Ambient CO₂ Capture and Storage in Bioelectrochemically Mediated Wastewater Treatment

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Abstract

This study reports that wastewater can be used to capture and store CO₂ directly from ambient air and produce energy. The proof-of-concept system consisted of an ion exchange resin column that captures and concentrates ambient CO₂ using a moisture-driven cycle. The concentrated CO₂ was then transferred into a microbial electrochemical carbon capture (MECC) reactor for carbon sequestration and hydrogen production. Data from an average batch cycle showed that approximately 8 mmol/L CO₂ was captured in the MECC cathode when 0.14 g/L COD was removed in the anode. With 90% hydrogen conversion efficiency, the energy intensity and CO₂ absorption from the process could be 11.3 KJ/gCOD and 0.49 gCO₂/gCOD respectively. If the proposed process is applied, over 68 million tons of atmospheric CO₂ can be captured yearly during wastewater treatment in the U.S., which equates to significant economic values if CO₂ taxes were to be implemented more widely.

Keywords

Microbial electrolysis, ambient CO₂ capture, wastewater treatment, carbonization, microbial electrolytic carbon capture

Introduction

The role that wastewater treatment plays in the water-energy-climate change nexus is beginning to be widely recognized: every year, approximately 100 trillion gallons of various types of wastewater are moved and treated in the U.S., which consumes somewhere between 2% and 4% of the entire country's electricity and generates 0.6 gigatons of CO₂ emissions (Lu et al., 2015; McCarty et al., 2011). While reducing the energy intensity remains important for the wastewater industry to mitigate its climate change impact, novel technologies exploring the sheer volume of wastewater may open new avenues to more proactively contribute to the water-energy-climate change nexus goals (Li et al., 2015; Logan & Rabaey, 2012; Wang & Ren, 2013).

Using wastewater as a carbon sink for direct CO₂ capture is a new concept that can potentially bring to scale the climate change benefits of wastewater. Among the options, biological utilization of CO₂ was first studied and has been demonstrated for liquid fuel and chemical production. For example, photosynthetic organisms grown in wastewater photobioreactors were used to fix CO₂ into biomass for biofuel and chemical production (Shoener et al., 2014), or integrated into microbial fuel cells to fix the CO₂ produced in anode respiration (Wang et al., 2010). Similarly, microbial electrosynthesis uses an electrode to feed electrons to autotrophic electrochemically active bacteria, which reduces CO₂ into biomass and short chain organics (Rabaey & Rozendal, 2010). More recently, electrolysis approach was reported efficient in carbon capture by utilizing the OH⁻ and metal ions to convert CO₂ into inorganic precipitates, which then can be separated from solution for long term storage and utilization (Lu et al., 2015; Rau et al., 2013; Zhu & Logan, 2014).

Despite the progress, most current CO₂ capture studies were demonstrated on either pure or highly concentrated CO₂, which makes the technology limited to CO₂ generated from point

sources such as power plants or refineries. It also limits the applicability of CO₂ capture to new emissions without tackling the existing problem – the over 1,000 Giga tons of cumulative anthropogenic CO₂ emissions since 1970, which has already posed great threat to the climate (Pachauri et al., 2014). It is estimated that even if 90% of CO₂ emitted from 90% of the point sources is captured, 50% of the total anthropogenic CO₂ emission will still be unaccounted for (Metz et al., 2005). This stresses the importance of ambient CO₂ capture in meeting the grand carbon management goals. Currently, feasible ambient CO₂ capture technologies are hampered by the high energy intensity of the process, and the lack of safe carbon storage options (Oldenburg & Unger, 2003).

This work presents a wastewater treatment process that can be directly used for ambient CO₂ capture and storage. The bio-electrochemically driven process integrates microbial electrolysis, which achieves organic removal, mineral dissolution, and alkalinity accumulation, into an ambient CO₂ capture module that spontaneously absorbs and releases ambient CO₂ using a moisture-driven cycle (Wang et al., 2011b). The study also demonstrated the feasibility of a modular design and evaluated its potential for future large-scale applications from energy and carbon neutrality perspectives.

