

Design and sustainability analysis of a combined CO₂ mineralization and desalination process

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Abstract: CO₂ mineralization sequesters CO₂ in a form of mineral carbonate through chemical reactions of CO₂ with metal oxide or alkaline solution. This process is attractive because it has no risk for a leakage of hazardous materials and requires a relatively small area for sequestering CO₂ compared to geological storage. In addition, generated mineral carbonate can be used as useful chemicals if its purity is high enough. One of the recent ideas in CO₂ mineralization is integrating it with desalination. Mineral ions needed for the CO₂ mineralization are separated from seawater, and generated deionized seawater is used as a feed to a desalination process, such as reverse osmosis (RO), to reduce its electric energy load. The goal of this study is to design the proposed process and examine its sustainability. The overall process is designed and simulated using Aspen plus® and Matlab. Based on the simulation results, techno-economic analysis (TEA) and CO₂ lifecycle assessment (CO₂ LCA) are conducted to verify the sustainability. In order to identify the improvement potential of the process, a best scenario study is performed. It turns out that this process can achieve about 230tonne CO₂ reduction/yr as well as relative economic benefit of almost 1million \$/yr compared to a benchmark process which comprises the stand-alone Solvay process and RO. From the best scenario result, it can be concluded that the proposed process has the potential for further investigation as means to reduce the CO₂ emission while generating economic benefits.

Keywords: Process modeling and identification; Sustainability study; CO₂ mineralization; Desalination; Techno-economic analysis; CO₂ lifecycle assessment;

1. INTRODUCTION

Greenhouse gas(GHG) emission has increased steadily since the industrial revolution and sharply in recent years, resulting in abnormal climate change (Stocker et al., 2013). Thus, carbon capture, storage and utilization (CCSU) process has attracted significant attention. Among a variety of CCSU technologies, CO₂ mineralization has been highlighted since it can be applied to a site where geological CO₂ sequestration is not viable. Also, it does not require CO₂ pre-separation which accounts for 70-75% of the cost of CCS chain. (Sanna, Uibu, Caramanna, Kuusik, & Maroto-Valer, 2014). It can sequester CO₂ using the chemical reaction of CO₂ with an alkaline solution or metal oxide. One of the examples of CO₂ mineralization is the SkyMine® project executed by Skyonic Co. In this project, the chlor-alkali process that produces sodium hydroxide and chlorine is combined with a CO₂ conversion process in which CO₂ is converted to stable a carbonate form (Jones, Barton, Clayton, Yablonsky, & Legere, 2011).

Motivated by this project, recently, our research team proposed a novel process that adopts CO₂ mineralization integrated with desalination (Fig. 1). The idea differs from

the Skymine project in two aspects. First, the new process uses seawater instead of brine as the ion source, and the ion-depleted seawater is fed to the reverse osmosis (RO) plant to reduce its electric energy load. Second, this process adopts electrodeionization (EDI) to separate ions from seawater. EDI is a hybrid technology that combines the advantages of both electrodialysis and ion exchange (Alvarado & Chen, 2014).

The main objective of this study is to design and analyze the sustainability of the proposed process in terms of both its economics and lifecycle CO₂ reduction. To this end, the overall process is simulated using commercial process simulators to come up with process data required for the analysis. Then, techno-economic analysis (TEA) and CO₂ life cycle assessment (CO₂ LCA) are carried out to assess the process performance quantitatively. It is noteworthy that the analysis is conducted as a best scenario study, since the purpose of this research is to identify improvement potential of the proposed process.

After the overall process description in section 2, detailed modelling of the key unit operations are presented in section 3. Based on the obtained mass & energy balance in section 3,

TEA and CO₂ LCA are carried out in section 4. Finally, conclusion of the research is described.

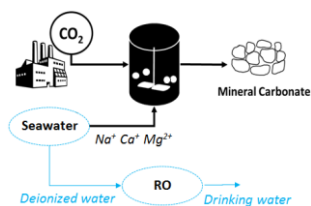


Fig. 1. Key idea of the process, adapted from (Jung, 2017)

2. PROCESS DESCRIPTION

Even though sodium chloride accounts for about 85wt% of solute in seawater, there are many other ions which can cause a serious problem in the process. For example, Mg²⁺, Ca²⁺, and SO₄²⁻ ions can lead to a scale formation in the process, which can cause the membrane in the process to break down. In addition, a small amount of K⁺ ion can lower the purity of the mineral carbonate product, which degrades the value of the product. Therefore, these unnecessary ions should be removed at the upstream of the process. In this research, an ion exchange resin is used to exchange these unnecessary ions with Na⁺ or Cl⁻ ions. For simplicity, minor ion components of which the overall portion is only ~0.75wt% are ignored in this process.

