Life-cycle and economic assessments of carbon-based fuels production via electrolytic conversion from (bi)carbonate and CO₂

Pengtao Yue[1,2], Zhongyin Kang[1,2], Qian Fu[*1,2], Jun Li[1,2], Xun Zhu[1,2], Qiang Liao[1,2]

1 Key Laboratory of Low-grade Energy Utilization Technologies and Systems, Chongqing University, Ministry of Education, Chongqing 400044, China

2 Institute of Engineering Thermophysics, School of Energy and Power Engineering, Chongqing University, Chongqing 400044, China

ABSTRACT

The energy consumption of carbon-based fuels production can be decrease through direct electrolytic (bi)carbonate conversion due to its lack of the energy-extensive process (CO_2 release, CO_2 compression and production separation), compared to electrolytic gaseous CO_2 conversion. In this study, life-cycle and economic assessments are performed to evaluate the energy conversion characteristics, environment impacts and economic benefits of CO production via the two pathways. The results show that the net energy input, greenhouse gas emissions and net present value of electrolytic (bi)carbonate conversion are 10.4905 GJ/(t CO_2 gas), −0.6287 t $CO_{2\text{-eq}}$/(t CO_2 gas) and \$ 42,264,560, respectively, whereas in CO production through electrolytic gaseous CO_2 conversion, the corresponding values are 32.5314 GJ/(t CO_2 gas), −0.2949 t $CO_{2\text{-eq}}$/(t CO_2 gas) and \$52,917,640, respectively. Additionally, according to the sensitivity analysis, the cell voltage and Faradaic efficiency have the maximum effects on the net energy input and net present value. The greenhouse gas emissions are affected mainly by the efficiency capture. This study demonstrates the prospect and provides a theoretical direction to promote the technical and economic benefits of carbon-based fuels production via electrolytic conversion from (bi)carbonate.

Keywords: Life-cycle assessment, Economic assessment, Carbon-based fuels, Electrochemical CO_2 reduction, Electrolytic (bi)carbonate conversion

NONMENCLATURE

Abbreviations	
FE	Faradaic efficiency
NPV	Net present value
LCA	Life-cycle assessment
GHG	Greenhouse gas
NER	Net energy ratio
$CO_{2\text{-eq}}$	CO_2 equivalence

1. INTRODUCTION

Electrochemical CO_2 reduction reaction (CO₂RR) combined with renewable electricity is a potential approach for converting emitted CO_2 from industry (e.g., power plant) into carbon-based fuels (e.g., CO, HCOOH, C_2H_4) [1-2]. High rates of products formation based on a number of cell configurations (e.g., flow cell) have been demonstrated using gaseous CO_2 as the feedstock [3-4]. However, to obtain pure CO_2 gas requires additional energetic steps, such as CO_2 release, CO_2 compression, and production separation. Direct electrolytic (bi)carbonate conversion from CO_2 capture solution provides a new perspective to bypass these high energy-intensive steps.

To date, producing CO and formate through direct electrolytic (bi)carbonate conversion has been proposed in a flow cell using a bipolar membrane (BPM). A study by Li and coworkers showed that CO can be produced from electrochemical (bi)carbonate with a 37% Faradaic efficiency (FE) at 100 mA cm⁻² [5]. In another work, Li and coworkers generated pure syngas product at a 3:1 H_2:CO ratio with a current density of

150 mA cm^{-2} [6]. Benefited from electrodes design engineering, Lees and coworkers demonstrated an optimized silver gas diffusion electrode (GDE) enhance this conversion with a FE_{CO} of 82% at current densities greater than 100 mA cm^{-2}. However, to our knowledge, few studies focused on a "cradle to grave" approach to evaluate resource consumption, pollutant emissions, and economic value of the two pathways.

This situation motivated us to unify the uncertainties caused by each step in the carbon-based fuels production. Thus Life-cycle and economic assessments are carried out to compare the two pathways of electrolytic conversion from (bi)carbonate and gaseous CO_2. Net energy ratios (NER) and greenhouse gas (GHG) emissions are quantified to assess the life-cycle impacts. Net present value (NPV) is investigated to explore the economic value impacts. In addition, the sensitivity of key parameters is conducted. This work provides a new perspective to carbon-based fuels from electrolytic conversion for the industrially feasible direction.

2. Methods

2.1 Evaluation methodology

The life-cycle assessment (LCA) of CO production via the two pathways is carried out based on the ISO guidelines using a software of OpenLCA [7]. In this work, the ISO guidelines provides us a consensus framework, including goal and scope definition, inventory analysis, impact assessment, and improvement assessment.