Materials and Methods

Reactor Systems

The reactor **consisted** of two modules: a microbial electrolytic carbon capture (MECC) module for wastewater treatment and mineral dissolution (calcium silicate), and an ambient CO₂ capture module (Fig. 1). The electrolysis module **consisted of** two cylindrical chambers (4 cm long × 7

cm diameter) made of polycarbonate and separated by a cation exchange membrane (CEM) (CMI-7000, Membrane International). Graphite-fiber brush (5cm in diameter and 7cm long) and carbon cloth (38cm², 30% wet proofing; Fuel Cell Earth, Woburn, MA) containing a 0.5 mg/cm² of Pt catalyst were used as the anode and cathode, respectively (Liu & Logan, 2004; Lu et al., 2015; Wang et al., 2011a). The ambient CO₂ capture and release module consists of a glass tube (Ace glass Inc., Vineland, NJ) filled with 75g strong base ion exchange resin comprising quaternary ammonium functional group (Marathon™ A OH Form, DOWEX, US). The ion exchange resin spontaneously absorbs and releases CO₂ in a moisture-driven cycle as previously described (Wang et al., 2011b). A three-way valve was used in the ambient CO₂ capture module to switch between the moisture cycle (CO₂ release) and the dry cycle (CO₂ capture).

Reactor startup and operation

The MECC anode was first acclimated in a microbial fuel cell using acetate substrate and sludge inocula (Lu et al., 2015). After stable voltage was reached for more than 30 days, the anode was transferred to the MECC module. An external 0.8 V was applied to the reactor using an external power source (denoted as DC in Fig. 1) to catalyze microbial electrolysis (Chookaew et al., 2014). A 10 Ω resistor was connected between the power source and cathode for voltage recording with a Keithley 2700 data logger (Keithley, Cleveland, OH). The MECC was then acclimated using acetate substrate in the anode (4.5 g/L NaCl, 0.8 g/L CH₃COONa, 0.31 g/L NH₄Cl and 0.13 g/L KCl, conductivity 9.33 mS/cm) and 4.5 g/L NaCl solution (conductivity 7.16 mS/cm) in the cathode, until stable microbial electrolysis performance was achieved (Haeger et al., 2014) (Luo et al., 2010). The MECC CO₂ capture experiment was then started by adding fresh anode and cathode solutions to respective chambers and adding 1mL CaSiO₃

suspension (1g silicate/50mL DI water) to the anode solution to provide carbon silicate for mineral dissolution.(Lu et al., 2015) The MECC experiment was then performed in the fed batch mode over a 24-hour period for each run. The anode and cathode pH was measured approximately every two hours. The cathodic CO₂ capture was performed approximately every six hours and 1mL CaSiO₃ suspension was added to the anode chamber every time cathodic CO₂ capture was performed to ensure the availability of CaSiO₃. An abiotic control was done using a bacteria-free anode following the same protocol as the MECC experiment.

For CO₂ capture operation, the glass column was first isolated from the MECC cathode by a 3-way valve. Air was directly pumped into the glass tube at 120ml/min until the ion exchange resin was completely dried and CO₂ saturated (approximately 4 hours). The column with saturated CO₂ was then connected to the cathodic solution through the valve and air-tight tubing, and 100mL DI water was added to the column to facilitate CO₂ release (Fig 1). The CO₂ was carried by pure nitrogen gas into the cathode chamber at 120ml/min for 30 minutes. After that, air capture process was repeated in the column while at the same time MECC was operated to convert the concentrated CO₂ into carbonate salts. Such operation allows mass balance calculation and scientific characterization though not necessarily presents an efficient way of operation.

Analytical methods

The conductivity and pH of anode and cathode solutions were measured periodically with appropriate probes (Thermo Scientific, Waltham, MA) to monitor the microbial electrolysis

activities. The anode was refilled with 1ml CaSiO_3 suspension every time the anode pH dropped below 7.0 to ensure calcium availability. The COD of the anode solution was measured at the beginning and the end of the experiments using Hach HR COD vials (Hach Company, Loveland, CO). The inorganic carbon concentrations of anode and cathode solutions were measured at the end of experiments using the standard titration method as detailed in previous studies (Lu et al., 2015). The result was reported in mmol/L per the titration method and normalized to the volume of the cathode solution.