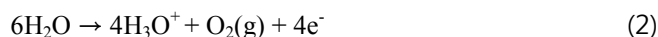
Table 1. Ion composition of seawater (Ienntech, 1998)

Ions	Concentration[mg/L]
Chloride(Cl ⁻)	18980(55.04 wt%)
Sodium(Na ⁺)	10556(30.61 wt%)
Sulfate (SO ₄ ²⁻)	2649(7.68 wt%)
Magnesium (Mg ²⁺)	1262(3.66 wt%)
Calcium(Ca ²⁺)	400(1.16 wt%)
Potassium (K ⁺)	380(1.10 wt%)
The others	256(0.75 wt%)
Total dissolved solids(TDS)	34483(100 wt%)

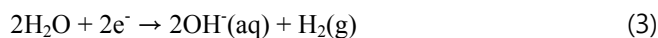
The brine (about 3.36wt% NaCl solution) from the pretreatment goes into the EDI where electrochemical reactions (equations 1~5) occur. As a nominal case, 10% deionization in the EDI is assumed. The main characteristic of EDI is that the dilute compartment is separated from the concentrated one by two membranes, anion exchange membrane (AEM) and cation exchange membrane (CEM). It allows for a selective passage of cations and anions to the anode and cathode respectively. When seawater enters the EDI, ions are migrated to each electrode under an electric field resulting in generation of H₂, Cl₂, O₂, and NaOH.

Since the ions in seawater are partially eliminated at EDI, deionized water that can be fed to RO is produced. At RO, there are two effluent streams, fresh water and concentrated brine. Concentrated brine is used to make high purity of NaCl using the solar evaporation method. At the same time, the generated NaOH is transported to the CO₂ conversion process and reacts with CO₂ in the flue gas (equations 6~7) producing carbonates. The overall process pathway is depicted in Fig.2.

Anode reaction:



Cathode reaction:



Overall reaction:



Reaction of CO₂ with NaOH:

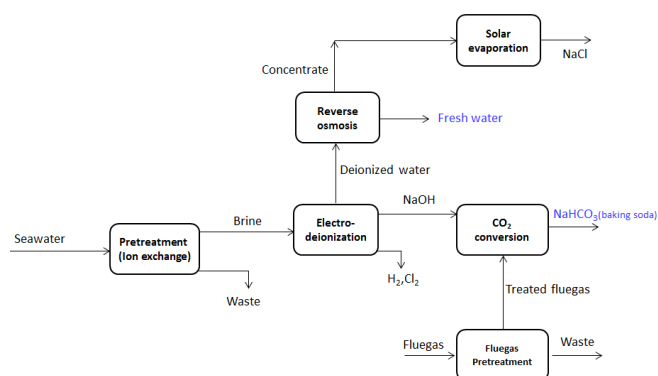
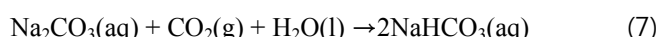


Fig. 1. Overall process pathway

3. PROCESS MODELLING AND SIMULATION

3.1 Overall description

Aspen plus[®], widely used commercial software in the chemical engineering discipline, is used as the main simulator since it provides many necessary chemical unit operations in the library. Since various ions (e.g. Cl⁻, Na⁺, OH⁻) exist in the system, eNRTL is selected as the thermodynamic property method (Chen & Song, 2004). In addition, Matlab and Excel are used to calculate and simulate the EDI unit operation, which is difficult to be simulated in Aspen plus.

3.2 Modelling of seawater pretreatment

Exchangeability between two ion species is identified based on selectivity difference of each ion to the ion exchange resin (Table 2). Since the selectivity of Na⁺ is much lower than that of Mg²⁺, Ca²⁺, K⁺, it is expected that these three cations in seawater are exchanged with Na⁺ in the cation exchange resin. Also, possibility of exchanging SO₄²⁻ with Cl⁻ is reported in (Julien Dron, 2011). Therefore, all the unnecessary ions in seawater (Mg²⁺, Ca²⁺, K⁺, and SO₄²⁻) can be exchanged with Na⁺ or Cl⁻ ions. As an optimistic approach (best scenario study), it is assumed that these four ions can be completely removed under an excess amount of cation and anion exchange resin whose functional ion is Na⁺ and Cl⁻ respectively. To regenerate the ion exchange resin, NaCl solution in high concentration is used in excess amount.

