No. A-4-1E

PJ : Development of Combined Carbon Capture and Conversion System for Utilization of Atmospheric CO_2

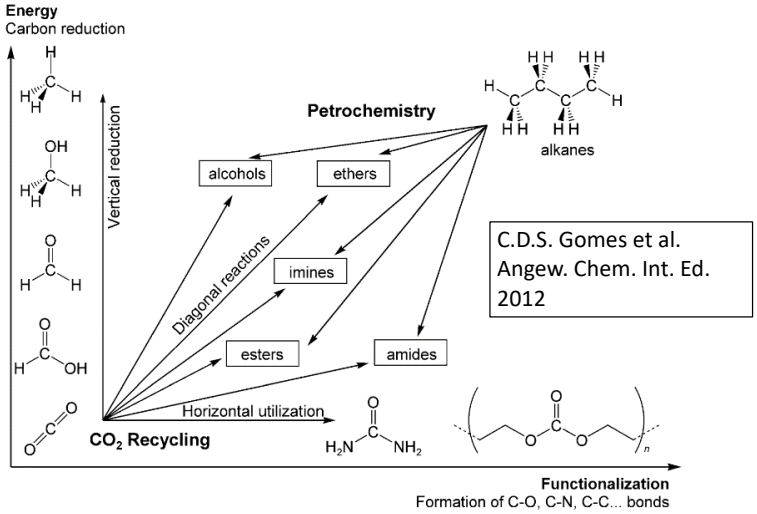
Project Overview

Organization: Tohoku University, Osaka Metropolitan University, Renaissance Energy Research

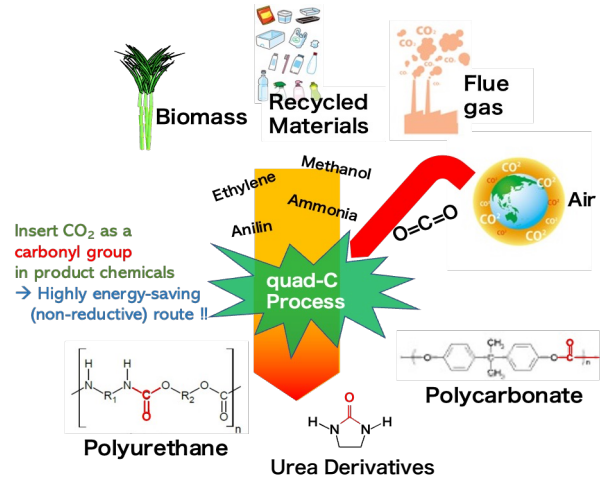
Contact: Prof. Yasuhiro FUKUSHIMA fuku@tohoku.ac.jp (PM)

Background

CO_2 utilization can be more energy saving (horizontal utilization)
… , but needs to target variety of downstream chemicals with smaller market size



C.D.S. Gomes et al.
Angew. Chem. Int. Ed.
2012



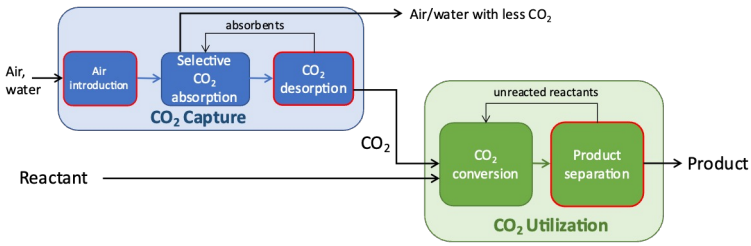
Our strategy for atmospheric CO_2 utilization:

Energy Saving Strategy

Common challenge for DAC-U:
Energy requirements in:
1) Air introduction
2) CO_2 desorption.
3) Product separation

→ Use of **Dual Function Materials (DFMs)** for capturing CO_2 from the atmosphere

→ Eliminate need of 2) CO_2 desorption



Catalyst

$A + CO_2 \rightarrow P + B$

Reactant — Product — Byproduct

Please visit Poster A-4-4E for more details

quad-C Type I process:
DFM: Reactant × Absorbent of CO_2
with aid of CO_2 selective membrane
to avoid loss of reactant by evaporation

Please visit Poster A-4-2E for more details

quad-C Type II process:
DFM: Catalyst × Adsorbent of CO_2
Our project not only explored CeO2 as DFMs in detail, but also identified other potential DFMs.

