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Manipulating the hydrogen production from acetate in a microbial electrolysis cell-microbial fuel cell-coupled system

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ABSTRACT

A biological hydrogen-producing system is configured through coupling an electricity-assisting microbial fuel cell (MFC) with a hydrogen-producing microbial electrolysis cell (MEC). The advantage of this biocatalyzed system is the *in-situ* utilization of the electric energy generated by an MFC for hydrogen production in an MEC without external power supply. In this study, it is demonstrated that the hydrogen production in such an MEC–MFC-coupled system can be manipulated through adjusting the power input on the MEC. The power input of the MEC is regulated by applying different loading resistors connected into the circuit in series. When the loading resistance changes from 10Ω to $10 k\Omega$, the circuit current and volumetric hydrogen production rate varies in a range of 78 ± 12 to 9 ± 0 mA m⁻² and 2.9 ± 0.2 to 0.2 ± 0.0 mL L⁻¹ d⁻¹, respectively. The hydrogen recovery (R_{H_2}), Coulombic efficiency (CE), and hydrogen yield (Y_{H_2}) decrease with the increase in loading resistance. Thereafter, in order to add power supply for hydrogen production in the MEC, additional one or two MFCs are introduced into this coupled system. When the MFCs are connected in series, the hydrogen production is significantly enhanced. In comparison, the parallel connection slightly reduces the hydrogen production. Connecting several MFCs in series is able to effectively increase power supply for hydrogen production, and has a potential to be used as a strategy to enhance hydrogen production in the MEC–MFC-coupled system from wastes.

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1. Introduction

Hydrogen is widely recognized as an efficient renewable energy carrier because of its numerous advantages. Microbial hydrogen production using fermentative, photosynthetic bacteria, or algae is an environmentally friendly and energy saving process, and it has recently attracted considerable attention as a way of converting organic wastes to hydrogen effectively [1–3]. Recently, microbial electrolysis cell (MEC) has been demonstrated to be a promising technique for biological hydrogen production from wastes [1–9]. Compared with the fermentative hydrogen-producing reactor from wastes, the MEC has a higher hydrogen recovery and a wider substrate diversity. A lower energy consumption and no need of expensive anodic catalysts are the main advantages of the MEC over the water electrolyer [9]. The MEC is developed on the base of microbial fuel cell (MFC) [10-13], which employs electrochemically active microorganisms to oxidize hydrogen-containing substances that are otherwise unable to be utilized in a usual fuel cell under

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mild conditions [14–18]. In an MFC, the microorganisms in the anode oxidize organic matters and produce electrons and protons, and the electrons flow from the anode to the cathode through an electrical circuit and then react with the protons in solution and the oxidant such as oxygen. In such a system electricity can be produced spontaneously. In an MEC, no oxidant is present in the cathode, and thus the electrons combine with the protons to generate hydrogen. In this process external power supply is needed to circumvent thermodynamic constraints. Usually, an external voltage of 0.6 V or more is applied for microbial hydrogen production [5–7].

Since the open circuit voltage of an MFC reaches as high as nearly 0.80 V [19], the extra energy needed in an MEC can be supplied by an MFC. With such an idea, we have developed an MEC–MFC-coupled system for biological hydrogen production from wastes [20]. This system was composed of one coupled MEC and MFC: the electrolysis was performed in an MEC designed according to Liu et al. [1], whereas the extra electricity for the electrolysis was supplied by an MFC with air cathode. The advantage of this system is the *in-situ* utilization of the electric energy of the related MEC–MFC as well as hydrogen production without an external power supply.

In MEC-oriented studies, attention is usually paid on the relationship between applied voltage and hydrogen production. It has

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been widely accepted that the hydrogen production and MEC performance are enhanced at a higher applied voltage [1,2,5,8]. It is because the current density is increased with the increasing voltage and a larger amount of energy is input into the MEC [21]. For an MEC, the external power supply helps to overcome the thermodynamic barrier and thus realize hydrogen generation. In theory the hydrogen production should be positively associated with the power input. In a conventional MEC, the power input can be easily manipulated by imposing an appropriate level of voltage. However, in the MEC–MFC-coupled system, the input power of the MEC is affected by both performance of the MFC and MEC, and can only be regulated indirectly. Therefore, an introduction of appropriate methods to manipulate the input power, and thus hydrogen production, is of great benefit to the successful application of an MEC–MFC-coupled system.

