV) at a scan rate of 1 mV s^{-1}, using a multichannel electrochemical workstation (CHI 1030C, Chenhua Instrument Co., Shanghai, China) (Chu et al., 2020b). During the polarization curve test, the cathode was selected as the working electrode, and the anode was served as both the reference and counter electrode. The current density and power density were calculated with the values normalized to the projected surface of the chamber. The microbial diversity was analyzed using the extracted DNA from both the bioanode and biocathode, and the PCR amplification was conducted with the primer 338F/806R before sequencing using the

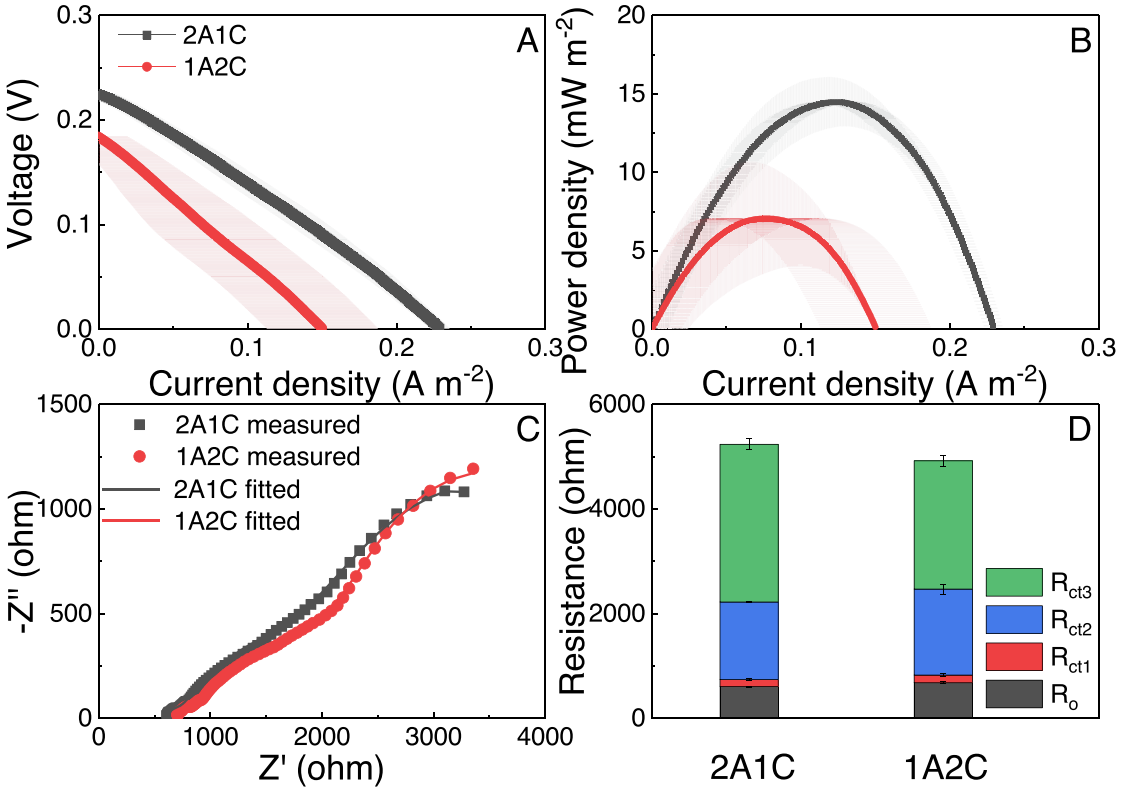

Fig. 2. Polarization curves (A), power density curves (B), EIS data (C), and internal resistance analysis with fitted results (D). Light colored bands indicate the range of the data for duplicate biosensors.

commercial Illumina MiSeq platform (Majorbio Bio-pharm Technology Co., Ltd., Shanghai, China).

To reflect the fluctuation of electric signal during toxic challenging, the *Response* (mV $\%^{-1}$) of the microbial electrochemical sensor was calculated according to (Jiang et al., 2018b; Xing et al., 2020):

$$Response = \frac{|U\max - U\min|}{c}$$

where U_{max} (mV) and U_{min} (mV) refers to the maximum voltage and minimum voltage during toxic events, while c (%) is the formaldehyde concentration. Absolute values were calculated here, as generally the electric signal was increased firstly, and then decreased immediately with the injection of formaldehyde.