Please visit Poster A-4-3E for more details

Summary of Outputs:

4 students received (incl. expected) Ph.D. degree

Dr. Jie Peng (Catalyst Development, Tomishige Lab., 2023)
Dr. Xing-Yi Yang (DFM development, Yoshioka Lab. – Kameda Gr., 2023)
Mr. Koki Yagihara (Process simulation, Fukushima Lab., 2024)
Mr. Ryotaro Fujii (Catalyst Development, Tomishige Lab., 2024)

Patents

Applied for 2 patents (1 PCT, 1 Japanese Domestic)
More in preparation

Publications: *More are in preparation*

1. CeO2-Catalyzed Synthesis of 2-Imidazolidinone from Ethylenediamine Carbamate, ACS Omega, 2021
2. Analyzing flue gas properties emitted from power and industrial sectors toward heat-integrated carbon capture, Energy, 2022
3. CeO2-catalyzed transformation of various amine carbamates into organic urea derivatives in corresponding amine solvent, Applied Catalysis A: General, 2022
4. Continuous Flow Synthesis of 2-Imidazolidinone from Ethylenediamine Carbamate in Ethylenediamine Solvent over the CeO2 Catalyst: Insights into Catalysis and Deactivation, ACS Catalysis, 2023
5. Effective synthesis of ethylene urea from CO2 adsorbed cerium doped Mg–Al layered double hydroxide, Journal of Cleaner Production, 2023
6. Enrichment of carbon dioxide using Mg–Al layered double hydroxides, Chemical Engineering Research, 2023
7. Adsorption behavior of atmospheric CO2 with/without water vapor on CeO2 surface, Applied Catalysis B: Environmental, 2024
8. Assessing economic trade-off for advances in amine-based post-combustion capture technology, Journal of CO2 Utilization, 2024

Spin-out from the Moonshot Program

Collaboration with TRE Holdings: Appl. to flue gas from waste incineration


2023年 | プレスリリース・研究成果

「TREホールディングス×東北大学WX（Waste Transformation）共創研究所」を開所 -廃棄物処理の革新的プロセスの開発とCCU技術の社会実装-

2023年12月 1日 11:00 | プレスリリース・研究成果

【発表のポイント】

- 国立大学法人東北大学と、TREホールディングス株式会社は、「TREホールディングス×東北大学WX（Waste Transformation）共創研究所」を本日開設しました。
- 廃棄物の焼却処理とCO2回収処理を統合する革新プロセスの開発に取り組み、CCU（Carbon Capture Utilization）（注1）技術の社会実装を目指します。

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PJ : Development of Combined Carbon Capture and Conversion System for Utilization of Atmospheric CO_2

Type II quad-C: Conceptual Design and Energy Analyses

Organization: Tohoku University, Osaka Metropolitan University, Renaissance Energy Research

Contact: Prof. Yasuhiro FUKUSHIMA fuku@tohoku.ac.jp (PM)

Core Members

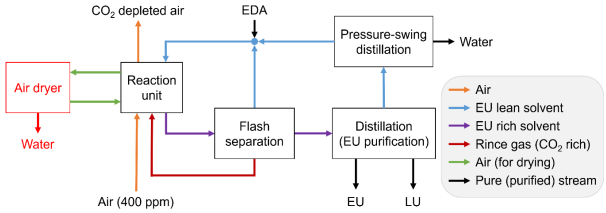
Fukushima Y. (Professor)
Ni J.-L. (Assist. Professor)
Yagihara K. (Ph.D. student)
Prospective assessment assisted by simulation technologies

