



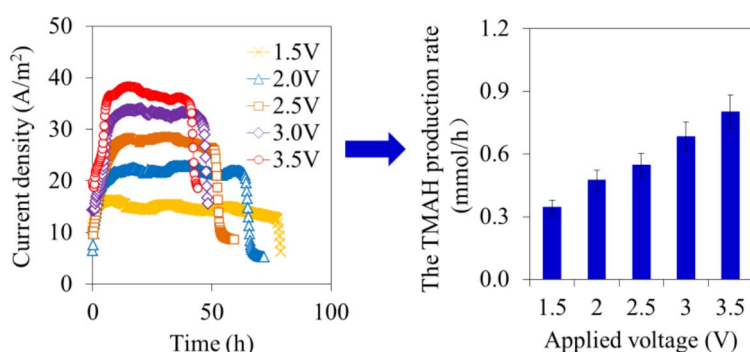
Short Communication

Tetramethyl ammonium hydroxide production using the microbial electrolysis desalination and chemical-production cell with long anode

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GRAPHICAL ABSTRACT



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ABSTRACT

The aim of this study was to investigate the feasibility to improve the tetramethyl ammonium hydroxide (TMAH) production in the microbial electrolysis desalination and chemical-production cell (MEDCC) with long anode of 48 cm. Different concentrations of tetramethylammonium chloride (0.3–0.7 M) and applied voltages (1.5–3.5 V) were tested in the MEDCC. With 0.6 M of tetramethylammonium chloride as the raw material and under the applied voltage of 3.5 V, the maximum TMAH production rate in the MEDCC reached 1.13 ± 0.12 mmol/h, which was 9.4 times higher than those previously reported in the MEDCCs. The maximum current density of 41.0 ± 4.0 A/m² in the MEDCC was obtained, which was the highest value in the bioelectrochemical systems using the carbon cloth or carbon brush as the anode so far. Our results should provide a promising method to improve the TMAH production and boost the MEDCC application.

1. Introduction

The bioelectrochemical system (BES) is a device that the anode or cathode reactions can be catalyzed by electrochemically active bacteria (EAB) (Rozendal et al., 2008). Diverse BESs have been developed for applications of wastewater treatment, desalination, and chemical production. As a typical BES, the microbial electrolysis desalination and

chemical-production cell (MEDCC) has been developed to utilize the electricity from substrate biodegradation by EABs for water desalination, and acid or alkali production. The MEDCC can produce the highest malic acid rate with the lowest energy consumption compared with other BESs, such as the microbial electrolysis desalination cell (MEDC) and the modified microbial desalination cell (MDC) (Liu et al., 2015). The distinguished performance of MEDCC can be attributed to the

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synergistic effect of EABs and bipolar membrane (BPM), which keeps a neutral pH in the anode chamber and results in high activity of EABs (Feng et al., 2008; Liu et al., 2015).

As a strong quaternary ammonium alkali, tetramethyl ammonium hydroxide (TMAH) is widely used in the processes of chemical synthesis and semiconductor production (Feng et al., 2008). The market value of TMAH is much higher than those of other products in the BESs, such as sodium hydroxide and hydrogen (Feng et al., 2008; Logan & Rabaey, 2012). Therefore, TMAH production in the MEDCC may balance the costs to construct the system and boost applications of the BESs (Liu et al., 2014; Logan & Rabaey, 2012). The main reactions involving TMAH production in the MEDCC have been reported by Liu et al. (Liu et al., 2014). To enhance the TMAH production and current density in the MEDCC, large surface areas of BPM, cation exchange membrane (CEM), and anion exchange membrane (AEM) are required, which increase the construction costs (Feng et al., 2008; Liu et al., 2014). Our recent results demonstrated that the maximum current density of MEDCC could be significantly increased by enlarging the anode length using 35 g/L NaCl solution in the desalination chamber (Ye et al., 2017). With an anode length of 48 cm, the maximum current density in the MEDCC reached 32.8 A/m² under the applied voltage of 2.5 V (Ye et al., 2017). Therefore, it should be useful to examine the TMAH production in the MEDCC with the enlarged anode. The objective of this study was to investigate the feasibility to improve the TMAH production in the MEDCC with anode length of 48 cm. The TMAH production rate, energy consumption, and current efficiency were calculated and discussed.

2. Materials and methods

2.1. MEDCC setup and operation

The MEDCC reactor was made of plexiglass with BPM (Fumasep-FBM, Fumatech, German), CEM (Ultrax CMI-7000, MI, USA), and AEM (Ultrax AMI-7001, MI, USA) as separators (Chen et al., 2012; Liu et al., 2014; Liu et al., 2015). The carbon brush anode of MEDCC was made with carbon fiber (SYT45, Zhongfu Shenying Carbon Fiber Co. Ltd., China). Each carbon fiber with a length of 48 cm and a width of 3 cm for the carbon brush anode was pretreated with 450 °C for 30 min. The cathode in the MEDCC was composed of a catalyst layer, a supporting layer with stainless steel meshes, and a diffusion layer. In the cathode, the activated carbon (SPC-01, Xinsen, China) was used as cathodic catalyst. The effective area of each membrane and cathode was 7 cm². The anode chamber in the MEDCC had an effective volume of 336 mL. A plastic magnetic pump (MP-6R, Shanghai Magnetic Pump manufacture Co. Ltd., China) was used to recycle the flow in the anode chamber of MEDCC with an outside storage tank (400 mL) at a flow rate of 2.8 L/min, resulting in the hydraulic retention time (HRT) of 7.2 s. The acid-production chamber, desalination chamber, and cathode chamber had the same effective volumes of 1 mL. In each of the acid-production, desalination and cathode chambers, a peristaltic pump (BT1-200, Shanghai Qite Pump manufacture Co. Ltd., China) was used to recycle the flow with corresponding storage tank (300 mL) at a flow rate of 0.2 mL/min, resulting in the same HRT of 300 s. An external resistance of 10 Ω was connected with the anode and cathode leads of the MEDCC.