The ion concentrations of anode and cathode solutions, including Ca^{2+} , K^+ , Na^+ , Mg^{2+} , Al^{3+} , and Cl^- , were measured at the beginning and end of experiments using inductively coupled plasma optical emission spectroscopy (ICP-OES). The carbonate precipitates on the CEM were rinsed off with 1M HCl solution and the ion concentrations were measured using ICP-OES. The results were normalized to the anodic volume (Forrestal et al., 2012).

The CO_2 capture capacity of 75g ion exchange resin was quantified using the following procedure: First, the resin was dried with air until CO_2 saturation; second, the resin column was immersed in water to release the CO_2 , which was then carried by N_2 gas into three 150ml 0.013M NaOH solutions that were sequentially connected. After 30 minutes of N_2 gas flushing, 50ml of each NaOH solution was taken for inorganic carbon quantification using the standard titration method.

Energy balance of the modular CO_2 capture reactor

The energy balance of the modular CO_2 capture reactor was calculated as:

$$Energy\ balance = (E_{H_2} - E_{electricity})/COD \quad (kJ/gCOD)$$

where E_{H_2} , $E_{electricity}$, and COD were the energy recovered in hydrogen gas, the electricity consumed by the MEC module, and the COD consumption respectively. The electricity consumed was calculated by integrating the multiplication of applied voltage and current over time based on experiment data, $E_{electricity} = \int VIdt$. The energy recovered in hydrogen was calculated as $E_{H_2} = 286 \cdot \phi$, where 286 was the heat content of hydrogen gas (kJ/mol) and ϕ was the hypothetical hydrogen gas recovery percentage based on electron balance. (Lu et al., 2015; Lu et al., 2009) In the sensitivity analysis, ϕ was varied from 50% to 90% to understand the sensitivity of energy balance to the hydrogen gas recovery percentage.

157

158 CO₂ balance of the modular CO₂ capture reactor

Both anode and cathode can capture CO₂ due to the alkalinity generated in the MECC. In the anode, alkalinity was generated via the adsorption of protons by calcium silicate, while in the cathode, alkalinity was generated from electrolysis.

The CO₂ balance of the process was calculated as:

$$CO_2\ balance = (CO_{2-capture} - CO_{2-release})/COD \quad (gCO_2/gCOD)$$

The total CO₂ captured in the process was calculated as:

$$CO_{2-capture} = CO_{2-cathode} + CO_{2-anode} + CO_{2-avoided} \quad (gCO_2/gCOD)$$

where $CO_{2-anode}$ and $CO_{2-cathode}$ were the inorganic carbon captured by the anode and cathode at the end of experiments. The avoided CO₂ emission, $CO_{2-avoided}$, was calculated by assuming

that the hydrogen gas generated in the MEC process is used to replace natural gas on the basis of heating value:

$$CO_{2-avoided} = E_{H_2} \cdot \varphi_{NG}$$

Where E_{H_2} was the heat content of hydrogen gas as detailed in the previous section, and φ_{NG} was the emission factor of natural gas based on previous reports (53.6 gCO₂/mmBTU).

The total CO₂ released in the process (gCO₂) was calculated as:

$$CO_{2-release} = CO_{2-COD} + CO_{2-electricity}$$

where CO_{2-COD} was the CO₂ released from COD oxidation, and $CO_{2-electricity}$ was the embedded CO₂ emission from electricity consumption. It was assumed that 2 mol CO₂ was produced when 1 mol of acetate was completely oxidized according to stoichiometry ($CO_{2-COD} = COD/32 \times 44$). $CO_{2-electricity}$ was calculated based on the emission factor of electricity consumption in the US:

$$CO_{2-electricity} = \varphi_{electricity} \cdot E_{electricity}$$

Where $\varphi_{electricity}$ was 558g CO₂/kWh, the average emission intensity of electricity production in the US according an EPA report;(EPA, 2014) and $E_{electricity}$ was the electricity consumption as per the previous section.