Tomishige K. (Professor)
Yabushita M. (Assist. Prof.)
Peng J. (Ph.D. student, graduated)
Fujii R. (Ph.D. student, graduated)
Development of catalytic reaction systems

Kitakawa-Shibasaki N. (Professor)
Takahashi A. (Assoc. Professor)
Hiromori K.(Assist. Professor)
Nakamura M. (Researcher)
Endo T. (Researcher)
Development of reactors and reaction systems

Conceptual Design of quad-C process (Type II)

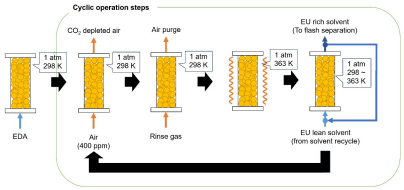
Schematic diagram



Assumptions from experimental study

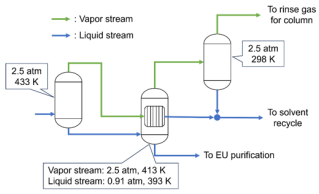
- Ratio of EDA-CA/EDA (for EU synthesis): 9.2 mol%
- Yield - CO_2 capture ・・・ CO_2 conversion: 100%
  - EU synthesis ・・・ EU: 51%, LU: 2.5% (EDA-CA basis)

Reaction unit



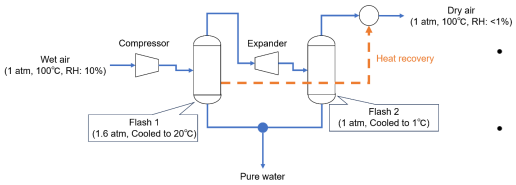
- Air-loading time is identical to reaction time (4.5 h)
- Recycled EU ratio: 0.1～0.9
  - Ratio of the EU-rich solvent remaining in reaction unit

Flash separation



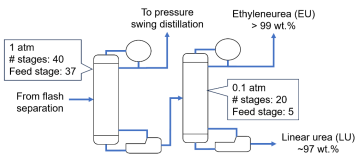
- Flash separation has three outlet streams
  - Rinse gas (CO_2 rich)
  - Solvent recycle (EU lean)
  - EU purification (EU rich)

Air dryer

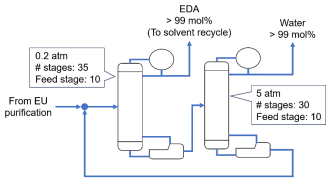


- Water is the only substance discharged from reaction unit
- Air-drying ratio: 0.1～0.9