2.2 Goal and scope definition

The goal of this work is to evaluate the technical and economic impacts of carbon-based fuels production from electrolytic conversion from (bi)carbonate and CO_2. Fig 1 shows the system boundary of CO production from electrochemical conversion with two different pathways. The inventories include CO_2 capture, CO_2 release, CO_2 compression, CO_2 electrolytic conversion and separation, but exclude the combustion of fuels. In this study, the energetic effectiveness and environment impacts caused by the use of carbon-based fuels are not considered. To compare the two process conveniently, we define the functional unit is 1 ton flue gas (CO_2) injection.

2.3 Life-cycle inventories

2.3.1 CO_2 capture, CO_2 release, and CO_2 compression

For the electrolytic CO_2 process, the CO_2 source is captured through chemical absorption using a solvent of monoethanolamine (MEA). After absorbing the flue gas (CO_2), it is accepted that desorbing CO_2 from the saline solvent through contacting with steam, which is an energetic-process. Subsequently, the gaseous CO_2
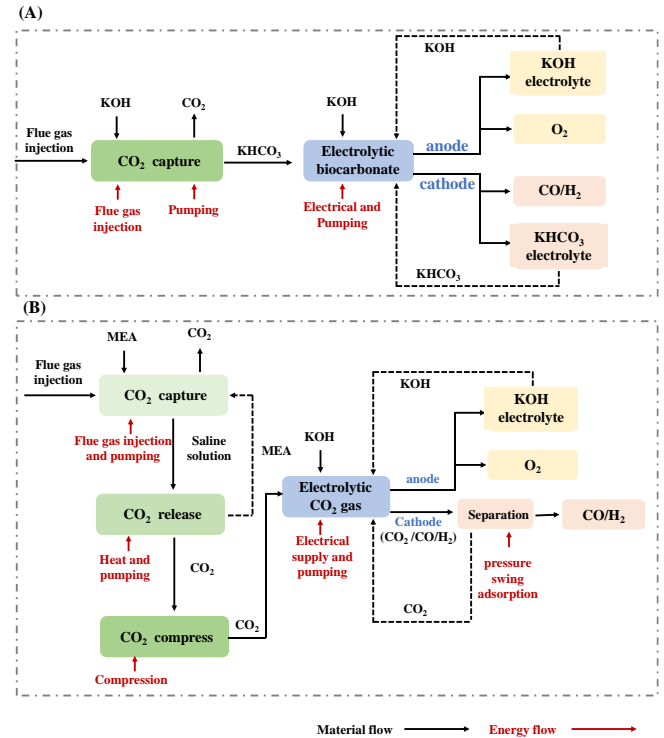

Fig 1 System boundary of CO production from electrolytic conversion from (bi)carbonate (A), and CO_2 (B).

are compressed to provide for the electrochemical process. Differently, CO_2 for the direct electrochemical (bi)carbonate can be captured using a chemical absorption solvent potassium hydroxide.

2.3.2 CO_2 electrolysis

To date, most fundamental studies for electrolytic CO_2 process have made much progress with a high FE (>90%) and current density (>200 mA cm^{-2}) based on the flow cell. There are just few researches focused on the electrolytic (bi)carbonate conversion with a lower FE (<80%) and current density (<200 mA cm^{-2}).

2.3.3 CO_2 separation

The $CO_2/CO/H_2$ gas mixture is separated through a technology of pressure swing adsorption (PSA). A fixed specific energy consumption of 201.6 KJ/mol is assumed on the basis of recent reports [6].

3. Results and discussion.

3.1 Life-cycle assessment

Fig 2(A) shows the net energy input of the CO production via electrolytic conversion from (bi)carbonate and CO_2. It is assumed that energy input is positive (e.g., pumping, electricity, heating, compression and separation) and the energy output is negative (e.g., CO, H_2 and O_2). Suffering from the high overpotential of electrolysis, electrolytic (bi)carbonate
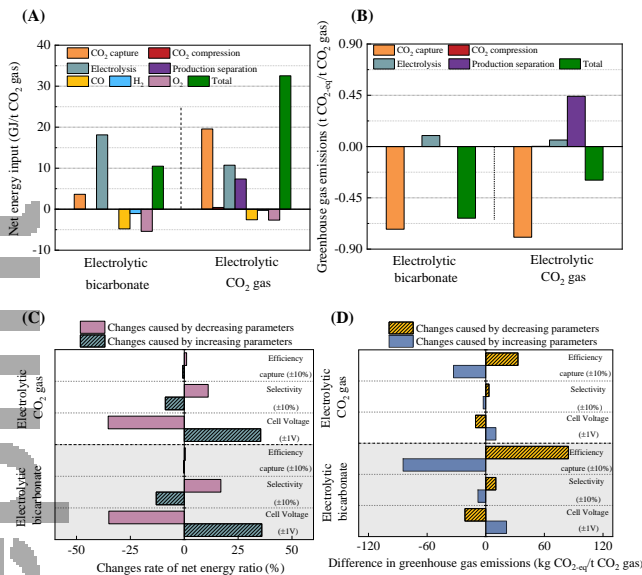
Fig 2 The net energy input and GHG emissions of CO production via electrolytic conversion from (bi)carbonate (A), and CO_2 (B).The changes rate of NER and difference in GHG of CO production via electrolytic conversion from (bi)carbonate (C), and CO_2 (D).