Results and Discussion

Current generation and pH change

185 The current density of the MECC reactor reached approximately 5 A/m³ after 5 hours and
186 remained stable during the experiment. Occasional spikes were observed when the reactor
187 operation was interrupted for measurements and CO₂ injections (Fig 2a). This current was much
188 higher than the abiotic control, which was less than 0.7A/m³ (Fig 2a), indicating active
189 bioelectrochemical reactions in the MECC. The current profile in this study was similar to a
190 previous study using continuous flow high strength wastewater (Lu et al., 2015), but the current
191 density was lower, mainly due to the difference in reactor conditions, such as the larger volume
192 and distance between electrodes and the use of CEM.

193 Cyclic pH pattern was observed in both anode and cathode chambers. Similar to other
194 electrolysis process, the anode pH decreased initially in each cycle due to the proton release
195 during microbial substrate oxidation, but the protons were then consumed during silicate
196 dissolution to release mineral ions, which kept a relatively stable pH overall without the addition
197 of high strength buffers (Fig 2b,c). The MECC anode pH ranged between 7.1-7.8 during stable
198 operation, while the pH in the abiotic control increased to nearly 10 due to the lack of biological
199 reaction with the presence of silicate mineral. On the other hand, cathode pH increased at the
200 beginning of each cycle due to the consumption of H⁺ and release of OH⁻, but CO₂ injection
201 subsequently reduced pH due to the capture and reduction of CO₂, which created the carbonate
202 buffer (HCO₃⁻/CO₃²⁻) (Lu et al., 2015). The MECC cathode pH stabilized between 8.2-9.4,
203 similar to a previous study using 15% CO₂ injection. In contrast, a much smaller pH change in
204 the cathode chamber was observed during the operation of the abiotic control, indicating limited
205 reactions (Fig 2b,c). The capability of keeping stable pH solves a major challenge faced by
206 abiotic or biotic electrolysis process, reduces the cost of buffer solution, and prevents the
207 production of hazardous chlorine gas.

The COD removed in the anode was 139 ± 6 mg/L or 14% during a 24-hour batch period. The batch period was determined by CO₂ cycling time **instead of** COD removal, so higher COD removal could be achieved with operation optimization based on traditional MFC practices. Hydrogen gas production was not directly monitored as in previous studies due to the mixing with flushing gas containing large amounts of N₂. Instead, H₂ recovery was calculated based on the current density using established methods described in previous work.(Logan et al., 2008; Lu et al., 2015; Luo et al., 2010) Further studies are needed to more accurately recover H₂.

Carbon capture efficiency

In the cathode chamber, 8.00 mmol/L gaseous inorganic carbon (CO₂) was captured at the end of the experiment, including 7.48 mmol/L in solution and 0.52 mmol/L as deposits on the cathode side of the CEM (Fig 3). In the anode chamber, the inorganic carbon concentration at the end of the experiment was 3.31 mmol/L, including 2.67 mmol/L in solution and 0.63 mmol/L on the anode side of the CEM. The total inorganic carbon captured in the anode accounted for 77% of the CO₂ released from acetate oxidation (assuming complete conversion of acetate to CO₂) and 29% of the CO₂ captured by the ion exchange resin respectively (Fig 3). In a follow up experiment, nearly doubled efficiency (51%) was obtained from the cathode by sequentially connecting three 150mL cathodic solutions for CO₂ absorption (Fig 3). In contrast, the CO₂ captured in the abiotic control only accounted for 4% of the CO₂ captured by the ion exchange resin (Fig 3).