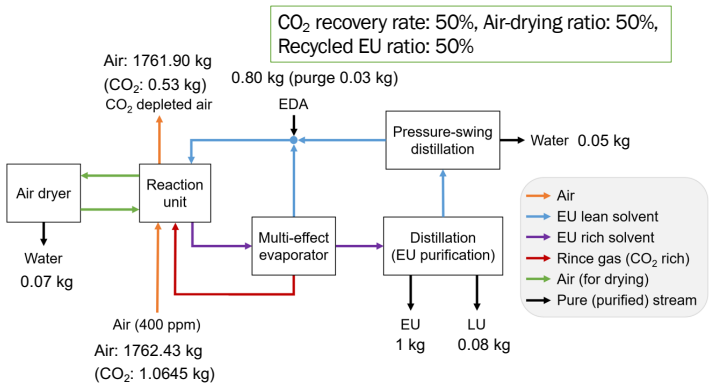
Distillations for EU purification

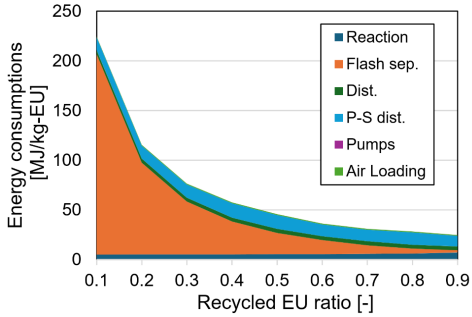


Pressure-swing distillation

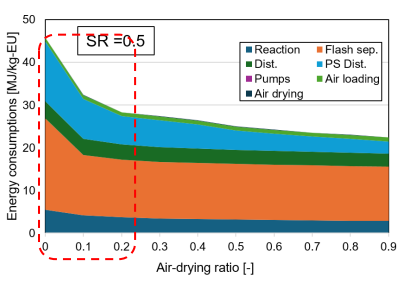


Simulated material balance and energy analyses

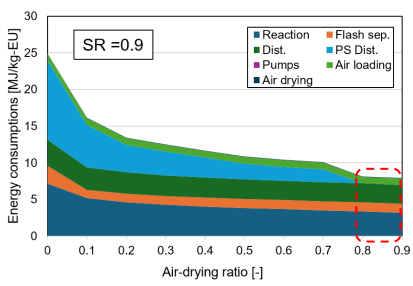




EU enrichment by inner EU recycle achieves significant energy saving. (without air dryer)
Excess retention may cause EU→LU, need experiments



for low inner recycle ratio (SR)
10~20% air drying
achieves effective energy saving



for high inner recycle ratio (SR)
>80% air drying
achieves uncontinuous energy saving due to simplification of pressure swing distillation

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Dual Function Materials and their Adsorption Mechanisms

Organization: Tohoku University, Osaka Metropolitan University, Renaissance Energy Research

Contact: Prof. Yasuhiro FUKUSHIMA fuku@tohoku.ac.jp (PM)

Core Members

Tamura M. (Assoc. Professor)
Akatsuka M. (Assist. Professor)

Understanding catalytic and adsorption mechanisms using advanced spectroscopic methods

Tomishige K. (Professor)
Yabushita M. (Assist. Professor)
Peng J. (Ph.D. student, graduated)
Fujii R. (Ph.D. student)

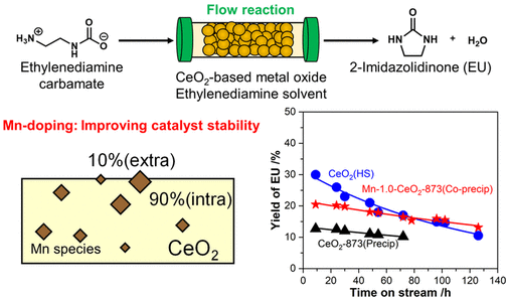
Development of catalytic reaction systems

Kameda T. (Professor)
Motswaiso F. (Researcher)
Yang X. (Ph.D. Student, graduated)
Rahman F. (Researcher)
Primeia S. (Researcher)

Development of Dual Function Materials other than CeO_2 (ex. Layered Double Hydroxides, $Ti_xZr_yO_2$, etc.)

DFM Development

Maintaining catalyst activity



R. Fujii et al. ACS Catalysis, 2023

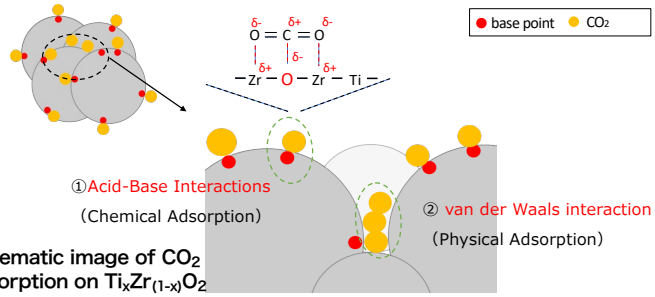
Poly-urea-like substance is observed when CeO_2 catalyst is used continuously. Mn-doping slows down the deactivation.