Excess amount means more than theoretically required amount.

Table 2. Relative selectivity of various cations to ion exchange resin: AG 50W-X8 resin(BIO-RAD)

Ions	Relative selectivity
Sodium(Na ⁺)	1.5
Magnesium (Mg ²⁺)	2.5
Calcium(Ca ²⁺)	3.9
Potassium (K ⁺)	2.5

3.3 Modelling of electrodeionization (EDI)

Matlab and Aspen plus are used together to simulate the EDI. In fact, these two simulators are linked using Excel as a bridge (Fontalvo, 2014). After input stream data to the *User 2* unit in Aspen plus are transported to Excel, Excel sends that information to Matlab. Then, desired output stream data and physical parameters of the *User 2* unit calculated by Matlab are sent back to Aspen plus. In Aspen plus, conversion and selectivity of each electrochemical reaction is calculated using Faraday's law and the Butler-Volmer equation (Tilak, Tari, & Hoover, 1988). Mass, energy, and voltage balances are considered together by referring to (Chandran & Chin, 1986), (Oh, 2015). Since EDI uses electricity to remove ions in seawater, required electrical energy in order to achieve 10% deionization should be calculated. For this, two partial differential equations, (8) and (9) are solved simultaneously in Matlab.

$$\frac{\partial \eta_i}{\partial w} = D \frac{F}{J} \frac{3\alpha\beta\gamma}{r^2(1-\alpha)} (C_i^b - C_i^o) \quad (8)$$

$$\frac{dC^b}{dA} = -\frac{\eta_w J}{Ff} \quad (9)$$

These two equations indicate steady-state mass balance for the resin particle (8), and for the total dilute compartment of EDI (9), respectively. The physical meaning of each parameter is presented in Table 3. Detailed derivation of the two governing equations is given in (Kurup, Ho, & Hestekin, 2009).

Table 3. Physical meanings of the parameters in equation (8), (9)

Parameter	Physical meaning
W(cm)	Width of dilute compartment
η_i	Local current utilization of i component
D(cm ² /sec)	Diffusion coefficient of ion in water
F(C/mol)	Faraday constant,
J(A/cm ²)	Current density
α	Packing ratio
β	Fraction of accessible particle surface area for ion exchange
γ	Ratio of cation to anion exchange resin
R (cm)	Radius of ion exchange resin
C ^b (mol/cm ³)	Bulk concentration of species i
C ^o _i (mol/cm ³)	Film concentration at the interface between liquid and resin solid particle of species i
A (cm ²)	Membrane cross sectional area
f (cm ³ /sec)	Feed flow rate of seawater

3.4 Modelling of CO₂ conversion process

The overall CO₂ conversion process in this system is designed using Aspen plus by referring to (Jones et al., 2011). Detailed process pathway of CO₂ conversion is described in Fig. 3. For the CO₂ absorption column, the reaction of CO₂ with NaOH is simulated by a rate based model in Aspen plus (Aspentech, 2008). Also, the following four reactions (equations 10~13) are considered and only the first two (equations 10, 11) are assumed to be in chemical equilibrium. Equilibrium constants and kinetic parameters given in (Aspentech, 2008) are adopted.

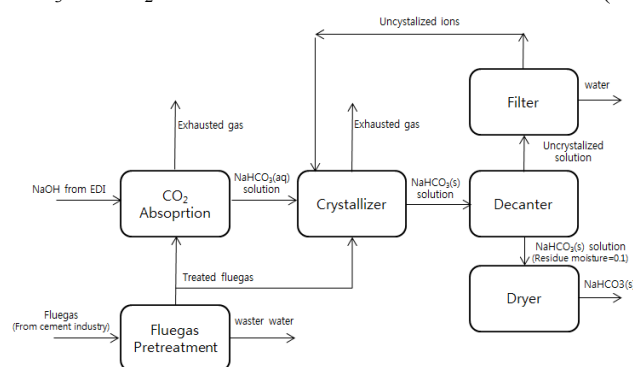
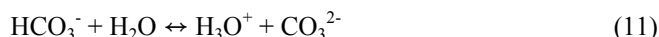