3. Results and discussion

3.1. Power generation and internal resistance analysis

The power generation determines the capability of electric signal output of these micro-microbial electrochemical sensors, while the internal resistance analysis indicates different contributions of kinetic limitations. From the polarization (Fig. 2A) and power density curves (Fig. 2B), it revealed that the maximum current of 2A1C biosensors was 0.23 ± 0.01 A m^{-2} in comparison to the 1A2C biosensors that reached 0.15 ± 0.04 A m^{-2}, showing 53% increase in current generation. The open voltage of 2A1C biosensors (0.22 ± 0.01 V) was slightly higher than that of 1A2C (0.18 ± 0.02 V). In addition, the maximum power density of 2A1C biosensors (14.5 ± 1.5 mW m^{-2}) was much higher than that of 1A2C (7.0 ± 3.5 mW m^{-2}). These values of maximum current and power density were close to previous studies on self-powered microbial electrochemical sensors (Jiang et al., 2018a; Zhao et al., 2019). However, the power generation of biosensors in the present study was one to two orders of magnitude lower than previous studies on microbial electrochemical sensors with a similar dimension but with the equipment of ions exchange membranes to prevent the organic substrate and oxygen crossover (Jiang et al., 2017a, 2017b). The EIS measured data and fitted results indicated that the internal resistance of 2A1C biosensors (5200 ± 130 Ω) was close to that of 1A2C (4900 ± 15 Ω) (Fig. 2C and Fig. 2D). The ohmic resistance (R_o) was about 600 Ω, which was mostly caused by the long electrode distance and the small area for ion diffusion. The charge transfer resistance (R_{ct}) accounted for the largest proportion of the internal resistance, which indicated that the electroactivity of both bioanode and biocathode were largely suppressed (Zhang et al., 2019).

3.2. Effects of flow rate and acetate concentration

The effects of acetate loading with different flow rates and acetate concentrations were detailly evaluated, considering that the medium was pumped into the micro-microbial electrochemical sensors in a single-pass mode. It revealed that the voltage output of these sensors was largely affected by both the flow rate and acetate concentration (Fig. 3). The voltage of 2A1C achieved its maximum value with an acetate concentration of 0.25 mM, while in the context of 1A2C, the voltage with an acetate concentration of 0.25 mM was close to that of 0.063 mM. The voltage signal achieved its maximum value at a low flow rate of 0.9 ± 0.1 mL h^{-1} for both biosensors. The electric voltage output of these biosensor was close to previous studies with similar dimension (Di Lorenzo et al., 2014), or with similar structure (Zhao et al., 2019).

Within such sensor equipped with a sequential flowing membrane-free channel, there is a clear trade-off between the electroactivity stimulation of bioanode vs. the electroactivity maintenance of biocathode. The concentration of organic matter could affect the electroactivity of bioanode during both the startup (Li et al., 2020) and operation processes (Cai et al., 2019). For example, the current output of an enriched bioanode was generally increased with the increase of organic matter concentration before saturation (Guo and Liu, 2020). A high acetate loading with the increased flow rate and acetate concentration could stimulate the electroactivity of bioanode. However, the undegraded acetate from the outlet of anodic chamber could negatively affect the inward EET of biocathode, which could be much more serious at a high flow rate and a high acetate concentration. Previous studies have observed that a high concentration of organic matter could stimulate the growth of aerobic heterotrophs with the consumption of dissolved oxygen (Jiang et al., 2017a), thus, deteriorate the cathodic electron consumption (Zhao et al., 2019). Collectively, the increase of acetate loading with the increased flow rate or acetate concentration could inhibit the electric signal generation of both 2A1C and 1A2C biosensors. In addition, the electroactivity of bioanode required further improvement to decrease the inhibition of undegraded acetate to biocathode.

3.3. Responses of biosensors to formaldehyde shocks

The responses of biosensors to formaldehyde shocks were tested under a flow rate of 0.9 mL h^{-1}, a acetate concentration of 0.25 mM, and an open circuit mode, to amplify the voltage output of these sensors. With the injection of 10 μL of 0.25%, 0.5%, and 1% formaldehyde into the inlet, both the 2A1C and 1A2C biosensors could deliver a clear toxic response (Fig. 4A). In addition, the 1A2C biosensors achieved a similar or even higher toxic response than that of 2A1C biosensors (Fig. 4B), although the baseline voltage signal of 1A2C biosensors (180 ± 4 mV) was much lower than that of 2A1C biosensors (216 ± 1). For instance, the *Responses* of 1A2C biosensors (125 ± 1 mV $\%^{-1}$) was higher than that of 2A1C biosensors (87 ± 17 mV $\%^{-1}$) with the injection of 10 μL of 0.25% formaldehyde. These results indicated that the biocathode