Improving tolerance to trace acid

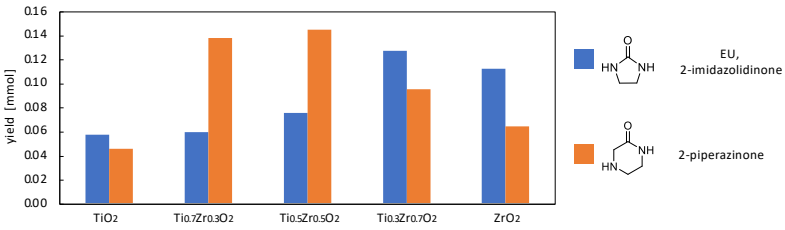
Acid substances such as NOx and SOx
→ Treated flue gas still contains trace amount that can be sufficient in affecting catalysts
→ Atmospheric gas may also contain trace amounts (CO_2 is also trace, so it can affect the catalysts)

We have acquired data on the acceptable level of NOx and SOx for flow synthesis of EU.

Enhancement of selectivity and adsorbed amount



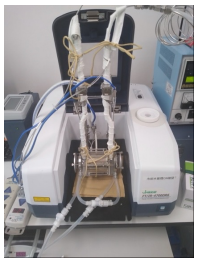
Schematic image of CO_2 adsorption on $Ti_xZr_{(1-x)}O_2$



- ✓ $Ti_xZr_{(1-x)}O_2$ is a good **promoter** and **CO_2 supplier** to the reaction
- ✓ Selectivity of EU increases as Zr content increases

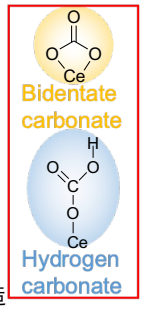
Adsorption mechanisms

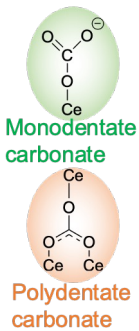
Akatsuka et al. Applied Catalysis B: Environmental, 2024

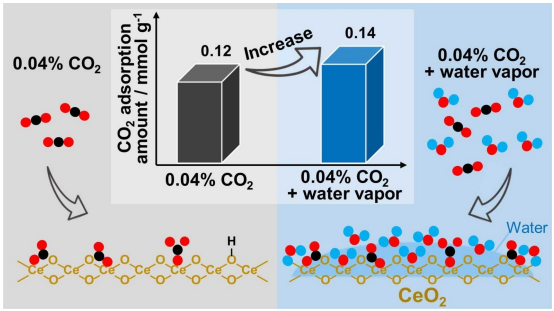


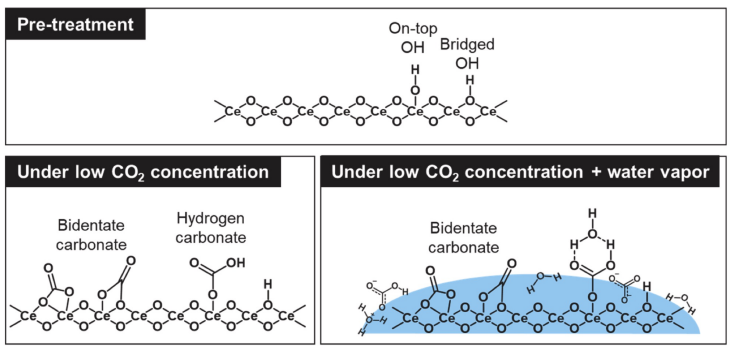
実験装置:
FT-IR, 本事業で改造

Major forms









- 0.04% (400 ppm), lean CO_2 can be adsorbed onto the CeO_2 surface
- Adsorbed species were bidentate and hydrogen carbonates
- Effect of moisture in the feed gas were clarified in terms of species and amount

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Type I process: Membrane-enabled use of amines as DFMs

Organization: Tohoku University, Osaka Metropolitan University, Renaissance Energy Research

Contact: Prof. Yasuhiro FUKUSHIMA fuku@tohoku.ac.jp (PM)

