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Microbial electrolysis desalination and chemical-production cell for CO_2 sequestration

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HIGHLIGHTS

- Acid and base solutions produced from renewable organic matter.
- Desalinated water produced at the same time.
- Acid solutions used to accelerate dissolution of the mineral serpentine.
- Magnesium/calcium carbonates formed that sequestered CO_2.

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ABSTRACT

Mineral carbonation can be used for CO_2 sequestration, but the reaction rate is slow. In order to accelerate mineral carbonation, acid generated in a microbial electrolysis desalination and chemical-production cell (MEDCC) was examined to dissolve natural minerals rich in magnesium/calcium silicates (serpentine), and the alkali generated by the same process was used to absorb CO_2 and precipitate magnesium/calcium carbonates. The concentrations of Mg^{2+} and Ca^{2+} dissolved from serpentine increased 20 and 145 times by using the acid solution. Under optimal conditions, 24 mg of CO_2 was absorbed into the alkaline solution and 13 mg of CO_2 was precipitated as magnesium/calcium carbonates over a fed-batch cycle (24 h). Additionally, the MEDCC removed 94% of the COD (initially 822 mg/L) and achieved 22% desalination (initially 35 g/L NaCl). These results demonstrate the viability of this process for effective CO_2 sequestration using renewable organic matter and natural minerals.

1. Introduction

Climate change is largely driven by increasing CO_2 emissions due to the combustion of fossil fuels (Crowley, 2000; Hansen et al., 2000; Kumar et al., 2011). Many non-fossil fuel alternatives, such as nuclear, biomass, and solar energy, continue to be more expensive than fossil fuels, resulting in a need for CO_2 sequestration methods that are sustainable, renewable, and cost effective (Mikkelsen et al., 2010; Ramanan et al., 2010; Cheng et al., 2013). The weathering of silicate minerals (such as olivine, serpentine, and wollastonite) is an important natural geochemical phenomenon through which atmospheric CO_2 can be transformed to carbonates (Brady, 1991; Drever, 1994; West et al., 2005), through reactions such as

Olivine:

$$Mg_2SiO_4 + 2CO_2 \rightarrow 2MgCO_3 + SiO_2 \quad (1)$$

Serpentine:

$$Mg_3Si_2O_5(OH)_4 + 3CO_2 \rightarrow 3MgCO_3 + 2SiO_2 + 2H_2O \quad (2)$$

Wollastonite:

$$CaSiO_3 + CO_2 \rightarrow CaCO_3 + SiO_2 \quad (3)$$

The formation of carbonates through these reactions is a geologically stable and environmentally safe way to store carbon. Additionally, silicate minerals are abundant on the earth. Rocks exposed on the earth's surface by plate tectonics and erosion can be used to capture and store billions of tons of CO_2 per year, but this can only be a practical approach if the rates for these natural processes can be accelerated (Kelemen and Matter, 2008; Matter and Kelemen, 2009). Natural mineral carbonation is extremely slow due to slow rates of mineral dissolution (≈0.3 mm/year) (Lackner et al., 1995; Kelemen and Matter, 2008). Various attempts have been made to enhance the mineral dissolution including heat treatment, dry or wet attrition grinding, and the use of chemical

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additives such as $NaHCO_3$/NaCl, acidic and alkaline solutions (Park and Fan, 2004; Krevor and Lackner, 2011; Bonfils et al., 2012). Dissolution of silicate minerals can be promoted using acidic solutions, but production of the acid can be expensive.

A microbial electrolysis desalination and chemical-production cell (MEDCC) is a bioelectrochemical system that has been used to desalinate salt water and produce acid and alkali using electricity generated by microorganisms at the anode and an external applied voltage (0.3 V–1 V) (Chen et al., 2012a,b). This system consists of an anode chamber, an acid-production chamber, a desalination chamber, and a cathode chamber (the alkali-production chamber). A bipolar membrane (BPM) is used to separate the anode chamber and the acid-production chamber. An anion exchange membrane (AEM) is placed between the acid-production chamber and the desalination chamber, with a cation exchange membrane (CEM) between the desalination chamber and the cathode chamber. With an electric field, water dissociation ($H_2O \rightarrow H^+ + OH^-$) occurs in the BPM. H^+ migrates through the cation exchange layer to the acid-production chamber to produce acid and OH^- migrates to the anode chamber through the anion exchange layer, neutralizing the H^+ produced by microorganisms and maintaining near-neutral pH. Simultaneously, anions (e.g., Cl^-) and cations (e.g., Na^+) in the desalination chamber are driven through the AEM and CEM into the acid-production and cathode chambers, resulting in salt water desalination. In addition, an alkaline solution is produced in the cathode chamber through oxygen reduction ($O_2 + 2H_2O + 4e^- \rightarrow 4OH^-$).