The results confirm that CO₂ was indeed captured in the MECC module as a result of the microbial electrolysis. Compared to the geological storage methods with largely unknown risks, the carbonized wastewater can be a less risky storage media.(Michael et al., 2010) The

carbonized wastewater can also be reused in wastewater treatment processes (*e.g.*, nitrification) for pH adjustment, or as a source of inorganic carbon for autotrophic processes.(Jiang et al., 2015)

Two design parameters can be further optimized to improve the carbon capture and storage efficiency of the current design. First, the cathodic CO₂ sorption efficiency can be further optimized. Currently, only 29% of the CO₂ released by the ion exchange resin was captured by the MECC module. This efficiency can be improved by increasing the MECC CO₂ capture efficiency that matches up with the resin CO₂ conversion rate. This was confirmed by an efficiency increase from 29% to 51% when increasing cathode solution from 150 mL to 450 mL (Figure SI-1). Second, the ion exchange resin had a diameter of approximately 600 µm, which could be further grinded to increase the CO₂ adsorption capacity. Previous studies have shown that the CO₂ capture capacity of the ion exchange resin can increase five-fold when the resin was ground into finer particles.(Wang et al., 2011b) Such change would also reduce the size of the CO₂ capture module and offer greater flexibility in the design and operation of future scale-ups.

Liberation and migration of cations for carbon capture

At the end of the experiment, the calcium ion concentrations increased from 0.30 ppm to 111.37 ppm in the anode chamber, and from 0.005 ppm to 23.46 ppm in the cathode chamber (Table 1). This indicates that the calcium ions were liberated by the protons generated in the anodic microbial activities and migrated to the cathode for carbon capture as designed. Given the calcium deposit on the CEM (Fig 3), further improvements are needed to reduce scaling. A membrane-less design, as illustrated in Rau *et al.* 2013, could facilitate the calcium migration and reduce scaling.(Rau et al., 2013) However, the membrane-less reactor design may also

facilitate the proton migration and negatively impact the alkalinity accumulation in the cathode. Other system optimizations may help alleviate fouling and increase CaCO_3 separation, such as the upflow operation and the membrane surface coating, and investigations are underway. Sodium and potassium migration from the anode to the cathode was also observed. The sodium ion concentration increased by 81 ppm in the cathode and decreased by 173 ppm in the anode; whereas the potassium ion concentration increased by 27 ppm in the cathode and decreased by 21 ppm in the anode (Table 1). This indicates competition between the calcium ion and sodium and potassium ions for CO_2 capture. More automated amendment of calcium minerals could help ensure the availability of calcium ions and minimize undesirable ion migration.

Scale-up potential of wastewater carbon capture

The sensitivity analysis suggests that the MECC process can be an energy positive and carbon negative process. As the hydrogen gas recovery efficiency improved from 70% to 95%, the energy gain increased from 9.8 to 11.3 KJ/gCOD and the CO_2 capture efficiency increased from 0.26 to 0.49 g CO_2 -capture/gCOD (Fig 4). This indicates that improving the hydrogen recovery efficiency of MEC is important to efficient energy production and carbon capture, which is an active area of research. (Heidrich et al., 2013; Pisciotta et al., 2012)

Assuming that the average BOD of domestic and industrial wastewater were 0.35 and 0.5 g/L respectively (Table 2 and SI-1), the total biodegradable BOD in the US was estimated at over 170 million tons/year. Assuming a carbon capture efficiency of 0.40 g CO_2 -capture/gCOD is achieved, over 68 million tons of CO_2 could be captured in US's wastewater treatment process annually (Table 2). In California, the auction price of CO_2 was approximately \$12/ton CO_2e in

November, 2015 (Table SI-1). If the entire US adopted a similar carbon tax rate to help achieve its carbon mitigation goals, the CO₂ captured in wastewater treatment **would** have a tax value of over 800 million USD/year (Table 2). This suggests the significant potential economic value in using wastewater treatment plants for carbon capture and storage, not to mention the additional benefits of added alkalinity in wastewater to save on alkaline uses in nitrification and the added value of calcium carbonate that can be potentially sold as construction materials. The tax value of the US's wastewater CO₂ capture will further increase to over 1 billion USD/year, if the US **was** to adopt similar tax rates that the UK and British Columbia governments are implementing (Table 2). The costs of MECC fabrication and operation are not included in this study. Major cost reductions can be achieved by reducing electrode distances, removing expensive membranes, increasing solution conductivity, and optimizing configuration and operation. Further work is needed to better understand the costs of MECC relative to the CO₂ tax benefits.