In this work, the performance of the MEC–MFC-coupled system, especially hydrogen production, was investigated with different power inputs on the MEC. The power input was regulated in the following two ways: (1) adjusting the loading resistors connected across the circuit in series; and (2) employing several MFCs as a power supply, which were connected in series or parallel. Based on the experiment results, a strategy for enhanced hydrogen production was proposed.

2. Materials and methods

2.1. Reactor and operation

The MEC–MFC-coupled system was composed of a hydrogenproducing MEC and an electricity-assisting MFC, as described previously [20]. In the MEC, the cathode electrode was made of carbon paper with Pt on it ($4 \text{ cm} \times 4 \text{ cm}$, 2 mg cm^{-2}). The anode was plain carbon paper ($4 \text{ cm} \times 4 \text{ cm}$, not wet proofed). In the MFC, the cathode was carbon paper with pt on it ($2 \text{ cm} \times 2 \text{ cm}$, 2 mg cm^{-2}). The anode was plain carbon paper ($4 \text{ cm} \times 4 \text{ cm}$, not wet proofed). All the electrode materials were purchased from the GEFC Co. (China).

The anode chamber of each reactor was filled with 350 mL of autoclaved medium containing (in 1 L of pH 7.0 phosphate buffer,): NaAc, 100 mg; NH₄Cl, 310 mg; KCl, 130 mg; CaCl₂, 10 mg; MgCl₂·6H₂O, 20 mg; NaCl, 2 mg; FeCl₂, 5 mg; CoCl₂·2H₂O, 1 mg; MnCl₂·4H₂O, 1 mg; AlCl₃, 0.5 mg; (NH₄)₆Mo₇O₂₄, 3 mg; H₃BO₃, 1 mg; NiCl₂·6H₂O, 0.1 mg; CuSO₄·5H₂O, 1 mg; ZnCl₂, 1 mg. The cathode chamber of the MEC was filled with 350 mL of pH 7.0 autoclaved phosphate buffer. The phosphate buffer solution was 10 mM when the effects of loading resistance on system performance were investigated, and 50 mM when two/three MFCs were applied as power supply.

The anode biofilms of the MFC–MEC had been cultivated for over 10 months with anaerobic sludge as inoculums and acetate as substrate. The MEC and MFC were connected in series with a 10 Ω resistor, unless mentioned otherwise. All tests were conducted in duplicate at 30 °C.

When additional one or two MFCs were introduced into this coupled system, in the series connection (Fig. 1A), where the MFCs and MEC were connected in series, the current was identical in the circuit. In the parallel connection (Fig. 1B) the two MFCs were initially connected in parallel and later connected with the MEC in series. The current in the MEC was the sum of currents in the two MFCs. The system with three MFCs had a similar current distribution.

2.2. Analysis and calculation

The circuit current was calculated based on the voltage across the loading resistor. The resistor voltage was continuously recorded with an electrochemical workstation (660C, CH Instruments, Inc., USA) connected to a computer. The output voltage of the MFC was measured by a multimeter with a data acquisition system (UT39A, UNIT Inc., China). Acetate concentration in the solution was measured using a gas chromatograph (Model 6890NT, Agilent Inc., USA) equipped with a flame ionization detector and a $30 \text{ m} \times 0.25 \text{ mm} \times 0.25 \mu\text{m}$ fused-silica capillary column (DP-FFAP). The hydrogen production was analyzed using another gas chromatograph (Model SP-6800A, Lunan Co., China) equipped with a thermal conductivity detector and a 1.5 m stainless-steel column packed with 5 Å molecular sieves [22]. The gas was sampled using a gastight syringe of 1 mL (SGE Syringe). The hydrogen amount in 1 mL gas sample was converted to hydrogen volume under standard conditions, and was then multiplied by headspace volume of MEC cathode chamber (100 mL) to get the total hydrogen volume.

System performance was evaluated in terms of volumetric hydrogen production rate based on the total MEC volume, the cathodic hydrogen recovery (R_{H_2}), the Coulombic efficiency (CE), the hydrogen yield (Y_{H_2}) of MEC (Y_{MEC-H_2}), and the overall systemic hydrogen yield (Y_{sysH_2}).