The objective of this study was to investigate the feasibility of using a MEDCC for CO_2 sequestration through the use of the acid and alkali solutions to accelerate mineral carbonation in three steps (Fig. 1). First, acid produced by the MEDCC was used to dissolve natural minerals rich in magnesium/calcium silicates to obtain Mg^{2+} and Ca^{2+} ions. Next, part of the alkali solution produced by the MEDCC was used to raise the pH for CO_2 absorption, and then the rest was used to adjust the pH for carbonate precipitation.

2. Methods

2.1. MEDCC reactor construction and operation

The MEDCC reactors (duplicates) were made from four cylindrical chambers all 3 cm in diameter, consisting of an anode chamber (30 mL), an acid-production chamber (15 mL), a desalination chamber (15 mL), and an alkali-production chamber (30 mL) (Fig. 1). Chambers were made by drilling a hole in a solid block of Lexan. A BPM (Fumasep-FBM) was used to separate the anode and acid-production chambers, an AEM (Selemion AMV) was placed between the acid-production and the desalination chambers, and a CEM (Selemion CMV) was used to separate the desalination and alkali-production chambers. The anode was a heat treated graphite fibre brush (2.5 cm diameter × 2.5 cm length) (Feng et al., 2010) placed horizontally in the centre of the anode chamber. The cathode with a projected surface area of 7 cm^2 was made from carbon cloth with four polytetrafluoroethylene (PTFE) diffusion layers and a 0.5 mg Pt/cm^2 catalyst layer (Cheng et al., 2006). Although Pt was used here, activated carbon can be used

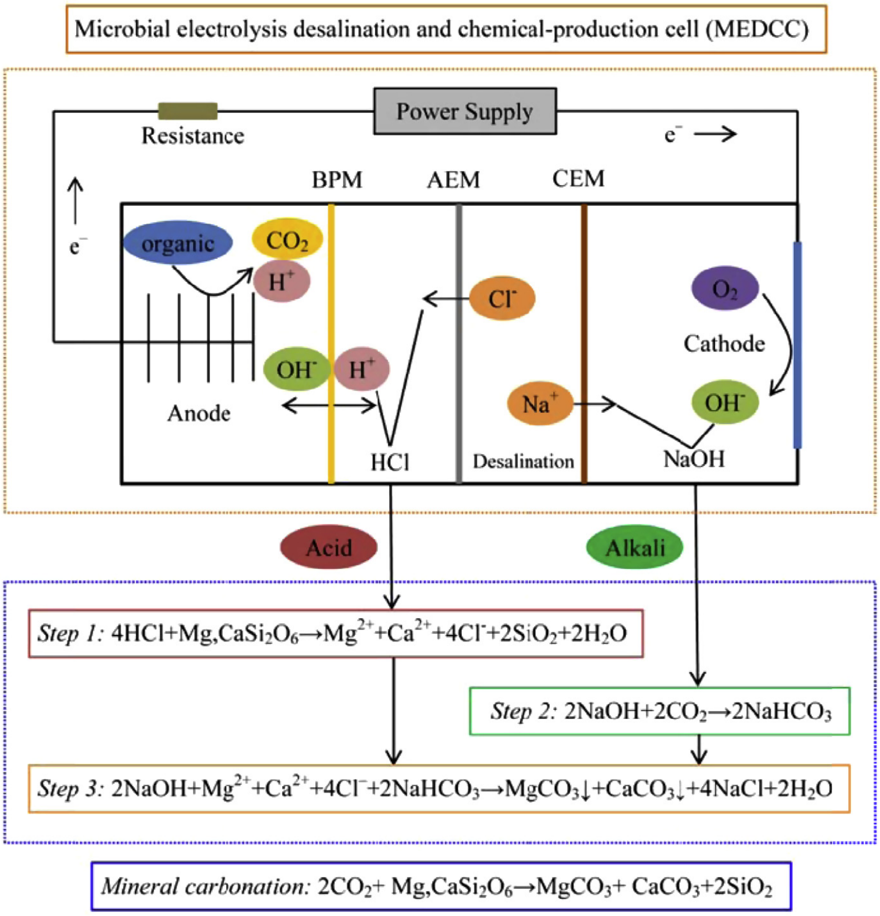

Fig. 1. Schematic representation of the MEDCC system and mineral carbonation processes.