The MEDCC started up at an applied voltage of 1.2 V as previously described (Chen et al., 2012; Liu et al., 2014; Liu et al., 2015). To reduce the internal resistance of MEDCC, the acid-production chamber with the storage tank was refreshed with 1 g/L of NaCl solution and the cathode chamber with the storage tank was refreshed with 0.01 M of the tetramethyl ammonium chloride (TMAH, Sinopharm Chemical Reagent Co. Ltd., China) solution at the beginning of each batch cycle. Different concentrations (i.e., 0.1–0.6 M) of tetramethylammonium chloride (TMAC) solutions with the initial pH of 7.0 were tested in the desalination chamber, respectively. Applied voltages in the MEDCC

gradually increased from 1.5 to 3.5 V using a power supply (Itech, IT6700, China). Acetate solutions of 1 and 2 g/L were used as substrate in the MEDCCs under the applied voltages of 1.5–2.0 V and 2.5–3.0 V, respectively. The anode solution was refreshed with sodium acetate, 50 mM phosphate buffer, and vitamins (Chen et al., 2012). The MEDCC was operated at least two cycles at each applied voltage. All the solutions in the MEDCC were refreshed when the current density in the MEDCC was lower than that in the abiotic MEDCC. All the experiments were carried out at 25 ± 5 °C in duplicate.

2.2. Analyses and calculations

The conductivity and pH were determined using a conductivity meter (FE 30, Mettler Toledo, Swiss) and a pH meter (FE 20, Mettler Toledo, Swiss) respectively. The TMAH concentration was measured using the titration method (Feng et al., 2008). The TMAH production rate (mmol/h) was calculated according to the final TMAH concentration, the effective volumes of the cathode chamber and storage tank (305 mL), and the operation time (h). The biomass on the carbon brush anode was determined using the Coomassie Brilliant Blue method (Ye et al., 2017). Voltages across the external resistance were recorded using a data acquisition system (model 2700; Keithley Instruments, Inc.) each 15 min. The current density (A/m²) was calculated with the current normalized by the projected area of cathode electrode (Luo et al., 2017). The current efficiency (CE) and total energy consumption (including the electricity consumption and bio-energy consumption from substrate utilization) were determined as previously reported (Luo et al., 2017).

3. Results and discussion

3.1. Effect of applied voltages on the performance of MEDCC

The current densities in the MEDCC varied with the different applied voltages (Fig. 1A). With the initial concentration 0.3 M of TMAC, the maximum current density in the MEDCC increased from 15.2 ± 2.3 to 35.5 ± 3.5 A/m² with increase of the applied voltages from 1.5 to 3.5 V. The maximum current densities in the MEDCC were 20.6 ± 2.0, 25.7 ± 3.2, and 31.4 ± 2.5 A/m² with 2.0, 2.5, and 3.0 V, respectively. However, the operation time for one cycle in the MEDCC decreased from 80 ± 20 to 46 ± 4 h with the applied voltages from 1.5 to 3.5 V, indicating that a higher applied voltage enhanced electrons released from the electrochemically active bacteria. The maximum current density in the MEDCC were comparable to that in previously reported (35.5 ± 3.5 vs. 32.8 ± 2.6 A/m²) (Ye et al., 2017). The applied voltage of 3.5 V in this study was the highest applied voltage on the MEDCC with a stable electricity generation so far.

The final TMAH concentrations increased from 0.13 to 0.17 M at the end of one cycle with the applied voltages from 1.5 to 3.5 V. The average TMAH production rate was calculated based on the TMAH concentration and running time. As shown in Fig. 1B, the maximum TMAH production rate of 0.80 ± 0.10 mmol/h was obtained under 3.5 V among all the tests, which was 2.3 times as that under 1.5 V (0.35 ± 0.05 mmol/h). The TMAH production rate was a linear function of the applied voltage ($R = 0.996$). Among the treatments, the minimum total energy consumption was 0.99 ± 0.06 kWh/kg under 1.5 V (Fig. 1C). With the increase of applied voltages, the total energy consumption increased from 0.99 ± 0.06 kWh/kg (1.5 V) to 1.69 ± 0.10 kWh/kg (3.5 V). The electricity consumption accounted for 53 ± 3% of the total energy consumption under 1.5 V. The electricity consumption under 2.0 and 3.5 V accounted for 64% and 75% of the total energy consumption, respectively. Correspondingly, the bioenergy consumption from the substrate utilization in the MEDCC was in a range of 0.39 ~ 0.46 kWh/kg, indicating that the activities of electrochemically active bacteria were relatively stable under the different applied voltages. The CEs in the MEDCC reached 80% ~ 85%

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