 R_{H_2} is calculated as $R_{H_2} = n_{H_2}/n_{Th}$, where n_{H_2} is the moles of hydrogen harvested, $n_{H_2} = V_{H_2}/RT$; and n_{Th} is the moles of hydrogen that could be produced from the measured current, $n_{Th} = C_p/2F$. C_p is the total Coulombs calculated by integrating the current over time, V_{H_2} is the measured hydrogen volume, R is the gas constant, T is the absolute temperature, and F is Faraday's constant. The mean circuit current density I is calculated as $I = C_p/86,400A_{MEC}$, where A_{MEC} is the anode surface area of the MEC (32 cm⁻²). CE, Y_{MEC-H_2} , and Y_{sysH_2} are respectively calculated as $C = n_{Th}/(4n_M)$, $Y_{MEC-H_2} = n_{H_2}/n_M$, and $Y_{sysH_2} = n_{H_2}/n_s$, where n_M and n_s are the moles of acetate consumed in the MEC and total system, respectively. The CE is calculated based on the assumption that 8 mol of electrons are produced from 1 mol of acetate in the MEC and [1].

3. Results and discussion

3.1. Hydrogen production under varied loading resistances

The ratio between the current generation and the cell voltage is partially governed by the external resistance [12]. A high external resistance shares a high cell voltage, while the circuit current is low [23]. The coupled system was operated with different resistors connected in series across the circuit. It was observed that a higher resistance resulted in a lower volumetric hydrogen production rate. A maximum hydrogen production rate of $2.9 \pm 0.2 \text{ mL L}^{-1} \text{ d}^{-1}$ was observed at a loading resistance of 10Ω , and a minimum of $0.2 \pm 0.0 \text{ mL L}^{-1} \text{ d}^{-1}$ at a resistance of $10 \text{ k}\Omega$. Similarly, the hydrogen yield decreased with the increase in loading resistance (Fig. 2). Comparing with the $Y_{\text{MEC-H}_2}$ of $1.98 \pm 0.14 \text{ mol-H}_2 \text{ mol-acetate}^{-1}$ at 10Ω and $2.12 \pm 0.05 \text{ mol-H}_2 \text{ mol-acetate}^{-1}$ at $10 \Omega \Omega$, it decreased by a factor of 7 to $0.26 \pm 0.03 \text{ mol-H}_2 \text{ mol-acetate}^{-1}$ at $10 \text{ k}\Omega$. The Y_{sysH_2} at 10Ω was almost nine times of that at $10 \text{ k}\Omega$, i.e., $1.02 \pm 0.14 \text{ mol-H}_2 \text{ mol-acetate}^{-1}$.

At various loading resistances tested in the experiment, the R_{H_2} varied from 105.71% to 44.50%, and the CE was in a range of 25.11–6.17% (Fig. 3). The R_{H_2} was over 90% when the resistance was below 1 k Ω ; and it declined gradually with the increment of resistance. When a higher loading resistor is applied, the electron transfer through the circuit might be lower than the electron consumption in the cathode [24]. Under that circumstance, the electron transfer rate becomes the rate-limiting step. When the electron transfer is limited, the electrons flowing to the cathode of MEC are preferably consumed by the alternative electron acceptors such as oxygen diffused through the sampling ports [20]. Furthermore, the hydrogen may be lost via diffusion through membrane or sampling



Fig. 1. Working principles of the MFC-coupled biocatalyzed electrolyzer with two electricity-assisting MFCs connected (A) in series; and (B) in parallel.



Fig. 2. Hydrogen production rate and hydrogen yield as a function of loading resistance.

ports [2]. When the hydrogen production rate is low, the hydrogen loss becomes appreciable and would of significant influence on the calculated R_{H_2} .

The index CE is an indicator for the cell ability to recover electrons stored in the substrate as currents [21]. It was significantly influenced by the loading resistance, given the fact that a higher resistance results in a lower CE. Some other electron-consuming



In summary, the circuit current decreased with an increase in loading resistance. When the resistance was shifted from $10\,\Omega$ to $10 \text{ k}\Omega$, the circuit current density decreased from 78 ± 12 to 9 ± 0 mA m⁻². Meanwhile, the output voltage of the MFC increased from 366 to 504 mV, whereas the input voltage of the MEC decreased from 364 to 224 mV (Fig. 4). Although a higher resistor can increase the output voltage of the MFC, it shares a higher voltage on it. Consequently, the input voltage of the MEC became even lower. For the MEC, an external power source was used to provide the energy input required for driving the hydrogen production, and the hydrogen production rate was partially dependent on the power input. Fig. 5 shows the distribution of the output power density of the MFC. At the loading resistance of 10Ω , the maximum output power density of the MFC reached 28.6 ± 4.5 mW m⁻². The power consumed on the resistor was the least, and the MEC got the most power input for hydrogen production. So, it was found the hydrogen production rate was the highest. Based on calculation, 99.3% of the power output of MFC was assigned to the MEC. With an increase in loading resistance, the power output of the MFC decreased, and a higher proportion of the power output was spent on the loading resistor. As a consequence, the power input on the MEC dropped. At the loading resistance of $10 \text{ k}\Omega$, the output