Members

Membrane Module Development

Dr. Osamu Okada
President,
Renaissance Energy Research corporation

Dr. Masaru Watanabe
Professor,
Tohoku University

Dr. Toshiyuki Nonaka
Project Associate Professor,
Tohoku University

Dr. Yuya Hiraga
Assistant Professor,
Tohoku University

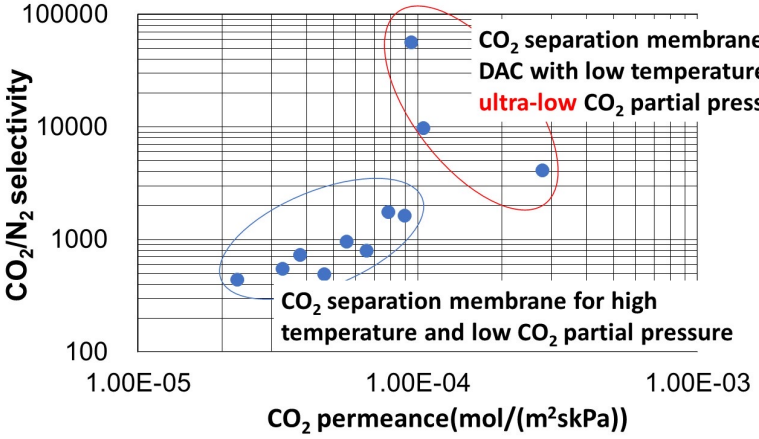
Process Simulation

Dr. Yasuhiro Fukushima
Professor,
Tohoku University

Dr. Jialing Ni
Assistant Professor,
Tohoku University

Type I quad-C process

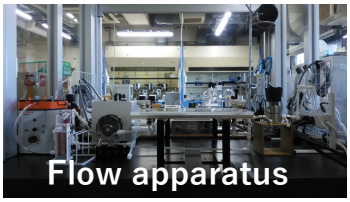
Successfully prototyped facilitated transport membranes with excellent CO_2/N_2 selectivity and sufficient permeance



✓Prototyped hollow fiber membrane module with many support materials with various d, and pore size

D = 1.2mm:
CO_2 permeance : 2.7×10^{-4} mol/m²skPa
CO_2/ N_2 Selectivity: 4,000

D = 3mm
CO_2 permeability : 9.5×10^{-5} mol/m²skPa,
CO_2/ N_2 Selectivity: 55,000



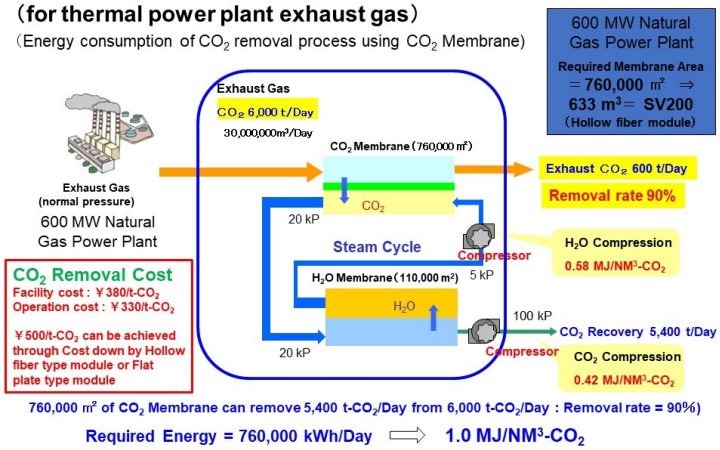
Development of membrane modules with ionic liquids

✓Exploring the optimal ionic liquid using novel in-situ Raman spectroscopy

✓DAC experiments [semi-batch system and flow apparatus] (time-average fluxes: 1～3 x 10^{-4} mol-CO_2/m²s at 120L/min)

Spin-out strategy

Study of CO_2 removal process
(for thermal power plant exhaust gas)
(Energy consumption of CO_2 removal process using CO_2 Membrane)



Existing CO_2 separation and recovery technologies, such as absorption and adsorption methods, are energy-intensive. In the membrane separation method using CO_2 selective permeation membranes, CO_2 is absorbed and released through the membrane, so that the energy generated during CO_2 absorption (absorption heat) is used as energy for CO_2 release, making it an energy-saving process. This technology will be applied to various CO_2 sources, and a model for a carbon-circulating society will be built by producing and utilising methane from the recovered CO_2 at high efficiency.