Fig. 3. Process pathway of CO₂ conversion (Jones et al., 2011)

Flue gas from a cement plant is selected as CO₂ source. Detailed composition of flue gas is presented in Table 4 (NETL, 2001). Target CO₂ capture rate is set as 90%. For simplicity, a small amount of SO₂ in the flue gas is ignored in this simulation. In the flue gas pre-treatment, water in the flue gas is removed using a flash drum. The lower the amount of water the better since it makes the reverse reaction of (11) occur more by Le Chatelier's principle, resulting in more HCO₃⁻ which is the desired ion.

Table 4. Composition of cement kiln gas(NETL, 2001)

Component	Composition (%)
CO ₂	18.9
O ₂	7.7
H ₂ O	12.8
N ₂	60.6

Aqueous sodium bicarbonate (NaHCO₃) solution goes into a crystallizer generating precipitated NaHCO₃. Additional CO₂ provided to the crystallizer enhances the conversion of CO₃²⁻ ion into HCO₃⁻ ion. To separate solid NaHCO₃ whose residue moisture is about 10% from the uncrystallized ion solution, a centrifuge type decanter is used. Uncrystallized ion solution enters the membrane filter to recover process water, and the concentrated ion solution is recycled to the crystallizer again. Finally, a dryer is used to increase the purity of NaHCO₃ product. The built-in units in the Aspen plus library used to model the aforementioned units are listed in Table5.

Table 5. Built-in unit in Aspen plus library used to model the CO₂ conversion process

Unit	Built-in unit in Aspen plus
Flue gas pretreatment	Flash2
CO ₂ absorber	RadFrac
Crystallizer	Crystallizer
Decanter	CFuge
Filter	Sep
Dryer	Heater

3.5 Modelling of reverse osmosis (RO)

Reverse osmosis is an option of desalination to obtain fresh water by applying electricity to overcome the osmotic pressure. Thus, the key information that should be calculated in modelling is the amount of electrical energy consumption in the RO. Fortunately, there is a useful spread sheet tool, Desalination Economic Evaluation Program version-5 (DEEP-5), developed by International Atomic Energy Agency(IAEA). This tool lets the user know the required amount of electrical energy to achieve reverse osmosis as a function of the amount of water to be desalinated and its concentration. This program is used to model the RO.

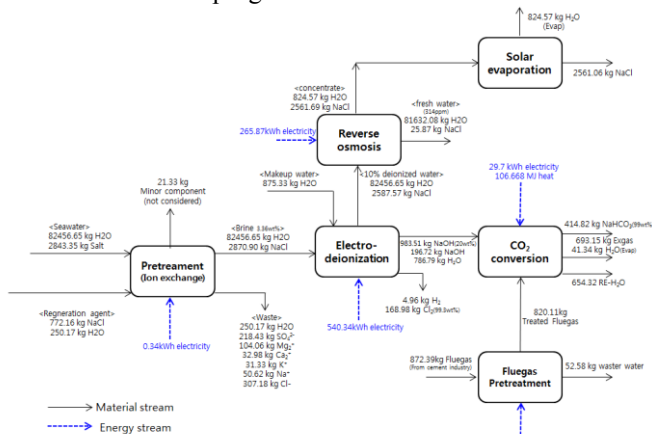


Fig. 4. Mass & energy balance information of a combined CO₂ mineralization and desalination process.

3.6 Mass & Energy balance information

Mass & energy balance information of the proposed process is derived through the previously explained modelling and simulation. Fig. 4 presents the detailed mass & energy balance information. One thing to note is that degree of freedom in deciding the process scale is just one due to the interdependency between the process unit operations. For instance, if the capacity of the desalination plant (RO) is fixed, the required amount of seawater at the upstream is determined. In addition, the generated amount of NaOH, H₂, and Cl₂ are decided automatically, since the EDI is assumed to use 10% of the ions in seawater. For the CO₂ conversion process, similar logic can also be applied. To achieve 90% CO₂ capture rate for a given amount of NaOH, the required amount of flue gas and the resulting produced amount of NaHCO₃ are fixed. In this context, scale of the process is set by designating the capacity of the desalination plant (RO) at a nominal value of 2000 m³ water/day.