Fig. 3. Cathodic hydrogen recovery and CE as a function of loading resistance.



Fig. 4. Circuit current, output voltage of electricity-assisting reactor, and input voltage of hydrogen-producing reactor at different loading resistances.



Fig. 5. Distribution of output power density of the electricity-assisting MFCs.

power density of the MEC decreased to 4.8 ± 0.2 mW m^-2, and only 2.4 ± 0.1 mW m^-2 power was distributed to the MEC.

3.2. Enhanced hydrogen production in a modified system with several electricity-assisting MFCs

The solution conductivity is partially dependent on the ionic strength. A higher ionic strength has been observed to enhance the current production through reducing the ohmic resistance of the electrolyte [25]. In order to increase ionic strength, the phosphate buffer concentration was increased from 10 to 50 mM as usually did in many studies [1,5]. The hydrogen production rate significantly increased to 7.86 ± 0.31 mLL⁻¹ d⁻¹, and the Y_{MEC-H2} and Y_{sysH2} also elevated to 2.50 ± 0.22 and 1.25 ± 0.25 mol-H2 molacetate⁻¹, respectively (Table 1). This improvement was attributed to the increase in circuit current, hence the power input, for hydrogen production. The circuit current was 241 ± 13 mA m⁻² and the power input was 85.7 ± 4.4 mW m⁻². Accordingly, the CE increased to $31.9 \pm 7.2\%$.

In a conventional MEC with an external power supply, the power input could be increased through increasing the input voltage [7]. However, in our coupled system the MEC input voltage was limited to the power output provided by the MFC. The power output could be increased by increasing the cell voltage or the current. Aelterman et al. [26] have tried to increase the voltages and currents produced by MFCs through using series or parallel stacked MFCs. A desired current or voltage could be obtained by combining the appropriate number of series and parallel connected fuel cells or power sources. Connection of several fuel cells in series adds the power supply by multiplying the voltages, while parallel connection increases the power supply by multiplying the currents. In a previous study the stacked MFCs were applied as energy source for the water electrolysis [27]. In this work, 10 fuel cells with pure cultures were connected in two parallel series of five cells to supply sufficient voltage for water electrolysis. The successful production of hydrogen demonstrated that the stacked MFCs were a good energy source for the water electrolysis, although only an energy efficiency of 11% was obtain because of the poor performance of the MFCs.

In order to increase the overall power input on the MEC, additional one or two electricity-assisting MFC units were introduced into the coupled system, which were connected in series or parallel. As shown in Table 1, such an introduction resulted in a current of 334 ± 22 or 418 ± 3 mA m⁻² for the series connection and 263 ± 9 or 253 ± 9 mA m⁻² for the parallel connection. The hydrogen production rate reached a rate of 10.95 ± 0.64 or 14.54 ± 0.12 mLL⁻¹ d⁻¹ for

the series system, and a rate of 7.50 ± 0.09 or 7.50 ± 0.14 mL L⁻¹ d⁻¹ for the parallel system. In the series connection, the *R*, CE, and $Y_{\text{MEC-H}_2}$ all increased, whereas in the parallel connection all of them slightly decreased. The Y_{sysH_2} reflected the amount of hydrogen recovery from the substrates in the whole system including the MEC and the MFCs. The utilization of more MFCs caused a decline of Y_{sysH_2} , and an increase in the number of MFC reduced the Y_{sysH_2} . In each MFC–MEC, a considerable quantity of energy in the substrates was lost in the circuit or consumed by other competitive processes [12], and only a limited amount of energy could be utilized for hydrogen production. Thus, the introduction of more MFCs would decrease the overall hydrogen yield of the whole system.