conversion shows a net energy input of 18.1285 GJ/(t CO_2 gas), which is larger than the other approach (10.7233GJ/(t CO_2 gas)). Nonetheless, due to the energy-extensive consumption of CO_2 capture (19.563 GJ) and separation (7.3617 GJ), it is shown that the whole net energy input of electrolytic CO_2 gas (32.5314 GJ/(t CO_2 gas)) is greater than the opposite (10.4905 GJ/(t CO_2 gas)). Additionally, it is worth mentioning that the process of electrolytic CO_2 gas reveals a low energy output owing to the loss of CO_2 during separation.

As shown in Fig 2(B), it is indicated that the total GHG emissions of the electrolytic (bi)carbonate and CO_2 gas are −0.6287 and −0.2949 t $CO_{2\text{-eq}}$/(t CO_2 gas), respectively. This difference in GHGs is mainly attributed to the energy requirements for electrolytic CO_2 gas to support the production separation. The GHG emissions in separation is 0.4410 t $CO_{2\text{-eq}}$/(t CO_2 gas). Noticeably, GHG emissions in CO_2 compression can be negligible, owing to its low energy requirements.

3.2 Sensitivity analysis

The effects of efficiency capture, FE and cell voltage on the NER and GHG emissions of CO production via electrolytic conversion from (bi)carbonate and CO_2 are investigated. As shown in Fig 2(C), NER is significantly affected by cell voltage and FE. Limited by the high overpotential of electrochemical process, it has been a bottleneck to break through this issue in condition of high current density (> 100 mA cm^{-2}). Therefore, it is imperative to decrease the overpotential and enhance the FE of CO for further NER decreased.

Fig 2(D) shows the difference in GHG emissions affected by the variation in parameters. The efficiency energy has the most important influence on GHGs. It is visible that the GHG emissions in direct electrolytic (bi)carbonate can be decreased deeply after enhancing the efficiency capture because of the primary stage of the chemical absorption technology using potassium hydroxide. Inversely, it is indicated that the GHG emissions are affected slightly by the cell voltage and FE.

In summary, through sensitivity analysis, it is shown that NER can be close to 1 via decreasing overpotential and improving FE of CO. When integrated with renewable electricity such as solar energy, CO production via electrolytic conversion from (bi)carbonate is still prospective in the future. Moreover, GHG emissions can be minimized via direct electrolytic (bi)carbonate conversion accompanied by carbon-based fuels production. To our knowledge, CO and HCOOH are the two types products with a high FE (> 60%) and current density (> 100 mA cm^{-2}) via direct electrolytic (bi)carbonate conversion. With the development of catalysts and interface engineering, other C_{2+} products (e.g., C_2H_4, C_2H_5OH, C_3H_8) can be obtained, which will make this technology more competitive.

3.3 Economic assessment and sensitivity analysis

Fig 3(A) and 3(B) presents the construction cost and NPV of electrolytic conversion from (bi)carbonate and CO_2 gas. Obviously, the cost of electrolytic CO_2 conversion ($ 6,271,569) was less than electrolytic (bi)carbonate conversion ($ 13,768,862). After 20 years operation of the industry, the NPV of electrolytic (bi)carbonate and CO_2 gas conversion are $ 42,264,560 and $ 52,917,640, respectively. This difference in the cost and NPV is mainly ascribed to the low current density (<100 mA cm-^{2}) via direct electrolytic

(bi)carbonate conversion compared to the other approach (>200 mA cm^{-2}).