4. PROCESS ANALYSIS AND EVALUATION

4.1 Selection of benchmark process

Generally, sustainability of carbon capture, storage, and utilization (CCSU) process is evaluated by comparing it with a conventional benchmark process whose main functionalities are same with the CCSU process in question. For example, let us consider the two processes in Fig. 5. These two have same main functionalities (product A and B). Under this situation, if the conventional process is replaced by the CCSU process to produce product A and B, is it better in terms of CO₂ emission and economics? This is the key question that should be answered in evaluating a CCSU process.



Fig. 5. Example: selection of a benchmark process

The main functionalities of a combined CO₂ mineralization and desalination process are CO₂ sequestration and desalination. The corresponding main products of these functions are NaHCO₃ and fresh water. Therefore, the stand-alone Solvay process and RO are selected as a benchmark process, as these two are the most commonly used process to produce NaHCO₃ and fresh water respectively. Even though the main product of the Solvay process is Na₂CO₃, NaHCO₃ can also be generated by reacting Na₂CO₃ with CO₂ and H₂O. Since the process of interest is modelled in a best-case scenario, the conversion rate of Na₂CO₃ into NaHCO₃ is assumed to be 100%. Mass & energy balance information for the benchmark process is obtained from open literature, Fig. 6 (Roland Hischer, 2007).

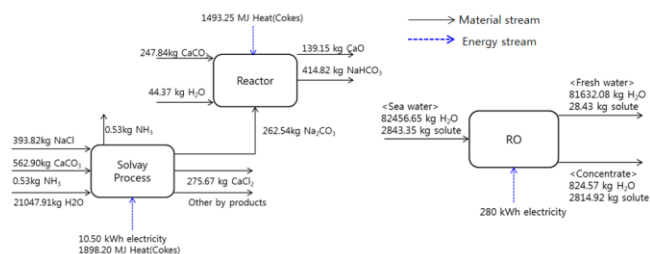


Fig. 6. Mass & energy balance information of the benchmark process (Solvay process+ RO)

4.2 Techno-economic analysis (TEA)

Table 6 shows the main assumption in TEA. In the TEA, CAPEX, OPEX (fixed operating cost + variable operating cost), and revenue are considered. Among various methods to calculate CAPEX and fixed operating cost, the method described in (Seider, 2010) is adopted. Fig. 4 and Fig. 6 are basically used as a system boundary for analysis.

Table 6. Main assumptions in the TEA

Assumption	Value
Plant life time	20yr
Discount ratio	5%
Desalination capacity	2000m ³ /day
Annual operation time	8760hr/yr

Table 7. Prices of raw materials, utility, and product

Component	Price
Raw material/Product(US \$/tonne)	
Water	0.076
NaCl	61.5
H ₂	988
Cl ₂	115
NaHCO ₃	217.5
CaCO ₃	45.85
NH ₃	339.5
CaCl ₂	110
CaO	145
Utility(US \$/kwh)	
Heat(Coal)	0.032
Electricity(Coal)	0.08

Price of raw material, utility, and product are listed in Table 7. Electricity cost presented in (Kost et al., 2013) is adopted in this research. Cost for heating is indirectly calculated by applying 40% efficiency of energy conversion from heat to electricity. For the prices of raw material & product, values in (Alibaba, 2018) and (Lee et al., 2014) are mainly used as representative values. Prices from some other website are also explored to increase the reliability of the representative values.