Theoretically, the circuit current in the coupled system can be calculated according to the Ohm's law. In the system with one MFC, the current I_{single} is calculated as:

$$I_{\text{single}} = \frac{V_{\text{MFC}} + V_{\text{MEC}}}{R_{\text{MFC}} + R_{\text{mec}} + R_{\text{resistor}}}$$
(1)

where V_{MFC} and V_{MEC} are the open circuit voltage of the MFC and MEC (the V_{MFC} is positive, and the V_{MEC} is negative). R_{MFC} , R_{MEC} , and $R_{resistor}$ are the resistance of the MFC–MEC, and loading resistor, respectively.

In the system with two identical MFCs (Fig. 1), the current I_{s-2} in series connection and I_{p-2} in parallel connection are calculated respectively as follows:

$$I_{s-2} = \frac{2V_{MFC} + V_{MEC}}{2R_{MFC} + R_{MEC} + R_{resistor}}$$
(2)

$$I_{\rm p-2} = \frac{V_{\rm MFC} + V_{\rm MEC}}{R_{\rm MFC}/2 + R_{\rm MEC} + R_{\rm resistor}}$$
(3)

In the system with three identical MFCs, the current I_{s-3} in series connection and I_{p-3} in parallel connection are calculated respectively as below:

$$I_{\text{s-3}} = \frac{3V_{\text{MFC}} + V_{\text{MEC}}}{3R_{\text{MFC}} + R_{\text{MEC}} + R_{\text{resistor}}} \tag{4}$$

$$I_{p-3} = \frac{V_{MFC} + V_{MEC}}{R_{MFC}/3 + R_{MEC} + R_{resistor}}$$
(5)

According to the calculations above, addition of other MFCs as a power supply was able to increase the circuit current, and an increase in the number of MFC led to a higher circuits current, no matter the MFCs were connected in series or parallel. In practice, because of the unique property of the electricigens, the MFCs are different from the usual fuel cells. Moreover, the several MFCs in the system are not independent, but were influenced by each other [21]. Fig. 1 illustrates the electron flows in the systems with two electricity-assisting MFCs. The system with three electricityassisting MFCs has the similar electrons flows. The electrons from the anode substrate oxidation in one MFC do not participate in the cathode reaction of the same MFC, but move to be consumed in the cathodes of the other MFCs or MEC. Additionally, the origin and destination of electron flows are different in the series and parallel connections.

3.3. Difference between the MFCs in series and in parallel connection

Table 1 shows that in the series connection the hydrogen production and circuit current were much higher than the others. In the system with two MFCs, the input power density of the MEC was twice as that with single MFC. The introduction of another MFC resulted in additional power input increase by 80%. The series connection greatly increased the input voltage of the MEC, and a higher voltage enhanced the MEC performance, and thus led to a higher

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Index	Hydrogen production rate (mL L ⁻¹ d ⁻¹)	R _{H2} (%)	CE (%)	Y_{MEC-H_2} (mol- H ₂ mol-acetate ⁻¹)	Y_{sysH_2} (mol- H ₂ mol-acetate ⁻¹)	Current density (mA m ⁻²)	Input voltage of MEC (mV)	Input power density of MEC (mW m ⁻²)
One MFC	7.86 ± 0.31	92.0 ± 1.8	31.9 ± 7.2	2.50 ± 0.22	1.25 ± 0.25	241 ± 13	356	85.7 ± 4.4
Two MFCs Series connection Parallel connection	$\begin{array}{c} 10.95 \pm 0.64 \\ 7.50 \pm 0.09 \end{array}$	$\begin{array}{c} 93.2 \pm 0.01 \\ 85.2 \pm 3.6 \end{array}$	36.8 ± 10.2 30.3 ± 4.5	$\begin{array}{c} 2.76 \pm 0.36 \\ 2.05 \pm 0.06 \end{array}$	$\begin{array}{c} 1.09 \pm 0.14 \\ 1.04 \pm 0.14 \end{array}$	334 ± 22 263 ± 9	539 363	$\begin{array}{c} 180.2\pm11.8\\ 95.3\pm3.4\end{array}$
Three MFCs Series connection Parallel connection	$\begin{array}{c} 14.54 \pm 0.12 \\ 7.50 \pm 0.14 \end{array}$	98.0 ± 1.9 82.3 ± 1.5	41.1 ± 1.9 31.5 ± 5.5	$\begin{array}{c} 3.22 \pm 0.06 \\ 2.08 \pm 0.09 \end{array}$	$\begin{array}{c} 0.70 \pm 0.19 \\ 0.85 \pm 0.04 \end{array}$	$\begin{array}{c} 418 \pm 3 \\ 253 \pm 9 \end{array}$	807 455	$\begin{array}{c} 337.9 \pm 2.5 \\ 115.2 \pm 4.3 \end{array}$