The influences of FE, cell voltage, cost of CO, electrovalence, cost of CO_2, and current density NPV of CO production via electrolytic conversion from (bi)carbonate and CO_2 are carried out. NPVs of both pathways are mainly affected by the cell voltage. Therefore, further reducing cell voltage is extremely significant to promote the economic benefit in both pathways. For the process of electrolytic (bi)carbonate conversion, it is found that FE and current density have a great impact on NPV. As the development of catalysts and systems, electrolytic conversion from (bi)carbonate with high current density (>100 mA cm^{-2}) and FE (>90%) will be competitive in the future. In addition, the NPV is also influenced by the price of CO_2, CO and electrovalence. Thus, to develop a low-cost CO_2 capture technology is important for improving the economic benefits of the two approaches.
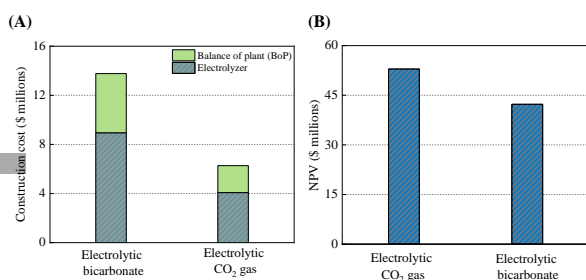

Fig 3 The construction cost and NPV of CO production via electrolytic conversion from (bi)carbonate (A), and CO_2 (B).

4. Conclusion

In this study, life-cycle and economic assessments of CO production via electrolytic conversion from (bi)carbonate and CO_2 are carried out. CO production through the two pathways show great environmental and economic performance, especially combined with renewable energy as the electricity. CO production from direct electrolytic (bi)carbonate conversion exhibits lower net energy input (10.4905 GJ/(t CO_2 gas)) and GHG emissions (-0.6287 t CO_{2-e}q/(t CO_2 gas)). Reversely, the net energy input and GHG emissions of CO production via electrolytic CO_2 conversion are 18.1285 GJ/(t CO_2 gas) and -0.2949 t CO_{2-e}q/(t CO_2 gas), respectively. However, due to the low current density, the process of direct electrolytic (bi)carbonate conversion represents a low NPV ($ 42,264,560) compared to the other pathway ($ 52,917,640). Furthermore, it is found that the improvement in efficiency capture can reduce GHG emissions. Reducing

the cell voltage can further decrease net energy input and promote the economic benefit through sensitivity analysis. This study provides a theoretical direction to promote the technical and economic benefit of carbon-based fuels production via electrolytic conversion from (bi)carbonate and CO_2.

REFERENCE

[1] Phil De Luna, Christopher Hahn, Drew Higgins, Shaffiq A. Jaffer, Thomas F. Jaramillo, Edward H. Sargent. What would it take for renewably powered electrosynthesis to displace petrochemical processes? Science 2019;350:364–373.

[2] Pengtao Yue, Qian Fu, Jun Li, Liang Zhang, Lei Xing, Zhongyin Kang, Qiang Liao, Xun Zhu. Chemical Engineering Journal 2021;405:126975-126984.

[3] Shaoxuan Ren, Dorian Joulié, Danielle Salvatore, Kristian Torbensen,Min Wang, Marc Robert, Curtis P. Berlinguette. Molecular Electrocatalysts can Mediate Fast, Selective CO_2 Reduction in a Flow Cell. Science 2019;365:367-369.

[4] Md Golam Kibria, Jonathan P. Edwards, Christine M. Gabardo, Cao-Thang Dinh, Ali Seiftokaldani, David Sinton, Edward H. Sargent. Electrochemical CO_2 Reduction into Chemical Feedstocks: From Mechanistic Electrocatalysis Models to System Design. Advanced Materials 2019;37:1807166-1807190.

[5] Tengfei Li, Eric W. Lees, MaxwellGoldman, Danielle A. Salvatore, David M. Weekes, Curtis P. Berlinguette. Electrolytic Conversion of Bicarbonate into CO in a Flow Cell. Joule 2019;3:1487-1497.

[6] Yuguang C. Li, Geonhui Lee, Tiange Yuan, Ying Wang, Dae-Hyun Nam, Ziyun Wang, F. Pelayo García de Arquer, Yanwei Lum, Cao-Thang Dinh, Oleksandr Voznyy, Edward H. Sargent. CO_2 Electroreduction from Carbonate Electrolyte. ACS Energy Letters 2019;4:1427-1431.

[7] Rebitzer G, Ekvall T, Frischknecht R, Hunkeler D, Norris G, Rydberg T, Schmidt W. P, Suh S, Weidema B.P, Pennington D.W. Life cycle assessment: Part 1: Framework, goal and scope definition, inventory analysis, and applications. Environment International 2004;30:701-720.