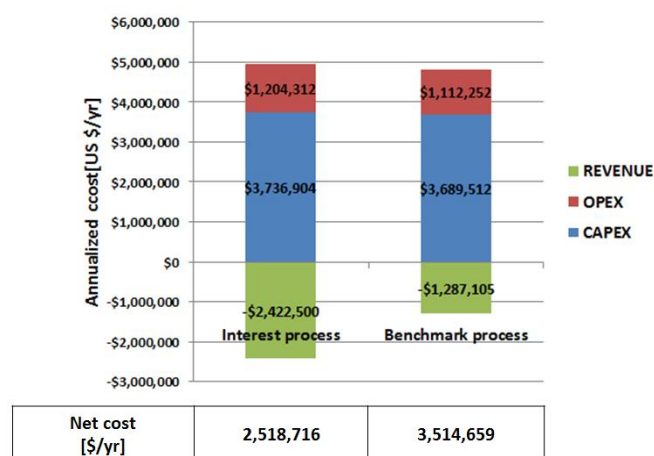


Fig. 7. Result of the techno-economic analysis

The result of the techno-economic analysis is given in Fig. 7. Comparing the OPEX and CAPEX of these two processes, they are almost the same. However, some differences exist in the revenue part. This is due to the revenue from selling NaCl. The process of interest can use the concentrated stream from the RO directly to produce high purity of NaCl, since the ion impurities are already removed at the upstream of the process using ion exchange. Contrary to this, the benchmark process cannot do this since there is no pretreatment step. The concentrated stream in the benchmark process is a mixture of various ions, not just NaCl solution. As a result, the proposed process can generate an additional economic benefit of about 1 million US \$/yr compared to the benchmark process.

4.3 CO₂ life cycle assessment (CO₂ LCA)

CO₂ life cycle assessment is a technique to quantify greenhouse gas (GHG) emission & consumption associated

with all of the stages in a process (Agency, 2010). In this research, CO₂ emission & consumption in Table 8 are considered. For CO₂ emissions due to transport, nominal distances of 100km and 50km are designated to the raw materials and product transport respectively. Note that there is indirect CO₂ emission as well as direct CO₂ emission. For instance, usage of electricity does not emit CO₂ directly. However, production of the electricity (e.g., through conversion of heat energy in fossil fuel into electricity in power plant), emits CO₂ and therefore its footprint is accounted for as indirect emission.

Table 8. Elements considered in CO₂ LCA

CO ₂ emission/consumption from
Raw material production
Raw material transport(100km)
Plant construction
Energy usage
Product transport(50km)
CO ₂ consumption in CO ₂ conversion

Table 9 shows the CO₂ emission factors for raw materials, utility, and transport. For utility, the CO₂ emission factors in (Bruckner et al., 2014) are selected, since they are most widely used values. For the others, the emission factors in (winnipeg, 2012) are used as representative values.

Table 9. CO₂ emission factors for raw materials, utility, and transport

Component	Emission factor
Raw material/Product (kg CO ₂ /kg)	
Water	0.00003
NaCl	0.2
CaCO ₃	0.01
NH ₃	2.11
NaHCO ₃	217.5
Utility (kg CO ₂ /kwh)	
Heat(Coal)	0.328
Heat(Cokes)	0.3445
Electricity(Coal)	0.82
Transport(kg/(kg*km))	
Lorry	0.00002

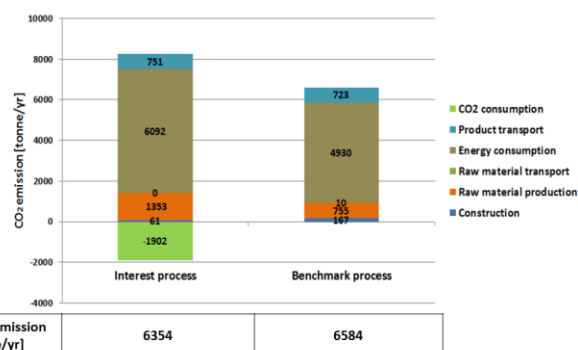


Fig. 8. Result of the CO₂ life cycle assessment

Fig. 8 presents the result of the CO₂ LCA. Even though the proposed process emits more CO₂ than the benchmark process mainly due to the energy consumption and raw

materials production, this amount of difference can be more than cancelled by the CO₂ consumption by the process. Thus, the net CO₂ emission, which is a summation of CO₂ emission and consumption, of the proposed process is lower than that of the benchmark process. It is expected that the process can achieve about 230 tonne CO₂ reduction/yr if it replaces the benchmark process.

5. CONCLUSION

A combined CO₂ mineralization and desalination process is designed using commercial simulators such as Aspen plus and Matlab. Feasibility of the process is verified by conducting TEA and CO₂ LCA. For a best scenario study, it is expected that this process can achieve CO₂ reduction as well as economic benefit compared to a benchmark process. The TEA/CO₂ LCA result of the best scenario study indicates that the proposed CO₂ mineralization process integrated with desalination has enough potential that it should be investigated further as means to reduce CO₂ emission while generating economic benefit. As a future work, optimistic assumptions used in the best scenario study such as the 100% ion removal rate in the ion exchange resin should be re-examined.

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