Table 1 Performance of an electrolyzer with several electricity-assisting MFCs at a phosphate buffer solution of 50 mM and a loading resistor of 10 Ω .

circuit current and boosted hydrogen production. However, the parallel connection has no positive effect on the hydrogen production (Table 1). It is known that only the cells with the same open circuit voltage and same internal resistance could be connected in parallel. Because of the diversity of the electricigens and the disparity of biofilm, the open circuit voltage and the internal resistance varied among the MFCs. When several electricity-assisting MFCs were connected in parallel, the MFC with the highest open circuit voltage was in discharging and the others were in charging. In the parallel system, only one MFC might virtually supply the power, but the others just behaved as capacitors. As a result, the system equals to a circuit composed of one power supply connected with several parallel electricity-consuming equipments. In other words, the additional MFCs were power consumers, rather than power supplies. In comparison with that in series connection, the unsuitable parallel connection caused a low power input and poor MEC performance. As shown in Fig. 1(B), the electrons for oxygen reduction in the two electricity-assisting MFCs were from the substrate oxidation in the MEC anode. Restricted by the MEC performance, the electricity-assisting MFCs did not work with their maximum ability. Therefore, the system efficiency was even lower than that with a single MFC.

Our study demonstrated that the series connection was more appropriate than the parallel connection for an MEC-MFC-coupled system. It is interesting that the introduction of more electricityassisting MFCs into the series system induced a higher hydrogen production. It is well known that one advantage of the MFC-MEC system is the simultaneous energy production and waste treatment [21]. However, the low electricity output has made the energy in the MFCs difficult to be extracted and utilized. Application of MFCs as a power supply for the MEC to produce hydrogen has provided a prospective way for in-situ utilization of the low energy produced in the MFCs. Furthermore, the waste treatment capacity of the coupled system increases with the introduction of more MFCs. Although the introduction of additional MFCs reduces the overall systemic hydrogen yield, such an introduction is practical feasible, because a higher hydrogen production rate and a more effective waste removal are highly desirable. Connecting several electricity-assisting MFCs in series as power supply has a potential to be used as a strategy to enhance the hydrogen harvest of MEC system from organic wastes.

In this study, acetate was used as the substrate for the MFC–MEC systems. Acetate is an ideal substrate for such a system, and the recalcitrance of many types of wastewater makes them more difficult to be utilized than acetate. When a complex wastewater is used as the substrate, the MFC–MEC has a higher internal resistance, and thus has a limited hydrogen production. In order to sort out this problem, an anaerobic acidogenesis system could be used as a pretreatment of wastewaters with refractory substances, which are able to readily be acidified into volatile fatty acids, accompanied with hydrogen production [28]. Acetate is usually the predominant species in the effluent of the anaerobic acidogenic reactor. Such an acetate-laden effluent could be used as the substrate of

the MFC–MEC system reported in the present paper. In addition, the elucidation of the microbiology of the anodic electricigens and the associated mechanisms of electron transfer to the electrode in the MFC–MEC system will be beneficial to the application of this system for the wastewater treatment and hydrogen production.

4. Conclusions

In this paper, we have demonstrated the hydrogen production in the MEC-MFC-coupled biocatalyzed system could be manipulated through adjusting the loading resistors or the electricity-assisting MFCs. The loading resistors connected into the circuit in series shared the power output of the electricity-assisting MFC and reduced the power input of the hydrogen-producing MEC. As a result, the hydrogen production rate decreased with an increase in loading resistance. When the loading resistor was shifted from 10Ω to $10 k\Omega$, the volumetric hydrogen production rate varied in a range of 2.9 ± 0.2 to 0.2 ± 0.0 mL L⁻¹ d⁻¹. The hydrogen recovery, CE, and hydrogen yield decreased with an increase in loading resistor. In the coupled system with two or three electricity-assisting MFCs, the series connection significantly increased the hydrogen production. In comparison, the parallel connection failed to enhance the hydrogen production. Connecting several electricity-assisting MFCs in series had a potential to enhance the hydrogen production of MECs from organic wastes.

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