



# A novel high performance configuration of electrochemical cell to produce alkali for sequestration of carbon dioxide



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## ABSTRACT

We propose a new configuration of a 3-compartment electrochemical cell for producing alkali that is subsequently utilized for mineralization of carbon dioxide. In this new membrane configuration, a second cation exchange membrane (CEM) replaces the anion exchange membrane (AEM) employed in a standard cell configuration. The electrochemical cell comprising of two cation exchange membranes (a CEM-CEM system) eliminates the issues associated with the AEM such as low ionic conductivity and inferior mechanical strength. All of tested CEM-CEM combinations exhibit superior electrochemical performance with considerable reduction of energy requirement for NaOH production in comparison to the standard AEM-CEM configuration. The optimized two-CEMs-based system presents a remarkable enhancement of output current density by 2.5-fold and a substantial input energy savings up to 740 kWh ton<sup>-1</sup> for mineralization of carbon dioxide to produce sodium carbonate.

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## 1. Introduction

Accelerated greenhouse gas emissions, primarily carbon dioxide (CO<sub>2</sub>), are proving extremely detrimental to global climate. Currently, the rate of anthropogenic CO<sub>2</sub> emission is far greater than what natural carbon cycle of earth can balance. As a result, the atmospheric CO<sub>2</sub> level is escalating rapidly and worsening the phenomenon of climate change. To cope with this challenge, a broad range of measures are being considered to implement, which include adoption of clean and renewable energy technologies, efficiency improvements of conventional power technologies, and capturing CO<sub>2</sub> gas from the industrial plants and other anthropogenic emission sources for preventing its exhaustion to atmosphere [1–5]. The captured CO<sub>2</sub> can either be utilized as a feedstock for producing value-added products such as fuels or other hydrocarbons, or can be stored into geological sites [6–11].

However, there are certain technical barriers associated with each of these techniques. For instance, converting CO<sub>2</sub> into useful fuels such as methanol or formic acid requires a large amount of input energy [12,13]. Such high energy requisites make the economic viability of such processes questionable. On the other hand, storage of carbon dioxide in sedimentary rocks within the earth's crust is not only dependent on the availability of proper geological sites but also raises certain safety concerns arising from the gradual gas leakage [14,15]. Another crucial factor that has to be considered for various CO<sub>2</sub> handling technologies is their feasibility for large-scale implementation. At the current stage of technology development, there is no single technique that can be universally applied for CO<sub>2</sub> sequestration. Depending on the technical and economical feasibility, one or more technologies may fit better than the others. An approach that integrates different methods may be the most practical solution for many applications.

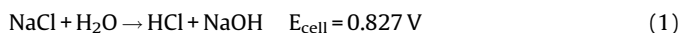
A relatively different approach that involves conversion of CO<sub>2</sub> to carbonate minerals offers advantages in terms of significant energy savings and storage safety as solid carbonate minerals alleviate the risk of gas leakage over extended period of time [7,16]. In the CO<sub>2</sub> mineralization process, carbon dioxide reacts with oxides/hydroxides/chlorides of Ca, Mg or Na to form carbonates or

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bicarbonates [17,18]. The overall energy required for carbonate formation process is considerably lower (if not zero) as compared to the other CO<sub>2</sub> conversion routes, for instance, production of hydrocarbons. The carbonate minerals are commercially valuable and can be used in a variety of household and industrial applications. In that regard, converting CO<sub>2</sub> into sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>) or sodium bicarbonate (NaHCO<sub>3</sub>) using abundantly available sodium chloride (NaCl) salt is a promising option. Large resources of NaCl make this course economically viable for scaling-up. The process involves splitting of NaCl in an electrochemical cell to produce sodium hydroxide (NaOH), which then chemically combines with CO<sub>2</sub> to form Na<sub>2</sub>CO<sub>3</sub>/NaHCO<sub>3</sub>. The overall process in a continuous-flow electrochemical cell is schematically shown in Fig. 1 [19]. The continuous-flow electrochemical cell consists of three compartments. Anode and central compartments are separated by an anion exchange membrane (AEM), while a cation exchange membrane (CEM) separates cathode from the central compartment. Brine (aqueous NaCl solution) is supplied to the central compartment. From there, chloride ions (Cl<sup>-</sup>) move to the anode through AEM and combine with protons, that are generated by hydrogen oxidation reaction (HOR), to produce hydrochloric acid (HCl). On the other side, sodium ions (Na<sup>+</sup>) move to the cathode through CEM and combine with hydroxyl ions (OH<sup>-</sup>), that are generated by water electrolysis, to produce NaOH. At the cathode, hydrogen gas (H<sub>2</sub>) is also generated as a byproduct of water splitting. Outside of the electrochemical cell, H<sub>2</sub> gas is separated from the collected cathode effluent, which can be supplied to the anode. After the gas separation, water from the cathode effluent is evaporated to obtain a concentrated NaOH product. Subsequently, the concentrated NaOH reacts with the CO<sub>2</sub> stored in a separate chamber to form either Na<sub>2</sub>CO<sub>3</sub> or NaHCO<sub>3</sub> depending on the pH value of solution.