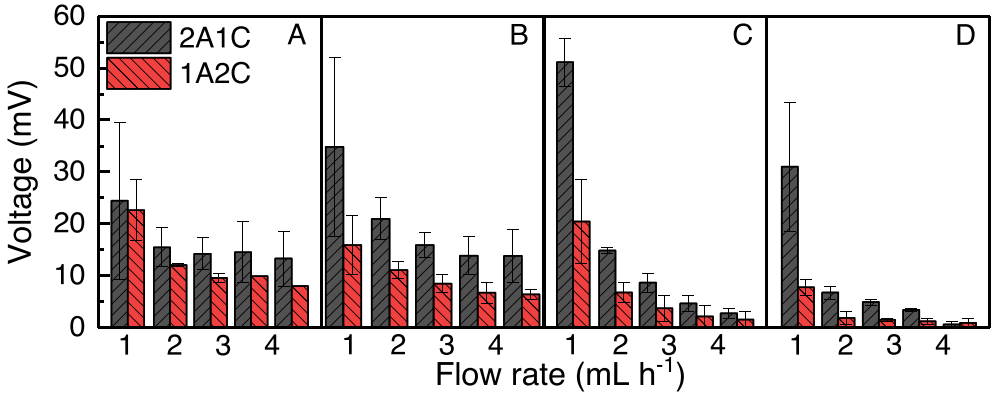

Fig. 3. The stabilized voltage generation at different flow rates and acetate concentrations. The acetate concentration was 0.016 mM (A), 0.063 mM (B), 0.25 mM (C), and 1 mM (D), respectively. The external resistance was 5000 Ω.

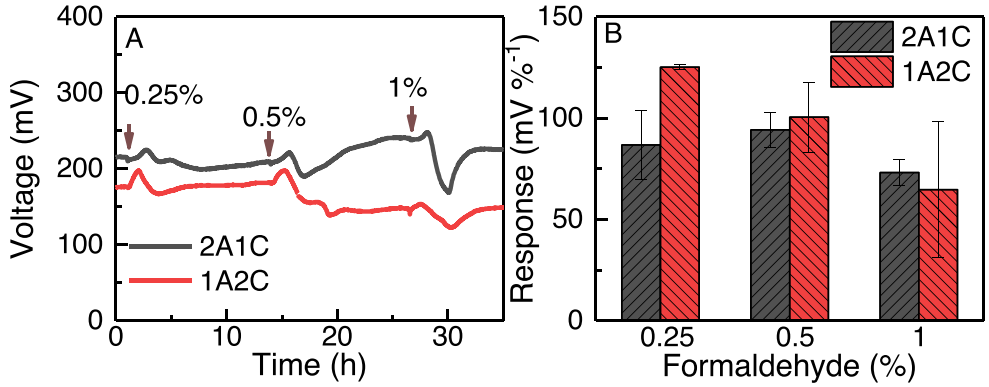

Fig. 4. Responses of biosensors to formaldehyde shocks under open circuit mode. Evolution of voltage during the injection of 10 μL formaldehyde solution into the inlet (A). The concentration of formaldehyde solution was marked with numbers. The response of biosensors to different concentrations of formaldehyde (B).

sensing element could have a high sensitivity than that of the bioanode sensing element, which was in agreement with previous studies (Jiang et al., 2017a; Prevoteau et al., 2019). In addition, these results revealed that the increase in the ratio of number of bioanode to biocathode could stimulate the power generation, while the decrease in such ratio might achieve a higher response for toxicant shocks. The constructed biosensors could also make response to the shocks with the injection of 100 μL of 0.025%, 0.05%, and 0.1% formaldehyde into the inlet. However, the injection process might cause the unnecessary fluctuation of signal, due to the instant change of flow rate. The concentration of formaldehyde, which could be detected by microbial electrochemical sensors, varied largely among previous studies in a range from 0.001% to 1.8% (Chu et al., 2021). In addition, most of these studies were conducted with a prepared medium with a known concentration of formaldehyde, which was different from the present study that a certain small volume of formaldehyde solution was injected into the inlet. The injection of a certain volume of solution for chemical analysis is commonly used in chromatography instruments (Do et al., 2020; Hao et al., 2020). The limit of formaldehyde that could be detected using such sensors was calculated to be 0.01% based on one-dimensional dispersion condition (Sauty, 1989). The application of open circuit mode with a low electrode potential might deteriorate the activity of exoelectrogen, because of the absence of electron acceptors (Logan et al., 2019). However, no significant degradation of electroactivity was observed in a previous study after operation for 3 months in open circuit mode (Wang et al., 2018). The micro-microbial electrochemical sensors had been operated for 2 months after the startup process, while the long-term performance was out of the scope of the present study. To avoid the potential limitation of open circuit mode, the micro-microbial electrochemical sensors could be connected to an appropriate external resistor to maintain the electroactivity during the recovery process after the toxic shocks.