To our best knowledge, this study is among the first to explore the economic values of MECC. It should be pointed out that most of bioelectrochemical systems are still in the liter scale. Efficiency challenges may exist with the scale-up of bioelectrochemical systems, as often encountered in chemical engineering. The scale-up of bioelectrochemical systems was discussed in more detail in a previous study (Logan, 2010). Further study on the scale-up should be attempted using chemical engineering principles such as the Buckingham π Theorem when a better understanding of the fundamental bioelectrochemical process is established (Duncan & Reimer, 1998). In the case of MECC, the theoretical estimates in this work suggest that improvements in the hydrogen recovery efficiency can improve the CO₂ capture efficiencies in the scale-up. Further, difference in wastewater characteristics, particularly the conductivity and biodegradability of COD, will likely affect the efficiency of the MECC process as they did in

bioelectrochemical systems (Logan, 2010). Further experimentation with different wastewater streams will help to better understand the scale-up potential of MECC.

Based on the simulated scale-up performance, the CO₂ efficiency is better on relatively concentrated media on a gCO₂ captured/ gCOD basis. This indicates that MECC would possibly be more suitable for integration into the sludge treatment process or concentrated wastewater from industrial sources. Further characterizations are needed to better understand the tradeoff.

Conclusions

This study successfully demonstrated that wastewater could be used for ambient CO₂ capture and storage using microbial electrolysis coupled with moisture-driven ambient CO₂ capture. An overall carbon capture efficiency of 0.49gCO₂/gCOD was achieved in the lab study. Preliminary economic analysis showed the targeted market size can be nearly 1 billion USD if the technology can be scaled up and adopted in wastewater treatment in addition to the benefits of wastewater treatment, carbon utilization, and energy production. Further studies are needed to improve system efficiency in larger scale in conjunction with comprehensive technoeconomic analysis.

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Figures and Tables

Table 1. Ion concentration changes at the beginning and end of experiment (ppm)

	Cl	Si	Mn	Fe	Mg	Ca	Al	Na	K
Cathode initial	2477	0.04	0.68	ND	0.50	0.005	0.003	1604	ND
Cathode final	2496	0.72	0.01	ND	0.089	23.46	ND	1685	27.0
Anode initial	2735	ND	0.01	ND	ND	0.30	ND	1918	70.7
Anode final	2505	74.81	ND	0.08	0.584	111.37	0.23	1745	49.2

(ND indicates below detection limit)

326 Table 2. The scale-up potential of wastewater CO₂ capture and storage

	Total Degradable BOD (tons/yr)	CO ₂ Capture (tons/yr)	Economic Values (million USD/yr)					
			California	British Columbia	Australia	France	UK	Japan
Domestic Wastewater	900,000	147,600	1.8	3.1	0.6	1.1	2.3	0.3
Industrial Wastewater	-	-	-	-	-	-	-	-
Three industries, 55 billion gallons	383,690	153,476	1.8	3.2	0.6	1.2	2.4	0.3
All industries, 0.5g/L BOD	170,100,000	68,040,000	816.5	1,428.8	272.2	520.5	1,071.6	136.1
Total	171,000,000	68,400,000	818.3	1,431.9	272.8	521.6	1,074.0	136.4

Fig 1 Schematic of the integrated ambient CO₂ capture and storage using resin and MECC.

Fig 2. (A) Current generation profile during a typical batch of experiment, (B) pH changes in the anode during the operation in the MECC reactor, and (C) pH changes in the cathode during the operation in the MECC reactor.

Fig 3. CO₂ balance in anode and cathode. The first two columns indicate the CO₂ release and capture in the anode chamber, the next two columns show the CO₂ capture in the cathode chamber, and the last two columns show the CO₂ capture in the abiotic control.

Fig 4. CO₂ and energy balance as a function of hydrogen gas recovery percentage

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