Regarding the process energetics, HOR at the anode is a spontaneous and kinetically fast reaction that does not require any input energy. The only energy driven reaction is the water splitting at the cathode. The thermodynamic equilibrium potential for splitting water into H<sub>2</sub> gas and OH<sup>-</sup> ions is -0.827 V versus the standard hydrogen electrode (SHE) in alkaline conditions (pH = 14). The overall cell reaction along with the reversible cell potential versus SHE is given as:



The extent of reaction given in Eq. (1) mainly depends on the amount of OH<sup>-</sup> ions generated at the cathode, which means that the water electrolysis at the cathode is the limiting step of the

overall NaOH production process. Theoretically, the amount of energy needed for producing NaOH is approximately 556 kWh ton<sup>-1</sup>, however, practically it upsurges to considerably higher magnitudes due to various types of voltage losses. These performance losses originate from poor kinetics of electrochemical reactions (activation loss), large resistances to ionic transports through polymer electrolyte membranes (ohmic loss) and resistances to flow of reactants and products (mass transport loss). These losses can be minimized by optimizing individual components, structural configuration of the cell and operating conditions.

In a 3-compartment electrochemical cell that employs two different types of polymer electrolyte membranes (AEM and CEM), a considerable performance drop is caused by the poor ionic conductivity of AEM. Commercially available AEMs exhibit inferior conductivities of anions as compared to what CEMs (mainly Nafion<sup>®</sup>) exhibit for cations. Another problematic aspect of AEMs is their inability to effectively prevent proton migration through them, as will be discussed in the following section. Additionally, currently available AEMs have quite poor mechanical strength and are highly susceptible to tears and ruptures, which makes them unqualified for the development of durable electrochemical cells that can operate for extended durations without undergoing large performance drops.

In the current study, we have investigated an alternative membrane configuration of the continuous-flow electrochemical cell by replacing the AEM with a second CEM (a CEM-CEM configuration) in order to address the issues associated with the commercially available AEMs. After substantiating the feasibility of newly proposed membrane configuration, several configurations of Nafion<sup>®</sup>-based CEMs have been subsequently scrutinized and compared with the standard AEM-CEM configuration in terms of output current densities, caustic efficiencies and process energetics. It is shown that as compared to its AEM-based counterpart, an optimized CEM-CEM combination offers a significant amount of energy savings for producing NaOH and converting CO<sub>2</sub> into sodium carbonates/bicarbonates. Various figures of merit such as specific energy consumption, normalized space velocity and space-time yield have been determined for the tested membrane configurations. Subsequently, the effect of different operating conditions on the caustic efficiency and caustic concentration has been examined using the optimized membrane combination. Finally, a successful mineralization reaction of CO<sub>2</sub> using NaOH has been demonstrated forming Na<sub>2</sub>CO<sub>3</sub> or NaHCO<sub>3</sub> depending on the reaction conditions.

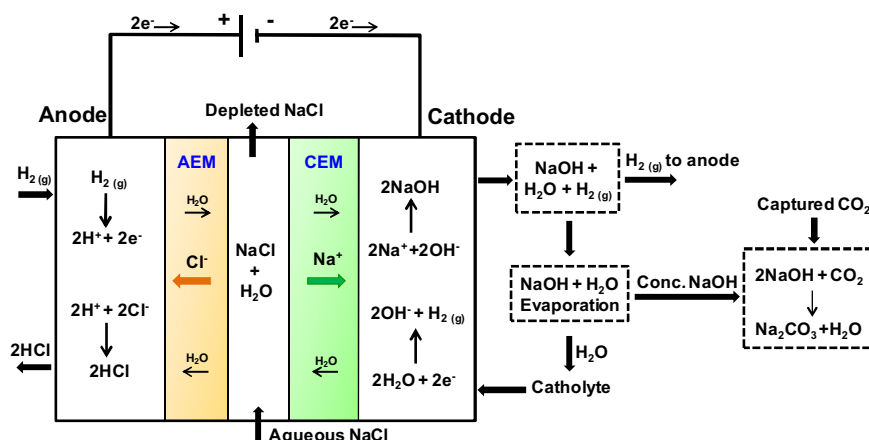


Fig. 1. Schematic diagram representing the salt splitting and CO<sub>2</sub> mineralization process in a 3-compartment continuous-flow electrochemical cell.

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