3.4. Microbial diversity

Most of the microbial community of bioelectrodes belonged to the following phyla (Fig. 5A): *Proteobacteria* (52.1–72.8%), *Bacteroidetes* (9.8–47.0%), *Actinobacteriota* (5.1–14.1%), *Firmicutes* (0.4–2.5%), and *Desulfobacterota* (0.5–2.9%). Following the flow direction of the medium, the relative abundance of *Bacteroidetes* was generally increased, with a decrease of *Proteobacteria* and *Actinobacteriota*. Some of *Bacteroidetes* and *Proteobacteria* have been proved to be electroactive microorganisms, and are commonly found to dominate in the biocathode of MFC reactors (Liao et al., 2018b; Xu et al., 2021a). At the genus level (Fig. 5B), *Geobacter*, a well-known electroactive microorganism (Jiang and Zeng, 2019; Logan et al., 2019), was found with a relative abundance of 0.01–0.5% in these bioelectrodes. Following the flow direction of the medium, the relative abundance of *norank_f_env.OPS_17* and *Acidovorax* generally increased, with a decrease of *Mycobacterium* and *Delftia*. The *Acinetobacter*, which has suggested to be capable of inward EET (Liao et al., 2018a), was also enriched at biocathodes in the present study (Liao et al., 2018a). A high microbial diversity of biocathode was found here, which agreed with previous studies on MFC sensors equipped with a biocathode sensing element (Liao et al., 2018b; Prevoteau et al., 2019). However, a very high microbial diversity of bioanode was also observed in the present study, which was different from previous studies on MFC reactors for high power generation (AlSayed et al., 2020). The low power generation and the application of open circuit mode could be the main reasons of the high microbial diversity, as these

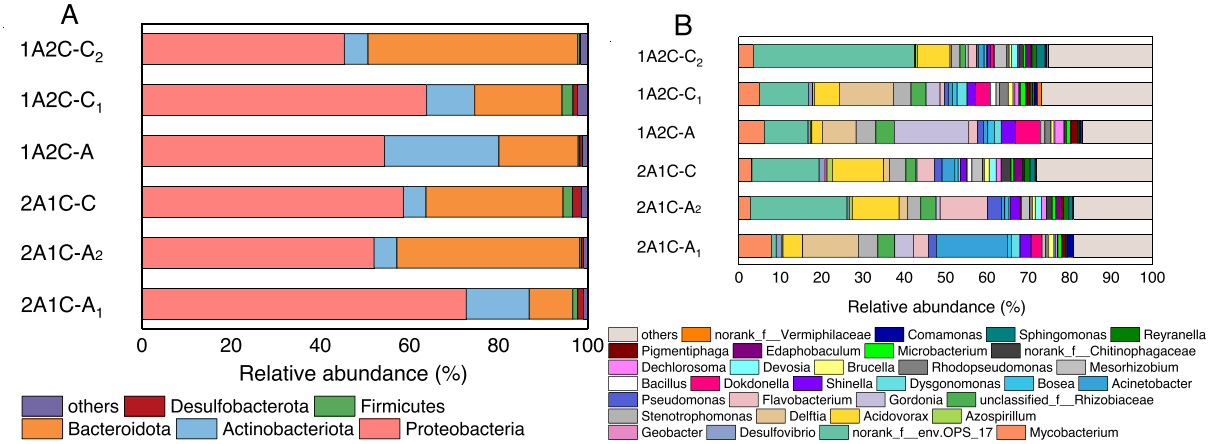

Fig. 5. Bacterial community at the phylum level (A), and at the genus level (B).

conditions might provide a poor selectivity for the enrichment of electroactive microorganisms (Chung et al., 2020; Logan et al., 2019). Further studies are needed for reactor configuration optimization to further increase the performance of these biosensors. For example, electrode structure design, which is rarely investigated in the fabrication of biocathode, could be used to enhance the mass transfer, decrease the startup-time, and promote the microbe-electrode interface (Xu et al., 2021b). Also, the effects of environmental parameters including the daily temperature cycles on the performance of such biosensor should also be tested (Prevoteau et al., 2019). Finally, the long-term performance of the low-cost micro-microbial electrochemical sensor should be assessed, and the emphasis might be on the tolerance mechanisms of bioelectrode (Qi et al., 2021).

4. Conclusions

Novel micro-microbial electrochemical sensors using both bioanode and biocathode as the sensing element were constructed, characterized by a sequential flowing membrane-free channel and a bilateral passive oxygen supply. The voltage output was largely affected by both the flow rate and acetate concentration. The 1A2C biosensors with a much poor power generation could achieve a similar or even higher toxic response than that of 2A1C biosensors. A high microbial diversity of bioelectrodes was observed. Further studies are needed for reactor configuration optimization with a shaped microbial community to further increase the performance of these biosensors.

CRediT authorship contribution statement

Na Chu: Conceptualization, Investigation, Writing - original draft, Writing - review & editing. **Qinjun Liang:** Writing - review & editing. **Wen Hao:** Writing - review & editing. **Yong Jiang:** Conceptualization, Investigation, Methodology, Supervision, Writing - original draft. **Raymond Jianxiong Zeng:** Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Notes

Declarations of interest: None.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.biortech.2021.124743.

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